VILNIUS GEDIMINAS TECHNICAL UNIVERSITY

Tim TOFAN

INVESTIGATION OF THE INKJET PRINTING PROCESS, EVALUATING THE DYNAMICS OF THE INK AND THE PROPERTIES OF SPECIFIC PRINTING SURFACES

DOCTORAL DISSERTATION

TECHNOLOGICAL SCIENCES MECHANICAL ENGINEERING (T 009)

Vilnius, 2023

The doctoral dissertation was prepared at Vilnius Gediminas Technical University in 2019–2023.

Supervisor

Assoc. Prof. Dr Raimondas JASEVIČIUS (Vilnius Gediminas Technical University, Mechanical Engineering – T 009).

The Dissertation Defence Council of Scientific Field of Mechanical Engineering of Vilnius Gediminas Technical University:

Chairman

Prof. Dr Dalius MAŽEIKA (Vilnius Gediminas Technical University, Mechanical Engineering – T 009).

Members:

Prof. Dr Sergiy ANTONYUK (University of Kaiserslautern-Landau, Germany, Mechanical Engineering – T 009),

Prof. Dr Habil. Rimantas BARAUSKAS (Kaunas University of Technology, Mechanical Engineering – T 009),

Prof. Dr Julius GRIŠKEVIČIUS (Vilnius Gediminas Technical University, Mechanical Engineering – T 009),

Prof. Dr Arnas KAČENIAUSKAS (Vilnius Gediminas Technical University, Mechanical Engineering – T 009).

The dissertation will be defended at the public meeting of the Dissertation Defence Council of the Scientific Field of Mechanical Engineering in the SRA-I Meeting Hall of Vilnius Gediminas Technical University at **10 a.m. on 20 December 2023**.

Address: Saulėtekio al. 11, LT-10223 Vilnius, Lithuania. Tel.: +370 5 274 4956; fax +370 5 270 0112; e-mail: doktor@vilniustech.lt

A notification on the intended defence of the dissertation was sent on 17 November 2023. A copy of the doctoral dissertation is available for review at the Vilnius Gediminas Technical University repository http://dspace.vgtu.lt and the Library of Vilnius Gediminas Technical University (Saulėtekio al. 14, LT-10223 Vilnius, Lithuania).

Vilnius Gediminas Technical University book No 2023-047-M

doi:10.20334/2023-047-M © Vilnius Gediminas Technical L

© Vilnius Gediminas Technical University, 2023 © Tim Tofan, 2023 *tim.tofan@vilniustech.lt* VILNIAUS GEDIMINO TECHNIKOS UNIVERSITETAS

Tim TOFAN

RAŠALINIO SPAUSDINIMO PROCESO TYRIMAS, ĮVERTINANT RAŠALO DINAMIKĄ IR SPECIFINIŲ SPAUSDINIMO PAVIRŠIŲ SAVYBES

DAKTARO DISERTACIJA

TECHNOLOGIJOS MOKSLAI, MECHANIKOS INŽINERIJA (T 009)

Vilnius, 2023

Disertacija rengta 2019–2023 metais Vilniaus Gedimino technikos universitete.

Vadovas

doc. dr. Raimondas JASEVIČIUS (Vilniaus Gedimino technikos universitetas, mechanikos inžinerija – T 009).

Vilniaus Gedimino technikos universiteto Mechanikos inžinerijos mokslo krypties disertacijos gynimo taryba:

Pirmininkas

prof. dr. Dalius MAŽEIKA (Vilniaus Gedimino technikos universitetas, mechanikos inžinerija – T 009).

Nariai:

prof. dr. Sergiy ANTONYUK (Kaizerslauterno-Landau universitetas, Vokietija, mechanikos inžinerija – T 009).

prof. habil. dr. Rimantas BARAUSKAS (Kauno technologijos universitetas, mechanikos inžinerija – T 009),

prof. dr. Julius GRIŠKEVIČIUS (Vilniaus Gedimino technikos universitetas, mechanikos inžinerija – T 009),

prof. dr. Arnas KAČENIAUSKAS (Vilniaus Gedimino technikos universitetas, mechanikos inžinerija – T 009),

Disertacija bus ginama viešame Mechanikos inžinerijos mokslo krypties disertacijos gynimo tarybos posėdyje **2023 m. gruodžio 20 d. 10 val.** Vilniaus Gedimino technikos universiteto SRA-I Posėdžių salėje.

Adresas: Saulėtekio al. 11, LT-10223 Vilnius, Lietuva. Tel. (8 5) 274 4956; faksas (8 5) 270 0112; el. paštas doktor@vilniustech.lt

Pranešimai apie numatomą ginti disertaciją išsiųsti 2023 m. lapkričio 17 d.

Disertaciją galima peržiūrėti Vilniaus Gedimino technikos universiteto talpykloje http://dspace.vgtu.lt ir Vilniaus Gedimino technikos universiteto bibliotekoje (Saulėtekio al. 14, LT-10223 Vilnius, Lietuva).

Abstract

The most well-known and commonly used printing techniques made the inkjet printing technology more popular among manufacturers for its simplicity and flexibility. Demand for printing products is increasing in such areas as textile printing, microchip production and personalised production, raising quality requirements. Thus, inkjet printing is continuously enhancing its capabilities. With the printing market expected to grow, there are many propositions to improve the productivity of inkjet process. The dissertation aims to study the dynamics of an inkjet droplet at different stages of its formation, considering its interaction with the surface to improve the quality and speed of high-class inkjet printing and improve inkjet printhead testing applications. The present dissertation reviews theoretical and numerical techniques of inkjet printing process modelling by applying the Computational Fluid Dynamics (CFD) module using the finite elements method. Inkjet printing inks are calculated as Newtonian fluid. The ink flow is modelled by the incompressible Navier-Stokes equations. The presented simulation model is solved using a level set method to determine the ink and air interface. Experimental investigation results are reported on the inkjet printing process colour reproduction on different linen fabrics, and modelling results are given for droplet ejecting, printing and interaction processes with the flat surface and the interaction with the inkjet droplet with physically treated polymer surfaces.

Based on systematic and analysed numerical simulations of research on the operation of inkjet printing heads, Article 1 describes the inkjet printhead models, which analyse how different inks impact droplet formation and movement at different time steps.

Article 2 presents an experimental realisation of colour reproduction by inkjet printing possibilities on different materials in different printing settings.

Article 3 presents the physical and numerical experiments on the impact characteristics of inkjet printhead droplets with flat surfaces. Different surface treatment methods were investigated to increase droplet adhesion with flat surfaces.

Article 4 investigates the numerical model of the print head modelled in threedimensional space. Various parameters of drop formation and fall were experimentally determined with a change in the time of the excitation impulse. A numerical analysis was carried out regarding the influence of the excitation time on the droplet formation and further movement.

The supplemented articles were published in four peer-reviewed scientific journals listed in the *Clarivate Analytics Web of Science* database with an impact factor. The study results were presented at three international conferences.

Reziumė

Atsižvelgiant į žinomus ir dažniausiai taikomus spausdinimo būdus, išlieka svarbi rašalinio spausdinimo technologija, kuria gamintojai vis dažniau taiko dėl jos paprastumo ir lankstumo. Didėjanti gaminių spausdinimo paklausa tokiose srityse kaip audinių marginimas, mikroschemų gamyba ar individualizuota gamyba kelia vis didesnius reikalavimus spausdinimo kokybei. Taigi, rašalinio spausdinimo kokybė ir spausdinimo proceso galimybės nuolat plečiasi. Tikimasi, kad spausdinimo rinka augs, todėl yra daug pasiūlymų gerinti rašalinio spausdinimo proceso efektyvuma. Disertacijos tikslas – ištirti skaitmeninės rašalinės spaudos metu susidariusio lašelio dinamika skirtingais jo formavimosi etapais, apimant šio lašelio saveika su paviršiumi, siekiant pagerinti spausdinimo kokybę ir greiti bei tobulinti spausdinimo galvučių testavimo programas. Disertacijoje aptariami teoriniai ir skaitmeniniai rašalinio spausdinimo proceso modeliavimo metodai taikant skaičiuojamaja skysčių dinamiką (CFD) ir baigtinių elementų metoda. Rašalo lašeliai modeliuojami kaip niutoninis skystis. Rašalo srautas modeliuojamas remiantis nespūdžiam skysčiui taikomomis Navjė-Stokso lygtimis. Pateiktas skaitinis modelis taikomas rašalo ir oro sasajai nustatyti, pasitelkiant lygio nustatymo (angl. *level set*) metoda. Pateikiami rašalinės spaudos ant įvairių lininių audinių spalyų atgaminimo proceso eksperimentinių tyrimų rezultatai bei lašelio išstūmimo, spausdinimo ir saveikos su plokščiuoju paviršiumi, rašalo lašelio saveikos su fiziškai apdorotais polimeriniais paviršiais procesų modeliavimo rezultatai.

Atlikus skaitinį eksperimentą, 1 straipsnyje aprašomas sudarytas skaitmeninės spausdinimo galvutės modelis. Modelis pritaikytas nagrinėti spausdinimo rašalų parametrų įtaką, spaudos lašelio parametrus.

2 straipsnyje aprašomas fizinis eksperimentas, atliktas nagrinėjant spalvų atgaminimo savybes ant skirtingų audinių esant skirtingiems spausdinimo parametrams.

3 straipsnyje pristatomi skaitinio ir fizinio eksperimento rezultatai, kai spausdinama ant skirtingų lygių paviršių. Išnagrinėti skirtingi paviršiaus apdirbimo metodai, siekiant nustatyti lašelio ir paviršiaus sąveikos aplinkybes.

4 straipsnyje sudarytas skaitinis spausdinimo galvutės modelis trimatėje erdvėje. Eksperimento būdu nustatyti skirtingi lašelio formavimosi ir kritimo parametrai esant skirtingiems galvutės sužadinimo parametrams (t; V_{in}). Atlikta skaitinė analizė ir nustatyta sužadinimo laiko įtaka generuojamam spausdinimo lašelio formavimuisi, kritimo greičiui ir masės pokyčiams.

Disertacijoje išvardyti straipsniai yra publikuoti 4 recenzuojamuose moksliniuose žurnaluose, įtrauktuose į *Clarivate Analytics Web of Science* duomenų bazę, su cituojamumo rodikliu. Disertacijoje aprašomi tyrimų rezultatai pristatyti trijose tarptautinėse konferencijose.

Notations

Symbols

m_i – mass of the particle *I* (liet. *dalelės masė*);

 \mathbf{x}_i – position vector of the particle *I* (liet. *dalelės pozicijos vektorius*);

 θ_i – orientation vector of the particle *I* (liet. *dalelės krypties vektorius*);

 \mathbf{F}_i – resultant force acting on the particle *I* (liet. *dalelę veikianti atstojamoji jėga*);

 \mathbf{T}_{i} – torque on the particle *I* (liet. *dalelės sukimo momentas*);

I_i – inertia moment of the particle I (liet. dalelės inercijos momentas);

d-diameter of the nozzle (mm) (liet. purkštuko skersmuo);

D_{d.max} – maximum droplet diameter (µm) (liet. *didžiausias lašelio skersmuo*);

D_h – droplet height (µm) (liet. *lašelio ilgis*);

 F_{st} – surface tension force (N) (liet. *paviršiaus įtempimo jėga*);

m_d – the mass of the droplet (kg) (liet. *lašelio masė*);

n – unit vector in the normal direction (liet. *vienetinis vektorius normaline kryp-timi*);

Z – ink printability number (dimensionless coefficient) (liet. *rašalų spausdinamumo rodiklis*);

Oh – Ohnesorge number (dimensionless coefficient) (liet. Onesorgo skaičius);

p – pressure (Pa) (liet. *slėgis*);

r – radius of the droplet (mm) (liet. *lašelio radiusas*);

g – gravitational constant (m/s²) (liet. gravitacijos konstanta);

Re-Reynolds number. Dimensionless coefficient (liet. Reinoldso skaičius);

t – time (s) (liet. *laikas*);

u – vector of fluid velocity (m/s) (liet. skysčio greičio vektorius);

 V_d – droplet volume (pL) (liet. *lašelio tūris*);

We-Weber number (dimensionless coefficient) (liet. Vėberio skaičius);

 W_a – work of the adhesion (J/m²) (liet. *adhezijos darbas*);

 β – maximum spreading factor (liet. *didžiausias sklaidos koeficientas*);

 γ – parameter which determines the repetition of initiations (liet. *parametras, ku*-

ris lemia iniciacijų pasikartojimą);

 δ – Dirac delta function (liet. *Dirac delta funkcija*);

 ε – representative mesh size in the area passed by the droplet (liet. *atvaizduojamas tinklo dydis per kuri praeina lašelis*);

к – curvature (liet. *kreivė*);

 μ – dynamic viscosity (Ns/m²) (liet. *dinaminė klampa*);

 ρ – density (kg/m³) (liet. *tankis*);

 σ – surface tension coefficient (mN/m) (liet. *paviršiaus įtempimo koeficientas*);

 ϕ – coefficient of level set interface between air and ink (liet. *sąveikos koeficientas tarp oro ir rašalo*);

s – standard deviation (liet. standartinis nuokrypis);

 ∇ – Nabla operator (liet. *Nabla operatorius*);

*V*_{in} – ink velocity at inlet (m/s) (liet. *rašalų įtekėjimo greitis*);

 θ – droplet contact angle with surface (°) (liet. *lašelio drėkinimo kampas paviršiuje*);

 ΔE – colour measurement difference (dimensionless number) (liet. *spalvos poky-tis*);

L*-colour value of the lightness parameter (liet. *spalvos šviesumo vertė*);

*a** – colour value from greenness to redness (liet. *spalvos vertė kintant nuo žalios iki raudonos spalvos*);

*b** – colour value from blueness to yellowness (liet. *spalvos vertė kintant nuo mėlynos iki geltonos spalvos*).

Abbreviations

DOD – drop on demand (liet. *lašelis pagal poreikį*);

DEM - discrete element method (liet. diskrečiųjų elementų metodas);

FEM – finite element method (liet. *baigtinių elementų metodas*);

CFD - computational fluid dynamics (liet. skaičiuojamoji skysčio dinamika);

NS – Navier-Stokes (liet. Navjė-Stokso);

HDPE – high-density polyethylene (liet. *didelio tankio polietilenas*);

- IPA isopropyl alcohol (liet. *izopropilo spiritas*);
- PET polyethylene terephthalate (liet. *polietileno tereftalatas*);
- PP polypropylene (liet. *polipropilenas*);
- SE surface energy (liet. *paviršiaus energija*);
- ST surface tension (liet. paviršiaus įtempimas);
- GT the classification value (liet. *klasifikacijos vertė*);
- OBR linen textile abbreviation (liet. lino tekstilės abreviatūra).

Contents

AUTHOR'S CONTRIBUTION TO THE PUBLICATIONS	XV
INTRODUCTION	1
Problem Formulation	1
Relevance of the Dissertation	2
Research Object	2
Aim of the Dissertation	3
Tasks of the Dissertation	3
Research Methodology	3
The Scientific Novelty of the Dissertation	4
The Practical Value of the Research Findings	5
The Defended Statements	5
Approval of the Research Findings	5
Structure of the Dissertation	6
1. REVIEW OF INKJET PRINTHEAD STRUCTURES AND SIMULATION OF	
DROPLET MOVEMENT APPLICATIONS	7
1.1. Constitutive models for numerical simulation of solid microparticle	
movement	
1.1.1 Concept of modelling with the discrete element method	8
1.1.2 Mathematical model of the narticle movement	9
1.1.3 Modelling the motion of a solid microparticle	ر و
1.2 Movement of a solid particle as a droplet	
1.2. Hovement of a sona particle as a dioplet	1 1

1.2.1. Modelling with computational fluid dynamics by COMSOL	11
1.2.2. Inkjet printhead structures and working principles	12
1.2.3. Inkjet droplet turnover during printing processes	13
1.3. Conclusions of the First Chapter	14
2. INKJET PRINTING PROCESS RESEARCH METHODOLOGY	15
2.1. Experimental evidence of numerical modelling	15
2.1.1. Application of the Navier-Stokes equation for droplet movement	16
2.1.2. Interface between the ink and air phases	16
2.1.3. Density and dynamic viscosity	17
2.1.4. Numerical realisation of the droplet motion	17
2.1.5. Droplet final mass calculation	18
2.1.6. Dimensionless numbers in inkjet printing parameters	19
2.1.7. Contact angle calculated from the stability of forces according	
to Young's equation	20
2.2. Characterisation of the colour reproduction measurement	21
2.2.1. Differences in ΔL^* , Δa^* and Δb^* values	22
2.2.2. Mean value and standard deviation measurement	22
2.3. Characterisation of the surface of polymers evidence of the inks and surface	
adhesion measurement	23
2.4. Conclusions of the Second Chapter	25
•	
3. INVESTIGATION RESULTS AND CONCLUSIONS OF THE INKJET	
DROPLET FORMATION AND MOVEMENT MODELLING	27
3.1. Investigation results of Article 1	27
3.2. Investigation results of Article 2	30
3.3. Investigation results of Article 3	33
3.4. Investigation results of Article 4	35
3.5. Conclusions of the Third Chapter.	37
GENERAL CONCLUSIONS	39
REFERENCES	41
AUTHOR'S PUBLICATIONS COLLECTION	51
Article 1. Tofan, T. et al. (2021). Numerical Modeling of the Motion and	
Interaction of a Droplet of Inkiet Printing Process with a Flat Surface	
(https://doi.org/10.3390/app11020527)	. 53
Article 2, Tofan, T. et al. (2021). Investigation of Colour Reproduction on	
Linen Fabrics when Printing with Mimaki TX400-1800D Inkiet with	
Pigment TP250 Dves (https://doi.org/10.3390/coatings11030354)	68
Article 3. Tofan, T. et al. (2021). Modelling of the Motion and Interaction of a	
Dronlet of an Inkiet Printing Process with Physically Treated Polymers	
Substrates (https://doi.org/10.3390/app112311465)	
~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~	

odeling 3D Droplet Movement Using	Article 4. Tofan, T. et al. (2022).
thead Model	a Drop-on-Demand Inkjet P
81467)	(https://doi.org/10.3390/pr1
	SUMMARY IN LITHUANIAN

# Author's contribution to the publications

Publication ¹	Formal contribution ²	Conceptualisation	Data curation	Formal analysis	Investigation	Methodology	Software	Validation	Visualisation	Writing – original draft	Writing – review & editing
Tofan et al., 2021	0.25	sole	joint	main	joint	main	main	sole	joint	joint	main
Tofan et al., 2021	0.34	main	main	sole	sole	sole		joint	sole	sole	sole
Tofan et al., 2021	0.5	sole	main	sole	sole	joint	sole	main	sole	sole	sole
Tofan et al., 2022	0.25	joint	joint	sole	main	sole	joint	joint	joint	sole	sole
Total or max ³	1.334	main	main	sole	sole	sole	main	joint	main	sole	sole

¹ The published articles have been used here with the permission of the relevant publishers.

 2  The formal contribution is calculated as a fraction  $- 1/N_{authors}$ .

³ The total sum of the formal contribution values or the highest contribution achieved (in the increasing order: none, joint, main, or sole) in the specified 10 of 14 roles (according to the CRediT taxonomy, https://credit.niso.org/).

All the above-mentioned articles' co-authors have no motive to use this published data to prepare other dissertations.

All the authors of the above-mentioned articles have agreed on the author's contribution statement.

### Introduction

#### **Problem Formulation**

The interaction behaviour of different matter states is important to understand from the viewpoint of mechanics. Microparticle handling is important for the pharmaceutical, food, chemical, printing and other industries. The improved understanding and the ability to simulate them numerically may help to better understand flow processes for science and industry. The microparticle considered in the investigation is a droplet that can deform and stick to the surface when moving and interacting. It is also important to understand droplet behaviour when it loses shape interacting with a surface. Quantification of particle behaviour requires experimental evidence. The printing industry is no exception.

According to Smithers. (2020) report, the inkjet printing industry will continue to expand faster than other printing forms, with decreasing circulation in existing markets and more flexible printing options. The expanding inkjet printing market proposes researching the productivity and quality of printing processes. Nowadays, inkjet printing technology is used globally in many manufacturing applications (Castrejon-Pita et al., 2013; Takagi et al., 2019; Huang et al., 2020; Maisch et al., 2021; West and Yoo, 2023).

Many research efforts are available, focused on different inkjet printhead structures, various jetting parameters, and inkjet droplet movement and impact with various surfaces (Son et al., 2008; Bussmann et al., 2000; Kang et al., 2020;

Yang et al., 2022; Li et al., 2022; Xiao et al., 2022; Mau and Seitz, 2023; Wang, and Chiu, 2023). Researchers are thereby concentrated on developing new printhead applications and ejection methods (Okuyama and Yoshida, 2018; Aqeel et al., 2019; Shah et al., 2019; Yoshida et al., 2019; Sabu, 2022; Wang et al., 2023). Many works explore the properties of the printhead nozzles, such as dimensions, orifice parameters, and droplet excitation parameters that determine the velocity and parameters of the formed droplets, as well as the conditions for the formation of satellite droplets (Jiang and Tan, 2018; Driessen, 2018; Li et al., 2019; Khan et al., 2021; Rump et al., 2023). Nowadays, there is growing interest in variable fluid inkjet droplet formation and movement control, reflecting many practical applications such as high-quality printing without the loss of speed (Wang and Chiu, 2020; Kamis et al., 2021; Peng et al., 2022; Wang et al., 2023).

Numerical experimental modelling could help evaluate droplet ejection, shape-changing droplet motion, various initial parameters, and effects influencing droplet motion, which are key components in the inkjet process. This dissertation concentrated on the experimental and theoretical investigation of the formation and movement of inkjet droplets. It is characterised by aspects of numerical simulation and characteristics of the conditions for their development.

#### **Relevance of the Dissertation**

The digital ink print method quickly gained popularity in the world as the printing circulation volumes decreased, yet the need to print on different materials remained. To match the high levels of print quality and print speed, it is necessary to provide the corresponding parameters of ink droplets, their movement and interaction. The studied parameters allow for determining which print speed can be chosen without losing print quality.

Aspects of the digital ink parameters and their numerical research are the novelty of this work. Relevance is based on the recommendations of the printing process and numerical studies related to the quality of the press.

#### **Research Object**

The object of the research is a droplet of ink used in the process of digital inkjet printing.

#### Aim of the Dissertation

The dissertation aims to investigate the dynamics of an inkjet droplet at different stages of its formation, considering its interaction with the surface.

#### **Tasks of the Dissertation**

Considering the review of well-known scientific literature in the collection of dissertation publications during the preparation of the dissertation, the following novelty was established in modelling and analysing the behaviour of the movement of an inkjet droplet in the process of digital inkjet printing:

- 1. To investigate the conditions of formation and movement of an inkjet droplet by physical and numerical research methods; determine the parameters according to which the ink droplet is formed and evaluate the influence of different factors on the ink printing process; to analyse droplet movement modelling methods.
- 2. To numerically investigate the modelling capabilities of the inkjet printhead, including droplets ejection parameters, droplets formation and movement as the droplet leaves the nozzle, so that to study their velocity, volume, shape variation, and satellite formation, as well as the influence of different surface parameters on the interaction of the droplet with the surface.
- 3. To determine the influence of ink physical parameters on droplet formation and movement towards the surface. Physically examine the influence of printing parameters on colour characteristics and investigate the circumstances of droplet interaction with different surfaces according to surface characteristics.
- 4. To create a numerical model to study the droplet formation of the ink printhead and the droplet movement in the air, including the parameters of different ink and ejection parameters such as droplet generation time and ink feed velocity. To offer solutions for developing inkjet print heads and improving print quality without performance loss.

#### **Research Methodology**

Experimental and numerical research methods were used to study the object of this research. Digital inkjet printing ink is generated in the form of a droplet, which is modelled by applying the finite element method using the COMSOL Multiphysics software. Reproduction of colour characteristics on different surfaces on different printing parameters was measured using a spectrophotometer and processed in the *Barbieri Rasterlink* software. Dyes of Plasmatreat, which have different surface tension, are used to determine changes in the surface energy of the printing surface. The adhesive properties of the printed surface were checked in accordance with the standard DIN ISO 2409. The obtained results are analysed by changes in various parameters depending on the initial parameters.

#### The Scientific Novelty of the Dissertation

During the preparation of the dissertation, the novelty of modelling the droplet formation and movement in digital inkjet printing was determined:

- 1. The effect of different printing inks on the printing process was numerically investigated. The results show that depending on the physical properties of the inks, the droplet interaction parameters and interaction duration change. The initial stages of the droplet formation in the inkjet printing process were determined, and the turnover at the droplet's shape and movement speed were recorded through the vertical axis.
- 2. The effect of different printing parameters on the reproduction of colour characteristics on surfaces of different characteristics was physically determined by evaluating the spectral properties of the print in the CIE  $L^*a*b*$  scale.
- 3. Physical changes were determined in the parameters of polymeric materials with different processing methods, and the adhesive properties of the inks were evaluated according to the DIN ISO 2409 standard. The interaction of an inkjet droplet with surfaces of different surface energy was numerically modelled by recording the droplet's dipping angles and movement speeds.
- 4. The ejecting process, formation, movement and interaction of inkjet droplets were modelled using COMSOL simulation CFD module software. The model allows for studying the droplet thread length and the moment when it is pulled out of the thread, as well as the shape, mass, volume and behaviour of the droplet when it hits the surface and its final state. In addition, the numerical model made it possible to configure different nozzle parameters and different ejection pressures and test different ink types.

#### The Practical Value of the Research Findings

The dissertation explains multiple issues, including experimental and theoretical studies, to determine, analyse and understand the ongoing processes of formation and movement of inkjet droplets under various operating conditions. A few important parameters, such as ink mass, droplet size and the shape of the ink droplet, have been obtained, and the contribution of different factors, such as ink characteristics, printhead ejection parameters and the nozzle form/shape, has been studied. The proposed numerical model of the inkjet printhead could be easily implemented/realised by using the COMSOL Multiphysics CFD software.

Applying the 3D CFD simulation model studied in the dissertation could help investigate the development of ink, the design of inkjet printheads and the creation of new digital printers. The presented numerical model and methods appreciate such parameters as the ejection dwelling time, the mass and diameter of the droplet, falling speed, and the change in the shape of the droplet.

#### **The Defended Statements**

The following statements are based on the results of the present investigation and may serve as the official hypotheses to be defended:

- 1. The proposed numerical model of the printhead allows for investigating the influence of parameters (*u*; D_{d.max}; D_h; m_d), which determines the shape of the ink droplet at each time step.
- 2. Regulating the ejection settings of the inkjet printhead allows for getting the same droplet covering area on the surface by different ink types.
- According to the numerical experiment, the physical properties of the ink (μ; ρ; σ; *Oh*; *Z*) have a decisive influence on the drop behaviour during printing, especially when the drop interacts with different surface types. This way, the effect on the formation and movement of the droplet was investigated by changing the physical ink properties.

#### Approval of the Research Findings

The results of the dissertation have been published in 6 scientific publications: 4 were published in journals, included in *Clarivate Analytics Web of Science* databases with a citation index, 2 publications in conference proceedings.

The author has made 3 presentations at international scientific conferences:

 International Conference of Numerical Analysis and Applied Mathematics 2020 (ICNAAM 2020), Rhodes, Greece.

- International Conference of Numerical Analysis and Applied Mathematics 2021 (ICNAAM 2021), Rhodes, Greece.
- 10th International Conference on Conveying and Handling of Particulate Solids (5–9 July 2022), Salerno, Italy.

#### Structure of the Dissertation

The dissertation includes an introduction, analytical literature review, discussion of research methodology, summarized investigation results and conclusions, references, four appended scientific articles and a summary in Lithuanian.

The first chapter reviews the studies of inkjet printhead structures, working principles and droplet formation parameters.

The second chapter reviews theoretical, numerical, and experimental study's methodology to identify the droplet formation of an inkjet droplet, modelling the movement of the ejected droplets and the impact condition of the droplet with different surfaces.

The third chapter summarises the experimental results of experimental and numerical investigations of appended articles 1–4 in general perception.

The total scope of the dissertation is 118 pages, and contains 34 equations, 15 figures, 3 tables and 104 references.

1

### Review of Inkjet Printhead Structures and Simulation of Droplet Movement Applications

This chapter provides a general review of the working principles of inkjet technology, discusses ink requirements and droplet formation fundamentals, deliberates on the constitutive models of numerical simulation, reviews the mathematical model of the particle movement, introduces the discrete element method modelling, investigates the movement of solid microparticle as a droplet by finite element method and applies computational fluid dynamics by COMSOL. Based on the published articles, various possible modelling methods are presented. The possibilities of modelling a micro-object are indicated, taking it as a solid and liquid object. Both cases are described in the chapter. The emphasis is on inkjet technology, which is the main focus of this work. This chapter concludes by the formulated main objective and tasks of the investigation Tofan, et al., 2021; Tofan, et al., 2022.

# **1.1. Constitutive models for numerical simulation of solid microparticle movement**

Before analysing the droplet motion, the microparticle should also be mentioned as a solid. Microparticles are considered to be microfine particles. Representing microparticles as a modelled object, a solid particle is also an important component in modern modelling, which can provide knowledge about contact mechanics (Tomas, 2000, 2004, 2007; Antonyuk et al., 2006; Dowding et al., 2020; Sarmadi et al., 2020; Leontev et al., 2023).

Particle handling is especially significant in pharmaceutical, food, cement, chemical, printing and other industrial processes (Jiang et al., 2021; Paul and Roy, 2022). Particle size reduction elevated particle adhesion to a new level of importance; therefore, understanding the underlying fundamentals is particularly essential in particle technology (Tykhoniuk et al., 2007; Maramizonouz et al., 2021; Kim and Lee, 2022; Wang et al., 2023)

Quantification of particle behaviour requires experimental evidence. For example, direct measurement of surface and contact forces by an Atomic Force Microscope. Experiments on colliding spheres with a particular emphasis on silica particles were conducted (Tofan, 2022; Rotter et al., 2023). Despite huge progress in experimental techniques, direct addressing of individual particles is still limited and expensive. In addition, the solution of an inversed problem is necessary for particle characterisation.

The stiff spherical particle is modelled by applying the Discrete Element Method, while initially, the spherical droplet was simulated with COMSOL software. Emphasis is put on studying various microparticle impacts by considering different matter states. The following paragraphs comprise problem motivation and description, simulation methodology and discussion of numerical results.

#### 1.1.1. Concept of modelling with the discrete element method

The discrete element method (DEM) introduced by Cundall and Strack (1979) has become a powerful alternative for solving scientific and engineering powder technology problems. This numerical method is based on the Lagrangian approach applied to a multi-particle system, meaning that particles are treated as individual objects, and all dynamical parameters (position, velocity, orientation, etc.) of each particle are tracked during the simulation (Zhu et al., 2008; Peng et al., 2020; El-Emam et al., 2021; Kuang et al., 2021).

With the discrete element method, the microparticle motion is described in the context of a Lagrangian framework influenced by nonlinear interaction forces. Based on DEM simulations, microparticle interaction models will be applied, which allow a description of the general characteristics of the particle behaviour, such as the coefficient of restitution for an elastic and elastic-plastic solid particle (Tofan et al., 2022; Wu et al., 2022; Zhu et al., 2023).

Thereby, the assumption is made that the microparticle behaviour during its impact is governed by mechanical deformation processes. Interaction models will be applied for the simulation of the liquid droplet movement when the initial interaction velocity with the surface is similar to that of the solid particle. From the modelling point of view, the microparticle behaviour can be described throughout the motion towards a surface, interaction at a distance and interaction when the microparticle has made contact.

#### 1.1.2. Mathematical model of the particle movement

The DEM methodology based on the Lagrangian approach is applied to simulate the dynamic behaviour of the cohesive particles under normal impact.

The motion of the arbitrary particle i is characterised by a few global parameters: particle position velocities (1.1) and accelerations of the mass centre and force applied (1.2).

$$m_i \frac{\mathrm{d}^2 \mathbf{x}_i}{\mathrm{d}t^2} = \mathbf{F}_i(t); \tag{1.1}$$

$$I_i \frac{\mathrm{d}^2 \mathbf{\theta}_i}{\mathrm{d}t^2} = \mathbf{T}_i(t). \tag{1.2}$$

Translational motion is described by Newton's second law applied to each particle *i*, where  $m_i$  is the mass,  $\mathbf{x}_i$  is the position vector;  $\mathbf{\theta}_i$  orientation vector while vector  $\mathbf{F}_i$  presents the resultant force acting on the particle *i* and  $\mathbf{T}_i$  present torque acting on the particle *i* during the contacts with *j* neighbour particles. I_i is the inertia moment of the particle *i*.  $\mathbf{F}_i$  may, thereby, comprise prescribed and field forces. The analysis and implementation of basic forces, which act on a particle, can be realised as part of a particle interaction model.

#### 1.1.3. Modelling the motion of a solid microparticle

Independently of the development of the DEM, a general contact model has been introduced for small particles comprising elastic-plastic, adhesion and dissipative behaviour (Jasevičius et al., 2007). Regarding the observation that ultrathin  $d < 0.1 \mu m$ , Iijima (1985) noted that there is no consensus. Many research efforts on microparticles are based on the theory (Tomas, 2007, 2009) of defining the diameter of ultrafine particles 0.1  $\mu m < d < 10 \mu m$ .

There are two different cases of particle motion in Fig. 1.1 (Jasevičius et al., 2015).



Fig. 1.1. Representation of non-attractive and non-dissipative particle movement (Jasevičius et al., 2015)

Fig. 1.2 represents particle motion, where adhesion and dissipation are not considered. Here, positive displacement h > 0 means contact compression, while negative h < 0 means approach or detachment. H₀ is the initial height. In this situation, the particle will rebound to its initial position. Such a situation could be characteristic of larger than ultrafine particles, for which the effect of adhesion is insignificant.



Fig. 1.2. Representation of attractive-dissipative particle movement (Jasevičius et al., 2015)

If a microparticle is modelled, the attractive zone in which the van der Waals force is applied must be considered during the particle motion. Due to the small microparticle mass, the attractive force can be large enough to bring such a small particle to a surface and let it stick there.

All main required forces for interaction, such as adhesive, viscous, elastic and elastic-plastic contributions, can be considered part of a unified model. Even deformations of biological microparticles, such as cells, can be described with the dissipative mechanism related to adhesion, which was originally developed for non-biological microparticles (Müller et al., 2013; Creton and Ciccotti, 2016). The 3D extension of the deformable discrete element method (DDEM) developed for 2D problems partly evaluates the deformation of the particles' deformability (Rojek et al., 2021).

#### 1.2. Movement of a solid particle as a droplet

Unlike the solid particle movement modelling, not all methods can be applied to study the droplet motion. Such motion additionally requires an assessment of the change in particle shape during its motion. Many scientific works have examined solid particles (Derby, 2011; Yoo & Kim, 2015; Hoath, 2016; Sun et al., 2020; Wang et al., 2022.). Meanwhile, when analysing the motion of the droplet, the change in the droplet shape and the possibilities of modelling are still unclear (Das et al., 2017; Zhang et al., 2023). Droplet motion analysis is associated with a printing process in which a droplet is ejected onto the surface. Therefore, the relevance of a droplet/surface contact is based on printing process recommendations and mechanical research related to print quality.

With the rapid growth in demand for printing on different materials, the digital inkjet printing method is becoming increasingly relevant in the manufacturing process. This presentation is related to the application of the digital inkjet printing method and the study of droplet mechanics applied in the printing process. An ink droplet is generated only when an image element needs to be formed. Appropriate droplet formation (ejection) settings must be selected to ensure print speed and quality. By controlling them, it is possible to maximise the print speed without losing print quality.

The ink droplets are not electrified and always move directly to the surface of the substrate point, so the writing head can be positioned at different heights to the surface (from 1 to 5 mm). The print head configuration and the frequency range of the piezoceramic element allow for precise droplet positioning and result in high print quality.

## 1.2.1. Modelling with computational fluid dynamics by COMSOL

Computational fluid dynamics is an integral part of a constantly growing number of development processes and is a well-established field within many different engineering disciplines: mechanical, chemical, civil, aeronautical, and more specialised areas, such as printing, biomedical engineering (Makowski et al., 2021; Mbanjwa et al., 2022; Lei et al., 2022).

Often, the flow itself is not the focus of a simulation. Instead, it is important to know how the flow affects other process and application parameters. In many situations, while the flow can add necessary operational parameters to a process or application, it is also affected by them (Nguyen et al., 2021; Wang et al., 2022).

A description combining multiple physical fields is often needed to produce precise models known as fluid flow applications. Being able to effectively simulate such models increases the understanding of the studied processes and applications, which in turn leads to the optimisation of the flow and other parameters. Today's engineers can easily use these software modelling tools and create realistic models.

Simulations can be useful to improve the understanding of the fluid flow and to predict the optimal design of an inkjet for a specific application. The purpose of this application is to adapt the shape and operation of an inkjet nozzle for a desired droplet size, which depends on the contact angle, surface tension, viscosity, and density of the injected liquid. The results also reveal whether the injected volume breaks up into several droplets before merging into a final droplet at the substrate.

#### 1.2.2. Inkjet printhead structures and working principles

Inkjet printing technology has gained prominence as the dominant printing technology for home, office and manufacturing. In addition, over the past decade, inkjet technology has acquired acceptance in the technical community as a highperformance tool for micromanufacturing processes (Korvink, 2012; Chen et al., 2013; Fischer et al., 2020; Giusti, 2022). The inkjet droplet physics description while in motion and its practical applications have been investigated and described in detail (Wijshoff, 2010; Lee, 2018; Liu and Derby, 2019; Huang, 2020).

The fundamental advantage of inkjet printing technology is the possibility of generating various-sized droplets with precision predetermined locations on the surfaces of many different materials without contact. The inkjet printing technology can produce low-quantity prints at high-quality requirements.

In the drop on demand (DOD) mode, an inkjet technology droplet is created only when preferred. It can be realised in two different ways: a piezoelectric transducer or heated resistor which, while heating, produces a bubble inside the nozzle (Oktavianty et al., 2019; Kang et al., 2020; Zhao et al., 2021; Shah et al., 2021; Garduno et al., 2023). These techniques trigger a pressure wave at the ink reservoir, which expels a droplet ejection from the printhead nozzle (Das et al., 2021; Oktavianty et al., 2022; Lohse, 2022;)

Since a droplet is created only when it is needed, DOD systems are conceptually far less complex than continuous mode systems. This system can generate different-size droplets by different signal data of the piezo driver to the piezoelectric transducer, regulating the dwelling time and energy magnitude (Fig. 1.3).



Fig. 1.3. Schematic of a demand printing system and droplet (Korvink, 2012)

#### 1.2.3. Inkjet droplet turnover during printing processes

During the ejection of a droplet from the inkjet nozzle, it falls by changing its shape and physical parameters. A numerical simulation can be applied to observe different parameters, such as shape, falling velocity, transformation speed, mass and volume turnover (Solanki et al., 2022; Sinha et al., 2023; Segura et al., 2023).

The droplet's form changes at different time steps, which are represented by considered periods (Fig. 1.4). These steps help to define what a droplet will look like over time.



Fig. 1.4. Main steps of droplet formation and impact on the surface (Tofan, 2021)

Droplet formatting phases are represented by the following steps: the first step (A) represents the maximum droplet velocity when ink is ejected; the second step (B) is when the droplet thread pitches off from the inkjet nozzle; at this point, the droplet has the maximum length size, and satellites could appear from the thread if incorrect parameters are set; the third step (C) represents the droplet in a sphere form; then, the droplet thread and satellites shrinking into the droplet by connecting to the final droplet; the fourth step (D) represents the droplet before the start of the interaction with the surface; the fifth step (E) represents the droplet at a maximum spreading diameter size; and the final point step (F) represents the droplet in the final-stable form after the oscillation processes.

#### 1.3. Conclusions of the First Chapter

Based on the literature investigation, the following conclusions have been drawn:

- 1. A stiff particle can keep the spherical shape during impact (of the stiff particle with a soft contact), and the discrete element method can be applied. During movement and impact, a droplet continually changes shape. Thus, for a single particle motion as a droplet, numerical modelling of FEM and CFD software must be applied.
- 2. By observing the changing shape of the droplet over time, the droplet motion and interaction can be better explained by considering the separate periods. It is suggested to divide the movement of a droplet into six stages: droplet ejection, droplet thread pull off the nozzle, droplet sphere form formation, droplet before interaction, droplet interaction (spreading) at the substrate surface and droplet stability phase.
- 3. During the droplet ejection and movement in the printing process, it is important to observe the droplet form and velocity turnover at the falling phase. The second important part is the droplet's impact on the surface. These parameters could be obtained and examined by applying the numerical simulation of the inkjet nozzle model;
- 4. The creation of a numerical model for studying the droplet formation and motion behaviour of droplets could be helpful for developers to improve inkjet printhead structure and inkjet ink creation. Using the numerical model of the inkjet printhead, it is possible to observe the droplet thread length turnover, the droplet parameter changes while detaching from the nozzle, the droplet form, mass and volume turnover, the droplet impact conditions, and the droplet characteristics at the stable state on the surface. Additionally, numerical modelling is reliable for examining different inks and ejection settings and configuring different nozzle parameters.

2

### Inkjet Printing Process Research Methodology

This chapter conducts a significant number of theoretical, numerical, and experimental studies to identify the most important parameters influencing the formation of an inkjet droplet, modelling the movement of the ejected droplet and the interaction condition of the droplet for such practical applications as printing. The dissertation methodology discusses and explains the data collection and analysis of numerical modelling methods used in this research. This chapter provides a reliable and fundamental technique of numerical modelling of the inkjet printhead working principles considering CFD. Research was performed by using the finite element method numerical simulations (COMSOL Multiphysics) and physical experiments published in the author's publication Tofan et al., 2022.

#### 2.1. Experimental evidence of numerical modelling

The Navier–Stokes equation defines the momentum conservation equation of a viscous incompressible fluid in movement. The dynamics of droplet motion were modeled using the Navier–Stokes (N–S) equation.

### 2.1.1. Application of the Navier-Stokes equation for droplet movement

Numerical simulation based on the continuity equation and the N-S equation reconciles two-phase flows of incompressible substances. The continuity equation for an incompressible fluid is calculated by the equation:

$$\nabla \cdot \mathbf{u} = \frac{\partial \mathbf{u}_{\mathrm{x}}}{\partial \mathrm{x}} + \frac{\partial \mathbf{u}_{\mathrm{y}}}{\partial \mathrm{y}} + \frac{\partial \mathbf{u}_{\mathrm{z}}}{\partial \mathrm{z}} = 0, \qquad (2.1)$$

here,  $\nabla$  is the vector differential operator, and **u** is the vector of fluid velocity. By the definitions of calculus, the gradient operator ( $\nabla$ ) is calculated by the following equation:

$$\nabla = \frac{\partial}{\partial x} \mathbf{i} + \frac{\partial}{\partial y} \mathbf{j} + \frac{\partial}{\partial z} \mathbf{k}, \qquad (2.2)$$

Here, **i**, **j**, and **k** are the unit vectors in the directions of the x, y and z coordinates, respectively. The Navier–Stokes equation is intended to describe the momentum conservation equation for a viscous incompressible fluid in motion as follows:

$$\rho(\mathbf{u} \cdot \nabla)\mathbf{u} = \mu \left( \nabla \left( \nabla \cdot \mathbf{u} + (\nabla \cdot \mathbf{u}^{\mathrm{T}}) \right) \right) - \nabla p + \rho \mathbf{g}, \qquad (2.3)$$

here,  $\rho$  is the substance density; p is the pressure; g is the acceleration due to gravity;  $\mu$  is the substance viscosity.

#### 2.1.2. Interface between the ink and air phases

To describe the interface between the ink and air phases, the convection of the reinitialised level-set function is considered:

$$\frac{\partial \Phi}{\partial t} + \mathbf{u} \cdot \nabla \Phi = \gamma \nabla \cdot \left( \varepsilon \nabla \Phi - \Phi (1 - \Phi) \frac{\nabla \Phi}{|\nabla \Phi|} \right), \tag{2.4}$$

here, the coefficient  $\phi$  represents the specified set level of the ink–air interface. Accordingly,  $\phi = 0$  represents air and  $\phi = 1$  represents ink. In the transition layer close to the interface,  $\phi$  changes fluently/smoothly from 0 to 1. The vector of fluid velocity **u** represents the motion of the interface as it interacts with the environment. Here,  $\varepsilon$  represents the thickness of the transition layer. For the simulation,  $\varepsilon$  is taken as half the typical cell size in the region through which the falling drop passes. The interface thickness is controlled by default and set to 1.25 µm.

The parameter  $\gamma$  is the amount of stabilisation of the levelling (set level) function. The appropriate value of  $\gamma$  is the maximum value in the velocity field. This re-initialisation parameter  $\gamma$  is set (by default) to  $\gamma = 10$  m/s for the current simulation.

#### 2.1.3. Density and dynamic viscosity

When defining an interface between different substrates, the levelling (level set) function is used to stabilise density and viscosity calculations across the interface (Osher & Tsai, 2003; Sethian & Smereka, 2003; Lesage et al., 2007). In the numerical simulation, it is necessary to consider the dynamic viscosity (shear viscosity)  $\mu$ . It shows the interface between shear rate and shear stress in the substrate. The density and dynamic viscosity in various finite elements depend on the interface coefficient  $\phi$ . Dynamic viscosity and density are written as:

$$\rho = \rho_1 + (\rho_2 - \rho_1)\phi;$$
 (2.5)

$$\mu = \mu_1 + (\mu_2 - \mu_1)\phi, \qquad (2.6)$$

here,  $\rho_1$  is the air density, and  $\rho_2$  is the ink density;  $\mu_1$  is the viscosity of air, and  $\mu_2$  is the viscosity of the ink.

#### 2.1.4. Numerical realisation of the droplet motion

In numerical simulations, ink and air are studied as substrates, starting from a flow where viscous stresses appear at each point. The Navier–Stokes equations describe mass and pulse/momentum transfer in an incompressible fluid. Surface tension and gravitational forces are considered. The droplet motion is based on the conservation of mass and is calculated using differential equations:

$$\nabla \cdot \mathbf{u} = \mathbf{0}; \tag{2.7}$$

$$\rho\left(\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} - \mathbf{g}\right) - \nabla \cdot \left(\mu \left(\nabla \mathbf{u} + \nabla \mathbf{u}^{\mathrm{T}}\right)\right) + \nabla p = \mathbf{F}_{\mathrm{st.}}$$
(2.8)

The surface tension force is calculated according to the equation:

$$\mathbf{F}_{st} = \sigma \delta \kappa \mathbf{n}, \tag{2.9}$$

here,  $\sigma$  – surface tension coefficient;  $\delta$  – the Dirac delta function;  $\kappa$  – curvature; **n** – the unit vector (normal direction). The unit vector is calculated according to the equation:

$$\mathbf{n} = \frac{\nabla \Phi}{|\nabla \Phi|}.\tag{2.10}$$

At the fluid interface  $\delta$  equals a Dirac delta function approximated by the following equation:

$$\delta = 6|\phi(1 - \phi)||\nabla\phi|. \tag{2.11}$$

Mass conservation must be considered to calculate droplet motion and an adaptive mesh refinement function is used to refine the ink–air interface mesh locally. To calculate droplet motion using FEM, the initial 3D FEM mesh within

both chambers was generated automatically. To reflect the real change of droplet shape, the re-meshing and adaptive local mesh refinement are involved in improving the mesh at the ink-air interface. Mesh properties during simulation. The variation of the element number, the mesh quality factor and the average elements' skewness is shown in Fig 2.1.



Fig. 2.1. Representation of an adaptive mesh refinement sequence (Tofan, 2022)

This function will essentially divide the simulation run into multiple time intervals and refine the mesh at the point (locally) of a phase interface in each interval to improve computational accuracy. At the beginning, there are about 220,000 finite elements, and the drop mesh changes 12 times during ejection. During the second mesh refinement, the number of finite elements reaches 800,000.

The average quality of the elements diminishes with the increase in the number of finite elements.

#### 2.1.5. Droplet final mass calculation

Several important characteristics, such as ink mass, droplet sizes and/or shape of the ink droplet, were obtained, and the contribution of different factors, such as inks, ejection parameters, nozzle shape sizes, and surface properties, were analysed. The final mass of the droplet could vary depending on different dwelling periods and fluid intake velocity. The droplet mass turnover is calculated using the level–set interface by the following equation:

$$m_{\rm d} = \rho(V\phi). \tag{2.12}$$

#### 2.1.6. Dimensionless numbers in inkjet printing parameters

Several dimensionless parameters are crucial to the stable DOD inkjet working process. The Reynolds number (Re) determines the proportion of an ink's inertia to its viscosity, while the Weber number (We) identifies the proportion of an ink's inertia to its surface tension (Seipel et al., 2020). These adjustable numbers characterise the initial spreading dynamics after the impact. These dimensionless numbers are calculated using the following equations:

$$Re = \frac{\rho du}{\mu}; \tag{2.16}$$

$$We = \frac{\rho du^2}{\sigma},\tag{2.17}$$

here, d is the length of the inkjet printhead nozzle diameter and u is the droplet movement velocity.

Several effects have been observed during the motion of the droplet. The fragment of a vertical droplet falls with the mesh, and a contour plot of the velocity magnitude and Cell Reynold's number is illustrated in Fig. 2.2.



Fig. 2.2. Vertical droplet falls with a mesh and distribution of velocities (Tofan, 2022)

Droplet velocity effects during inkjet printing can be eliminated by generating (forming) the Ohnesorge number (Oh). A solution based on the Navier–Stokes equations expresses droplet ejection constraints/limitations in terms of fluid interfaces, viscosity, and inertial properties (Seipel et al., 2020). It is independent of the waveform extraction impulse that controls the speed (Wang and Hasegawa, 2023). The Oh greatly influences the behaviour of the expanding droplet from the print head nozzle. The Oh value can be defined from the physical properties of the ink and the nozzle size scale or from the Re and We numbers.

$$Oh = \frac{\mu}{\sqrt{\sigma \rho d}} = \frac{\sqrt{We}}{Re}.$$
(2.18)

Ink printability specification can be characterised by the dimensionless (Z) number:

$$Z = \frac{1}{oh}.$$
 (2.19)

The initial specification of the printing ink is necessary for stable droplet ejection and should be within the acceptable range of 1 < Z < 10 (Fromm, 1984; Derby and Reis, 2003). If the number Z > 10, satellite droplets will form during printing, reducing the quality of the print. If the number Z < 1, then the viscous forces of the liquid will not allow the droplet to expand.

A proper printing process in a DOD inkjet printhead requires the right combination of physical parameters, which will also depend on the droplet size and velocities (through the value of Reynolds or Weber number). Some of the results include empirical data that are incorporated into numerical simulation calculations considering the dimensionless numbers *Oh*, *We*, *Z*, and *Re*.

### 2.1.7. Contact angle calculated from the stability of forces according to Young's equation

The terms surface free energy (SFE) and surface free tension (SFT) are physically corresponding. SFE is usually used to describe energy for solid surfaces, and SFT is used for liquid surfaces. SFT is accordingly defined as the work required to isothermally and reversibly increase the surface area by unit size/amount (Ebnesajjad, 2014; Zhang et al., 2022). Irregularly, however, the SFT of a solid is also referred to. The SFE has the unit  $mJ/m^2$  (millijoule per square meter) as the energy per area, whereby the equivalent unit mN/m (millinewton per meter), which is commonly used for the SFT (Ebnesajjad, 2014; Tiyyagura et al., 2021). Surface free energy  $\sigma$  respectively represents the tensions between the solid–air, liquid–air and solid–liquid substrates. The relation of these components is calculated by Young's equation:

$$\sigma_s = \sigma_{sl} + \sigma_l \cdot \cos\theta, \qquad (2.20)$$
here,  $\sigma_s$  is the surface free energy of the solid,  $\sigma_l$  is the surface free energy of the liquid,  $\sigma_{sl}$  is the interfacial free energy between the solid and the liquid, and  $\theta$  is the solid and liquid contact angle. The following illustration (Fig. 2.3) shows droplet height *h*, droplet diameter *d*, and contact angle calculated from the stability of forces according to Young's equation at the stable phase:



Fig. 2.3. Droplet on a substrate indicates the surface tension and contact angle of three media (Kinloch, 1980)

In this system, the strength of adhesion may be calculated through the work of adhesion ( $W_a$ ). This was defined by the Dupre definition, which is calculated as:

$$W_a = \sigma_s + \sigma_l - \sigma_{sl.} \tag{2.21}$$

The droplet's interaction with the surface was modelled with COMSOL Multiphysics software. The height and base of the droplet were taken from programming results, and the contact angle was calculated as (Bonadiman et al., 2008):

$$\theta = 2 \tan^{-1} \left(\frac{2h}{d}\right),\tag{2.22}$$

where *h* is the height of the droplet, *d* is the diameter of the droplet, and  $\theta$  is the contact angle.

# 2.2. Characterisation of the colour reproduction measurement

To determine the difference in colour reproduction by CIE  $L^*a^*b^*$  values,  $\Delta E$  was taken as a reference. The colour difference is calculated according to the following equation:

$$\Delta E = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2}, \qquad (2.23)$$

where  $L^*$  is the lightness value (0 for black and 100 for white);  $a^*$  – the position between the red and green value (negative–green, positive–red);  $b^*$  – the position between the blue and yellow value (Sidaravičius, 2012). CIE  $L^*a^*b^*$  colour system is represented in Fig.2.4.



Fig. 2.4. CIE *L***a***b** colour system (Sidaravičius, 2012)

### 2.2.1. Differences in $\Delta L^*$ , $\Delta a^*$ and $\Delta b^*$ values

Differences in colour characteristics  $\Delta L^*$ ,  $\Delta a^*$ ,  $\Delta b^*$  between two samples are calculated using the following equation:

$$\Delta L^* = \sqrt{(L_1^* - L_2^*)^2}; \qquad (2.24)$$

$$\Delta a^* = \sqrt{(a_1^* - a_2^*)^2}; \tag{2.25}$$

$$\Delta b^* = \sqrt{(b_1^* - b_2^*)^2},\tag{2.26}$$

where, depending on the comparison,  $L_1^*$ ,  $a_1^*$ ,  $b_1^*$  represents the primary measured colour values and  $L_2^*$ ,  $a_2^*$ ,  $b_2^*$  represents the second measure of colour values. These values must be found to calculate differences.

### 2.2.2. Mean value and standard deviation measurement

The mean value  $\bar{x}$  is calculated using the following equation:

$$\bar{x} = \frac{\sum x}{n}.$$
(2.27)

The standard deviation is s (or variance, which is the standard deviation squared,  $s^2$ ). This quantity is defined as that deviation value equal to the square root of the sum of the squares of all the deviations divided by the total number of such deviations n, or mathematically:

$$s = \sqrt{\frac{\sum_{n=1}^{\infty} x^2}{n}},\tag{2.28}$$

here, *s* represents the variance or deviation for a sample. From the definition of this quantity, its formula can be written for a finite sample of deviations rather than the infinite parent population as in the previous section:

$$s = \sqrt{\frac{\sum x^2}{n}},\tag{2.29}$$

where *s* replaces  $\sigma$  since we are dealing with a sample rather than a population. Here, *s* represents the variance or standard deviation for a sample: *x* – the deviation of an independent variable from the correct value; *n* – the total number of deviations.

When the correct or best value must be found, statistical theory shows that a more exact value of the standard deviation results from:

$$s' = \sqrt{\frac{\sum x^2}{n-1}}.$$
 (2.30)

The (n - 1) is used because the best estimate found by averaging x will certainly deviate from the correct value by some amount unless a complete population is used instead of a selection. Using (n - 1) instead of n partially corrects for this situation because the true value is not independently known.

The standard mean value error is calculated as (Crowle, 2017):

$$SE_{\bar{x}} = \frac{s}{\sqrt{n}}.$$
 (2.32)

The colour difference is described by the  $\Delta E$  value. This value is characterised by the numbers: 0–1 is the imperceptible difference; 1–2 has a very small difference, which could be evident only to a trained eye; 2–3.5 has a medium difference and could be evident to an untrained eye; 3.6–5 is a visible difference and coefficient larger than 5 is very noticeable.

# 2.3. Characterisation of the surface of polymers evidence of the inks and surface adhesion measurement

The DIN ISO 2409 test was performed by cross-cutting and taping the printed surface directly after printing and in 24 hours to assess the adhesion between printed inks and the surface. Cross-cut test classification is shown in Table 2.1.

Classification Value	GT0	GT1	GT2	GT3	GT4
Cross-cut result					
Percent of area removed	_	<5%	5-15%	15-35%	35-65%

**Table 2.1.** DIN ISO 2409 test classification (Tofan, 2021)

To realise the differences in DIN ISO 2409 test classification, printed and tested PET samples with values GT2 and GT4 are illustrated in Fig. 2.5.



Fig. 2.5. Printed layer tested on PET material. (a) Test classification value (GT2); (b) test classification value (GT4); (c) cross-cut mesh dimensions (made by the author)

The purpose of the test is to quickly test the adhesion of the coating by applying simple methods. It is recommended to test samples directly after printing and within 24 hours after printing. The DIN ISO 2409 test shows the sample that can obtain strong adhesion with a classification value GT0, and the opposite value GT4 shows poor adhesion of inks to the surface.

The free energy change of the substrate during the physical experiment was measured using Plasmatreat test inks (A series) called PLASMATREAT "Surface Test Inks C28 to C72". This ink is suitable for the SE determination of plasmatreated and untreated (non-treated) polypropylene (PP), high-density polyethylene (HDPE) and polyethylene terephthalate (PET) surfaces. To determine the surface energy of a particular surface, the experiment started with the lowest ST C28 ink and progressed to the higher C72 test ink, which was quickly applied to the surface using a built-in (integrated) bottle brush. If the brush stroke edges remained stable for 2 seconds, the surface was easily wetted. The surface energy of the substrate is then equal to the value of the test ink. Thus, in this work, the surface energy values of the substrates are gradually approached. This test ink was used to analyse/determine changes in surface energy. Experiments were performed at room temperature of 23 °C and 55% humidity.

# 2.4. Conclusions of the Second Chapter

The presented methodology of theoretical, numerical, and experimental investigations in this chapter includes the following findings:

- The COSMOL numerical model using the finite element method can be used to predict the physical effects found/detected in the printing process, such as the maximum velocity of the droplet at the moment of ejection, when the pressure inside the nozzle is the highest and when the thread of the droplet unhooks/detaches the nozzle of the inkjet, and the droplet velocity exchange/turnover during falling and colliding/impact with the surface.
- 2. The colour reproduction measuring research contains the formation of the required experimental facilities and the proper collection and processing of CIE  $L^*a^*b^*$  value data using mathematical-statistical methods.
- 3. Following the test procedure DIN ISO 2409, the ink layers' resistance to separation/detachment from the substrate when cut at a right-angle lattice pattern must be evaluated. This procedure does not directly measure the adhesion force but helps to empirically determine the adhesion of the inkjet-printed layer to the surface.

3

# Investigation Results and Conclusions of the Inkjet Droplet Formation and Movement Modelling

This chapter presents an experimental and numerical investigation of appended articles 1–4. The aim is to link the conclusions of the articles with the results of the final dissertation objectives. It is impossible to improve the printing quality and speed without analysing the formation of an inkjet droplet and movement modelling. Article 1 presents the numerical modelling aspects and inkjet printhead process modelled with different impact of ink parameters. Article 2 describes the physical investigations of the inkjet printing colour reproduction by different printing properties. Article 3 elaborates on the research of inkjet droplet impact with the different characteristics of the surface. Article 4 presents the inkjet printing process using different printhead ejection parameters. The research results presented in this chapter were published in the author's publications Tofan, et al., 2021; Tofan, et al., 2023.

# 3.1. Investigation results of Article 1

When performing inkjet printing, it is critical to observe the droplet form turnover, its velocity prior to the interaction with the surface and the final shape in the stable

phase on the substrate. Technically, all these characteristics have been achieved and realised in the numerical model of inkjet nozzle application. The movement of the droplet was modelled using Navier–Stokes equations. The droplet ejection, motion, and impact were investigated using the COMSOL CFD module. The presented model of the droplet ejection from the nozzle can calculate process parameters or even early stages of future issues of the ink or inkjet printhead developing process. The numerical model was used to evaluate every important step of the inkjet printing application.

Article 1 provides the occurrence comparison of the different inks with different viscosity and density values. The droplet velocity and length parameters of different inks are represented at different time steps during the printing process. The main phases of the printing process (Fig. 1.4) of droplet formation and movement to the surface are illustrated in Table 3.1.

Inkı			Ink ₂			Ink3						
Phases	t µs	u m/s	D _{d.max} μm	D _h μm	t μs	u m/s	D _{d.max} µm	τ D _h μm	t µs	u m/s	D _{d.max} μm	D _h μm
А	2	13.8	26.3	9.2	2	13.8	25.8	9.7	2	13.8	23.5	9.9
В	11	8.51	20.4	110.9	13	7.88	20.2	115.3	14	7.28	20.0	118.7
С	26	7.05	23.3	26.2	32	6.77	24.9	26.3	39	5.93	24.0	26.6
D	143	4.48	22.2	21.7	150	4.28	22.2	21.8	169	3.60	21.9	22.0
Е	149	0.18	47.7	4.3	161	0.12	42.8	7.2	183	0.05	37.5	11.6
F	174	0	36.0	11.6	172	0	36.1	12.1	181	0	36.3	11.9

**Table 3.1.** Characteristics of the different inks at different steps in time (Tofan, 2021)

After the droplet is generated, it is important to observe all these steps during the period while the droplet stagnates in the final state on the surface. The comparison of the printing processes of all three ink types revealed that  $Ink_2$  shows the most suitable result. A droplet from  $Ink_2$  has the smallest oscillation effect and has the lowest period to reach a stable phase on the surface. Droplets from Ink₁ reach the surface faster but, in the case of the lowest viscosity and density parameters, oscillate more strongly before they take their final form on the surface. For UV-curable inks, it is vital to undergo this step in the stable phase of the droplet.

The associated graphs illustrated the characteristics of turnover of the droplet during the formation and movement to the substrate, whereby no wettability is considered. The velocity turnover is an important factor for the control of print quality, as, for instance, the droplet velocity turnover plays a direct role in the distribution of the ink on the surface. Fig. 3.1 demonstrates the droplets from different inks and velocities during the ejection, fall and impact processes.



Fig. 3.1. Representation of the different inks formed droplet velocities turnover (Tofan, 2021)

The droplet reaches the highest velocity values at phase A when the pressure wave inside the nozzle reaches the highest level. After, the droplet velocity decreases while the droplet is stretching and finally pitches off from the nozzle at phase B. The velocity still decreases during the droplet transformation to the sphere form at phase C. During the droplet fall, the velocity of the droplet decreases continuously while the droplet reaches the surface at phase D, and the interaction with the substrate begins. Phase D illustrates the droplet's spread on the surface. The droplet diameter is the biggest. Phase F illustrates the droplet at the final stable stage when the velocity equals zero. Also, differences in the droplet forming parameters can be addressed depending on time steps. Fig. 3.2 illustrates the turnover of parameters  $D_{d.max}$  (the maximum droplet diameter by the horizontal cross-section calculated in Table 3.1) and  $D_h$  (a droplet height by vertical cross-section calculated in Table 3.1).



Fig. 3.2. Representation of size parameters' turnover of a droplet formed from different inks (Tofan, 2021)

The results show that different behaviours of droplets and the duration of interaction can be expected depending on inks. The comparison of different ink behaviours showed that inks with the lower viscosity and density have the biggest oscillation on the surface and the smallest jet size. The droplet's maximum spreading diameter on the surface is at phase  $E_1 = 47.7 \mu m$ . The sphere diameter of different droplets during the flight equals 22  $\mu m$ . After the impact with the substrate, the droplet loses its spherical shape and takes a different form on the surface. The minimum spreading diameter is particular to the droplet of Ink₃, with the highest density and viscosity parameters. This ink droplet does not oscillate at all and takes the final form immediately after the impact with the smallest oscillation parameters.

Even though numerical studies of a moving droplet have been carried out, the question remains not only about the dynamic behaviour but also about the colour characteristics. Numerical experiments showed that further physical studies are needed to analyse the effect of inkjet colour reproduction on various surfaces using various printing parameters applied.

### 3.2. Investigation results of Article 2

Experiments with digital inkjet printing on different linen textile surfaces were performed to find the best printing parameters of colour reproduction on the surfaces. To obtain more accurate results, it was considered that reproduced colours should be compared with digital colour values under daylight illuminance. Also, the inks' physical properties directly affect the printing quality.

Shades of linen fabrics were chosen in different colours: bleached, melange and natural linen. The surface colour and structure have a significant effect on the reproduction of the colours by printing with different colours of paint.

Printed colour  $L^*a^*b^*$  parameters were compared with reference digital colour parameters.

The whitened linen fabrics (OBR 840, OBR 0114, OBR 491, OBR 1542) have similar colour reproduction characteristics as white paper, and the natural linen fabrics (OBR 166, OBR 051, OBR 1041) have very restricted colour reproduction limits. While printing on whitened linen fabrics, the colours are the best reproduced on the thickest bleached linen fabric (OBR 1542) and the worst on the thinnest natural linen fabric (OBR 1041). The background colour of the material strongly affects the reproduction of inkjet colours. Colour reproduction on linen fabric by printing in four passes and two ink layers is illustrated in Fig. 3.3.



**Fig. 3.3.** Colour reproduction on linen fabric in two-dimensional space a*b* printing in four passes and coating in two ink layers (Tofan, 2021)

Parameters were measured over a period of 90 days for the evaluation of ink stability and suitability for digital inkjet printing. This was considered for the evaluation of ink stability and suitability.

The a*b* values of digital reference colours (black line in Fig 3.3) show that it is possible to reach the limits of maximum intense colours, and the range of colours was reproduced in the print on paper (white line in Fig 3.3). The following

are six colours for reproduction: Y - yellow, R - red, M - magenta, B - blue, C - cyan, and G - green. Limited colour impregnation reproduction on all types of linen fabrics is visible. The most affected are C and B colour saturations. Colour reproduction in prints on bleached linen fabrics is the closest to colour reproduction on paper, while there are very limited reproduction limits of colour saturation in printing on natural linen fabrics.

The research focus was on the investigation of different printer parameters (the number of printing layers 1–5 and printing passes 4, 8, 16) to affect colour reproduction on different natural linen textile surfaces. Colour  $L^*a^*b^*$  value reproduction by printing at different passes is very corresponding. Printing by different pass numbers does not affect colour reproduction parameters, but printing with a smaller pass number significantly increases printing speed. The colour difference  $\Delta E$  between the different passes printing is not greater than three.

Whitened linen fabric (OBR 840) was selected to better identify changes in colour characteristics depending on different ink layers and the printing pass number. The number of applied ink layers has the most influence on the change in lightness (*L**) parameters. There is a relevant difference between printed layers from 1 to 5 ( $\Delta E$  from 3% to 55%), while there is an insignificant difference between layers 3 and 4 ( $\Delta E \leq 5\%$ ) and between layers 4 and 5 ( $\Delta E \leq 4\%$ ). Printing with one layer of ink produces the worst colour reproduction, and the best colour reproduction is achieved with two ink layers. Using 4 or 5 ink layers increases colour characteristic  $\Delta E$ , and colour reproduction is less effective than using 2–3 ink layers.

Comparing the 4 and 16 pass printing settings, the same patterns of  $\Delta E$  colour characteristic changes remain. Comparing the difference in colour reproduction from the number of passes, the colour variation is inconsequential ( $\Delta E \leq 3$ ), except for the green colour.

After physical experimentation with different print settings and their effect on colour rendering, it was noted that further research is needed to analyse the effect of droplet impact from inkjet printers with various surface energy. This is necessary to find out the tendency of the droplet to stick to the surface. Further numerical studies should include droplet formation, movement velocity, and impact on various treated and untreated polymeric surfaces to analyse how quickly droplets reach a stable phase on the surface, droplet covering plot, and droplet wetting angles.

After conducting preliminary studies related to the movement of the droplet and the reproduction of colour characteristics by various printing parameters, further studies of the interaction of droplets are needed. Previous studies, given the main line of research, i.e., ensuring the quality of printing, require additional research indicating not only the cases mentioned but also cases of the droplet impact on the surface. This study will complement two previous publications and answer the question of the influence of surface energy on the spreading of a droplet over the surface. This will directly affect the improvement in print quality.

# 3.3. Investigation results of Article 3

To better analyse droplet impact on the surface, it is important to obtain appropriate droplet deposition conditions. For example, by applying and adjusting the droplet impact, it is possible to maximise the print speed without losing print quality. The impact of droplets during the inkjet printing process application can be characterised by the sequence of five consistent phases: interaction, spreading, relaxation, wetting and equilibrium.

Table 3.2 provides the results of the DIN ISO 2409 test method for the resistance of paint coatings and varnishes to separation from substrates. The change in free energy of the surface was measured with Plasmatreat test inks. The contact angles of the droplet and surface were modelled by using the COMSOL Multiphysics software.

Test No.	Sample No.	SE (mN/m)	Contact Angle (°)	Cross-Cut Test Direct after Printing	Cross-Cut Test after 24 h	Samples Treat- ment Method
1	1	32	82.3	GT1	GT4	—
	2	28	83.8	GT3	GT4	—
	3	32	78.1	GT4	GT4	—
	4	38	79.7	GT3	GT4	—
2	1	32	_	GT1	GT4	Cleaned with IPA
	2	28	_	GT2	GT4	Cleaned with IPA
	3	32	_	GT2	GT4	Cleaned with IPA
	4	38	_	GT2	GT4	Cleaned with IPA
3	1	70	52.4	GT0	GT0	Plasma treated
	2	58	55.3	GT0	GT0	Plasma treated
	3	72	48.7	GT0	GT0	Plasma treated
	4	72	49.1	GT0	GT0	Plasma treated

**Table 3.2**. Results on the resistance of paint coatings and varnishes to separation from substrates (Tofan, 2021)

The objective of this test is the adhesion of the printed layer and surface of different materials using different treatment methods. The cross-cut test shows which material can achieve stable adhesion by classification values of GT0–GT4. The classification GT0 means stable adhesion with 0% of the ink layer removed, and GT4 is used when all printed layers are fully removed. All samples had different results directly after the printing, but the final measuring was applied after 24 hours. Excellent adhesion results were noticed when samples were treated with

open-air plasma. This pre-treatment process gives all samples a stable GT0 classification value.

34

By applying the numerical modelling, the droplet velocity turnover is investigated at the impact on non-treated and treated surfaces (Fig. 3.4).



Fig. 3.4. Droplet velocity turnover during the impact on different surfaces (Tofan, 2022)

The velocity changes during the contact of the droplet with different surfaces are represented by every  $\mu$ s. Before reaching a stable phase on the surface, several processes of oscillation damping pass, and the droplet action appertains to the properties of the surface. It is important to note that the behaviour of droplet oscillations at the impact also depends on the properties of the fluid, but this numerical experiment used the same inks with the same properties. Different surface parameters determine a different droplet movement velocity during impact. At the end of the impact, when the velocity extends to zero, the droplet takes on its final form on the surface. The results show that on the non-treated surface, the velocity of droplet oscillations is twice as much as on the treated surface. The treatment methods that increase the surface energy affect the inkjet droplet's final form and reduce the splashing effect of the impact.

Droplets on the surface have different wetting (contact) angles by the different surface parameters. The droplet coverage area on the surface can be twice bigger and cover four times more area if the treated polyethylene terephthalate surface is considered. After the impact of a droplet with the untreated polyethylene terephthalate surface, the droplet diameter during the oscillation grows to 48  $\mu$ m and afterwards decreases to 32  $\mu$ m at a stable shape. In the case of a treated surface, the droplet slowly expands and stays in its final shape with a diameter of 64  $\mu$ m. The reduction of the droplet's contact area on non-treated surfaces can be predicted by processing the surface energies and interfacial energy.

In the process of inkjet printing on different surfaces, it is important to know the surface energy of the polymer to better understand the adhesion properties. The atmospheric pressure plasma treatment method was investigated to reach the GT0 stable adhesion classification value while testing the adhesion between the printed layer and the surface by the DIN ISO 2409 test. The investigated numerical modelling with COMSOL was used to examine the inkjet droplet velocity and diameter during the impact with treated and non-treated polymer surfaces. The oscillation of the droplet at the impact phase depends on the properties of the inks, printhead jetting parameters and the characteristics of the surface energies.

Regarding demanding plastics regulations, it is very important to investigate the recycling processes for printing applications on different polymer surfaces to safely use them for post-consumer recycling. It has been observed that the adhesion between the ink layer and the polymer surface should be appropriate for the application of circular economy.

Numerical modelling is related to improving the structure of printheads, print quality, print velocity and the study of new inks for inkjet printers. In future studies, this research information may be extended to other materials, combined surface treatments, and mixed adhesives. For additional research, it is important to analyse the droplet ejection process deeply and what parameters in control of the inkjet printhead play a direct role in droplet formation.

### 3.4. Investigation results of Article 4

This Article aimed to investigate the droplet ejection parameter impact on droplet formation and movement parameter turnover over time. An understanding was gained of the droplet behaviour at different ejection parameters while explaining the influence of dwelling time on the droplet formation process. The droplet mass and speed turnover were also observed. It is important to investigate the trajectory of the jet and the mass of the droplet to understand the final position and area covered by the droplet on the surface. These aspects affect the final printing quality. The numerical modelling in the three-dimensional space evaluates the impact of such components as the waveform dwelling time and fluid inlet velocity. If the inlet pulse and fluid velocity at the nozzle are too low, the droplet shrinks back to the nozzle. The kinetic energy is not high enough to jet the droplet from the nozzle. If the impulse for jetting the droplet is too strong, the droplet forms many malignant satellites and impacts the surface by splashing hard and harming the printing quality by uncontrolled impact at the surface.

The droplet ejection progress was taken from numerical simulation using the COMSOL Multiphysics software. The droplet formation behaviour is predicted by the strength and period of fluid inlet velocities. For these simulations, the inlet velocity was set to 0.5 m/s with different dwelling times of 1  $\mu$ s, 3  $\mu$ s, and 12  $\mu$ s. The results of droplet formation and movement are represented in Fig. 3.5.



**Fig 3.5.** Representation of the droplet formation and movement by different ejection parameters of waveform dwelling time: (a) 1  $\mu$ s; (b) 3  $\mu$ s; (c) 12  $\mu$ s (Tofan, 2022)

The droplet mass at the ejection phase increases proportionally to the printhead inlet velocity. The droplet moves forward to the surface and pitches off from the nozzle at different time steps after the waveform dwelling period ends. The droplet with the formatting time of 3  $\mu$ s reaches the surface at the same time step as the droplet with the formatting time of 12  $\mu$ s, but an obvious difference in the impact with the surface is observed. The droplet with the waveform dwelling time of 3  $\mu$ s forms a perfect shape on the surface, while the droplet with 12  $\mu$ s splashes a lot. This result shows that depending on the printed image's resolution, it is possible to use all three different dwelling times to increase printing speed and quality. If it is necessary to print in high-quality resolution, it is better to use the small dwelling period to reach small dots without any harmful satellites; but if high quality is not required and the need is to cover a thick layer of the same ink, the big dwelling period can be used.

The droplet mass formatting was investigated by different dwelling periods. The mass turnover of droplets is shown in Fig 3.6.



Fig. 3.6. Representation of the droplet mass turnover with different ejection parameters. Mass versus formatting time (Tofan, 2022)

The different dwelling times specify a direct effect on the droplet mass formation. The large formatting time of 12  $\mu$ s impacts the mass reduced at 30  $\mu$ s by droplet break up and increases after 35  $\mu$ s by jet tail connecting the main droplet. The mass of the different dwelling periods generated droplets before the impact are 1  $\mu$ s (2.5·10⁻³  $\mu$ g), 3  $\mu$ s (16·10⁻³  $\mu$ g), 12  $\mu$ s (65·10⁻³  $\mu$ g). The presented numerical simulation is an important alternative to the quality improvement analysis of different printhead settings.

### 3.5. Conclusions of the Third Chapter

- 1. The known effects of printing inks in the physical process of DOD inkjet printing were discovered and numerically investigated. The changed physical properties ( $\rho$ ,  $\mu$ ) of the ink directly affects the droplet formation time, falling velocity turnover, shape turnover, and the droplet surface oscillation after the impact.
- 2. It is observed that inks with lower viscosity and density have the largest surface oscillation and the smallest jet size. The minimum spreading diameter is characteristic for a droplet with the highest density and viscosity parameters. This ink droplet does not oscillate at all and takes its final shape immediately after impact with the smallest oscillation parameters.
- 3. The effect of different printing parameters on the reproduction of colour characteristics on surfaces of different characteristics was physically determined by evaluating the spectral properties of the print on the CIE  $L^*a^*b^*$  scale. Printing with different pass numbers does not affect colour reproduction parameters, but printing with a smaller pass number significantly increases printing speed. The difference in colour  $\Delta E$  between different printing passes does not exceed three. The greatest influence on change in lightness ( $L^*$ ) parameters is exerted by the number of applied layers of ink.
- 4. The interaction of an inkjet droplet with surfaces of different surface energy was simulated numerically by recording the dipping angles and the movement speeds of the droplet. After a droplet impact an untreated polyethylene terephthalate surface, the diameter of the droplet increases to  $48 \mu m$  as the oscillation increase, and then decreases to  $32 \mu m$  as the shape stabilizes. In the case of a treated surface, the droplet slowly expands and maintains its final shape of  $64 \mu m$  in diameter. Oscillations in droplet velocity are more significant on surfaces with lower surface energy.
- 5. The presented numerical simulation model is an important alternative to analyse the quality improvement of different printhead settings. A notice-

able difference in the formation of a droplet and its impact with the surface is observed at different dwelling periods. If high-quality resolution is required, it is recommended to use a short dwelling period to generate small droplets without any harmful satellites; but if high quality is not required and it is necessary to cover a thick layer of the same ink as quickly as possible, a longer dwelling period can be used.

# **General Conclusions**

- 1. As a result of a numerical experiment, it was found that the following parameters affect the process of inkjet printing: how fast the droplet forms a sphere during step C (Fig. 1.4) and what speed it will have before interacting with the surface during step D. It is also important to predict the results of the interaction of a droplet with a surface at steps D–F.
- 2. The impact of various inkjet inks on a flat surface was investigated. The results show that depending on the ink ( $\mu$ ;  $\rho$ ;  $\sigma$ ; *Oh*; *Z*), different behaviours of the droplets and the duration of the interaction can be expected. Changes in the presented parameters are important for controlling the printing process and for checking the print quality.
- 3. The turnover of droplet ejection in time has been studied. The main goal of understanding the behaviour of the droplet at various waveform ejection parameters has been achieved. The turnover of the droplet mass is also observed. It is important to investigate the droplet trajectory and droplet mass to understand the final location or place and diameter of the droplet on the surface.
- 4. The graphical results demonstrate that the change in droplet shape over time can be better explained by considering different time intervals. It is proposed to divide the droplet motion into six stages: droplet ejection, separation of the droplet thread from the nozzle, droplet sphere formation,

droplet before interaction, droplet interaction (spreading) over the substrate surface, and droplet stability phase. The proposed six-stage study allows for faster detection of changes in the shape of the ejected droplet during the corresponding time interval when it freely falls towards the interacting surface.

- 5. Droplet ejection, motion, and impact were analysed using the COMSOL CFD simulation software module, and the inkjet model was improved. The numerical experiments showed that the presented study is important for the analysis and improvement of behaviours of different inks, different ejection parameters and different nozzle parameters.
- 6. A numerical model was created for studying the behaviour and parameters of the motion of an inkjet droplet. Using the COMSOL software, the length of the droplet thread and the moment of separation of the droplet from the thread were found, as well as the spherical shape, droplet mass, droplet volume, droplet condition on impact, and final droplet state. In addition, it became possible to adjust different nozzle parameters and different ejection pressures and test different inks.
- 7. The use of 3D CFD modelling makes it possible to estimate such components as the ejection time of the ejection of the drop, the mass and diameter of the drop, the fall velocity of the droplet, and the parameters of its impact. In this work, the direct influence of various performance parameters was found on the impact of a droplet on the surface, the impact on the surface of a different dwelling period, i.e., 1  $\mu$ s (2.5 ng), 3  $\mu$ s (16 ng), and 12  $\mu$ s (65 ng).
- 8. The numerical experiment showed that the ejection parameters can be used to increase the printing speed and print quality, considering the resolution of the printed image. In cases where a print of the highest quality is required, it is recommended to set a low dwelling time to form small droplets of ink without harmful satellites. If a high-resolution print is not required and a thick layer of the same ink needs to be applied, a longer dwelling time can be set.

# References

Antonyuk, S., Khanal, M., Tomas, J., Heinrich, S., & Mörl, L. (2006). Impact breakage of spherical granules: experimental study and DEM simulation. *Chemical Engineering and Processing: Process Intensification*, *45*(10), 838–856. https://doi.org/10.1016/j.cep.2005.12.005

Aqeel, A. B., Mohasan, M., Lv, P., Yang, Y., & Duan, H. (2019). Effects of nozzle and fluid properties on the drop formation dynamics in a drop-on-demand inkjet printing. *Applied Mathematics and Mechanics*, 40, 1239–1254. https://doi.org/10.1007/s10483-019-2514-7

Bonadiman, R., Marques, M., Freitas, G., & Reinikainen, T. (2008). Evaluation of printing parameters and substrate treatment over the quality of printed silver traces. In *2008 2nd Electronics System-Integration Technology Conference* (pp. 1343–1348). IEEE. https://doi.org/10.1109/ESTC.2008.4684550

Bussmann, M., Chandra, S., & Mostaghimi, J. (2000). Modeling the splash of a droplet impacting a solid surface. *Physics of fluids*, 12, 3121–3132. https://doi.org/10.1063/1.1321258.

Castrejon-Pita, J. R., Baxter, W. R. S., Morgan, J., Temple, S., Martin, G. D., & Hutchings, I. M. (2013). Future, opportunities and challenges of inkjet technologies. *Atomization and sprays*, 23, 541–565. https://doi.org/10.1615/AtomizSpr.2013007653

Chen, F., Zhang, Y., Nakagawa, Y., Zeng, H., Luo, C., Nakajima, H., Uchiyama, K., & Lin, J. M. (2013). A piezoelectric drop-on-demand generator for accurate samples in capillary electrophoresis. *Talanta*, 107, 111–117. https://doi.org/10.1016/j.talanta.2012.12.058

Creton, C., & Ciccotti, M. (2016). Fracture and adhesion of soft materials: a review. *Reports on Progress in Physics*, 79(4), 046601. https://doi.org/10.1088/0034-4885/79/4/046601

Crowle, G. (2017). Effects of Cotton Structure and Surface Interactions on Dye Uptake. Ph. D. Thesis, Deakin University.

Cundall, P. A., & Strack, O. D. L. (1979). A discrete numerical model for granular assemblies. *Geotechnique* 29 (pp. 47–65). https://doi.org/10.1680/geot.1979.29.1.47

Das, A., Princen, M., Roudbari, M. S., Khalate, A., & Weiland, S. (2021). Soft Sensing-Based In Situ Control of Thermofluidic Processes in DoD Inkjet Printing. *IEEE Transactions on Control Systems Technology*, 30(3), 956–971. https://doi.org/10.1109/TCST.2021.3087576

Das, R., Mahapatra, A., Ball, A. K., Roy, S. S., & Murmu, N. C. (2017). Study of thermofluidic behavior of micro-droplet in inkjet-based micro manufacturing processes. In *AIP Conference Proceedings*, 1851(1), p. 020069. AIP Publishing LLC. https://doi.org/10.1063/1.4984698

Derby, B. (2011). Inkjet printing ceramics: From drops to solid. *Journal of the European Ceramic Society*, *31*(14), 2543–2550. https://doi.org/10.1016/j.jeurceramsoc.2011.01.016

Derby, B., & Reis, N. (2003). Inkjet printing of highly loaded particulate suspensions. *MRS bulletin*, 28(11), 815–818. https://doi.org/10.1557/mrs2003.230

Dowding, I., Hassani, M., Sun, Y., Veysset, D., Nelson, K. A., & Schuh, C. A. (2020). Particle size effects in metallic microparticle impact-bonding. *Acta Materialia*, 194, 40–48. https://doi.org/10.1016/j.actamat.2020.04.044

Driessen, T. (2013). Drop formation from axi-symmetric fluid jets. Ph. D thesis, University of Twente.

Dupré, A., & Dupré, P. (1869). Théorie mécanique de la chaleur. Gauthier-Villars. http://books.google.com/books?id=qD4JAAAAIAAJ&oe=UTF-8 (accessed on 11 November 2021).

Ebnesajjad, S. (2011). Material surface preparation techniques. In *Handbook of Adhesives and Surface Preparation* (pp. 49–81). William Andrew Publishing. https://doi.org/10.1016/B978-1-4377-4461-3.10004-5

El-Emam, M. A., Zhou, L., Shi, W., Han, C., Bai, L., & Agarwal, R. (2021). Theories and applications of CFD–DEM coupling approach for granular flow: A review. *Archives of Computational Methods in Engineering*, 1–42. https://doi.org/10.1007/s11831-021-09568-9

Fischer, A. C., Mäntysalo, M., & Niklaus, F. (2020). Inkjet printing, laser-based micromachining, and micro–3D printing technologies for MEMS. In *Handbook of silicon based MEMS materials and technologies* (pp. 531–545). Elsevier. https://doi.org/10.1016/B978-0-12-817786-0.00021-9

Fromm, J. E. (1984). Numerical calculation of the fluid dynamics of drop-on-demand jets. *IBM Journal of Research and Development*, 28(3), 322–333. https://doi.org/10.1147/rd.283.0322

Garduño, S. I., Sacramento-Orduño, A., Ramírez-Como, M., Reyes-Valderrama, M. I., Rodríguez-Lugo, V., & Estrada, M. (2023). Development of Drop-On-Demand Inkjet Process for the Fabrication of Thin-Film Printed Devices. In *2023 IEEE Latin American Electron Devices Conference* (LAEDC) (pp. 1–5). IEEE. https://doi.org/10.1109/LAEDC58183.2023.10208285

Giusti, D., Costantini, S., Brovelli, S., & Ferrera, M. (2022). Inkjet Printhead. In *Silicon Sensors and Actuators: The Feynman Roadmap* (pp. 603–649). Cham: Springer International Publishing. https://doi.org/10.1007/978-3-030-80135-9_19

Hoath, S. D. (Ed.). (2016). Fundamentals of inkjet printing: the science of inkjet and droplets. John Wiley & Sons.

Huang, J., Segura, L. J., Wang, T., Zhao, G., Sun, H., & Zhou, C. (2020). Unsupervised learning for the droplet evolution prediction and process dynamics understanding in inkjet printing. *Additive Manufacturing*, 35, 101197. https://doi.org/10.1016/j.addma.2020.101197

Huang, Z., Tang, Y., Guo, H., Feng, X., Zhang, T., Li, P., & Xie, Y. (2020). 3D printing of ceramics and graphene circuits-on-ceramics by thermal bubble inkjet technology and high temperature sintering. *Ceramics International*, 46(8), 10096–10104. https://doi.org/10.1016/j.ceramint.2019.12.278

Iijima, S. (1985). Electron microscopy of small particles. *Microscopy*, *34*(4), 249–265. https://doi.org/10.1093/oxfordjournals.jmicro.a050518

Jasevičius, R., Baronas, R., & Kruggel-Emden, H. (2015). Numerical modelling of the normal adhesive elastic–plastic interaction of a bacterium. *Advanced Powder Technology*, 26(3), 742–752. https://doi.org/10.1016/j.apt.2015.04.010

Jiang, H., & Tan, H. (2018). One dimensional model for droplet ejection process in inkjet devices. *Fluids*, *3*(2), 28; https://doi.org/10.3390/fluids3020028

Jiang, L., Yu, L., Premaratne, P., Zhang, Z., & Qin, H. (2021). CFD-based numerical modeling to predict the dimensions of printed droplets in electrohydrodynamic inkjet printing. *Journal of Manufacturing Processes*, 66, 125–132. https://doi.org/10.1016/j.jmapro.2021.04.003

Kamis, Y. E., Eral, H. B., & Breugem, W. P. (2021). Active control of jet breakup and droplet formation using temperature modulation. *Physical Review Fluids*, *6*(10), 103903. https://doi.org/10.1103/PhysRevFluids.6.103903 Kang, S. H., Kim, S., Lim, J. W., Sohn, D. K., & Ko, H. S. (2020). Study on fall velocity of continuously ejected micro inkjet droplet. *Journal of Mechanical Science and Technology*, 34, 3311–3315. https://doi.org/10.1007/s12206-020-0723-1

Kang, S. H., Kim, S., Sohn, D. K., & Ko, H. S. (2020). Analysis of drop-on-demand piezo inkjet performance. *Physics of Fluids*, *32*(2). https://doi.org/10.1063/1.5142023

Khan, A., Rahman, K., Ali, S., Khan, S., Wang, B., & Bermak, A. (2021). Fabrication of circuits by multi-nozzle electrohydrodynamic inkjet printing for soft wearable electronics. *Journal of Materials Research*, 1–11. https://doi.org/10.1557/s43578-021-00188-4

Kim, E., & Lee, H. (2022). Mechanical characterization of soft microparticles prepared by droplet microfluidics. *Journal of Polymer Science*, 60(11), 1670–1699. https://doi.org/10.1002/pol.20220110

Kinloch, A. J. (1980). The science of adhesion: part 1 surface and interfacial aspects. *Journal of materials science*, 15, 2141–2166. https://doi.org/10.1007/BF00552302

Korvink, G., Smith, P. J., & Shin, D.-Y. (Eds.) (2012). Inkjet-based micromanufacturing. Wiley-VCH Verlag GmbH & Co. KGaA. https://doi.org/10.1002/9783527647101

Kuang, D. M., Long, Z. L., Ogwu, I., & Chen, Z. (2022). A discrete element method (DEM)-based approach to simulating particle breakage. *Acta Geotechnica*, *17*(7), 2751–2764. https://doi.org/10.1007/s11440-021-01406-3

Lee, E. R. (2018). Microdrop generation. CRC press.

Lei, T., Han, J., & Liu, H. (2022). Numerical analysis and optimal CFD model verification of piezoelectric inkjet printhead. *Journal of Applied Fluid Mechanics*, *15*(4), 1087–1097. https://doi.org/10.47176/JAFM.15.04.33396

Leontev, A., Rozental, L., & Freger, V. (2023). Dynamics of underwater microparticle adhesion to soft hydrated surfaces: Modeling and analysis by time-dependent AFM force spectroscopy. *Journal of Colloid and Interface Science*, 651, 464–476. https://doi.org/10.1016/j.jcis.2023.07.185

Lesage, A. C., Allain, O., & Dervieux, A. (2007). On level set modelling of bi-fluid capillary flow. *International journal for numerical methods in fluids*, 53(8), 1297–1314. https://doi.org/10.1002/fld.1449

Li, D., Zhang, H., Zhang, L., Wang, P., Xu, H., & Xuan, J. (2019). Rapid synthesis of porous graphene microspheres through a three-dimensionally printed inkjet nozzle for selective pollutant removal from water. *ACS omega*, *4*(24), 20509–20518. https://doi.org/10.1021/acsomega.9b02249

Li, G., Liu, T., Xiao, X., Gu, M., & Liao, W. (2022). Numerical simulations of droplet forming, breaking and depositing behaviors in high-viscosity paste jetting. *Journal of Manufacturing Processes*, 78, 172–182. https://doi.org/10.1016/j.jmapro.2022.03.047

Liu, Y., & Derby, B. (2019). Experimental study of the parameters for stable drop-ondemand inkjet performance. *Physics of Fluids*, *31*(3). https://doi.org/10.1063/1.5085868 Lohse, D. (2022). Fundamental fluid dynamics challenges in inkjet printing. *Annual review of fluid mechanics*, 54, 349–382. https://doi.org/10.1146/annurev-fluid-022321-114001

Maisch, P., Tam, K. C., Jang, D., Steinberger, M., Yang, F., Brabec, C. J., & Egelhaaf, H. J. (2021). Inkjet printed organic and perovskite photovoltaics—review and perspectives. *Organic Flexible Electronics*, 305–333. https://doi.org/10.1016/B978-0-12-818890-3.00010-2

Makowski, Ł., Łaskowski, J., & Tyrański, M. (2021). Influence of modification of the geometry of the wave-plate mist eliminators on the droplet removal efficiency — CFD modelling. *Processes*, 9(9), 1499. https://doi.org/10.3390/pr9091499

Maramizonouz, S., Rahmati, M., Link, A., Franke, T., & Fu, Y. (2021). Numerical and experimental studies of acoustic streaming effects on microparticles/droplets in microchannel flow. *International Journal of Engineering Science*, 169, 103563. https://doi.org/10.1016/j.ijengsci.2021.103563

Mau, R., & Seitz, H. (2023). Influence of the Volatility of Solvent on the Reproducibility of Droplet Formation in Pharmaceutical Inkjet Printing. *Pharmaceutics*, *15*(2), 367. https://doi.org/10.3390/pharmaceutics15020367

Mbanjwa, M. B., Harding, K., & Gledhill, I. M. (2022). Numerical Modelling of Mixing in a Microfluidic Droplet Using a Two-Phase Moving Frame of Reference Approach. *Micromachines*, *13*(5), 708. https://doi.org/10.3390/mi13050708

Müller, C., Müller, A., & Pompe, T. (2013). Dissipative interactions in cell-matrix adhesion. *Soft Matter*, *9*(27), 6207–6216. https://doi.org/10.1039/C3SM50803J

Nguyen, V. T., Leong, J. Y. C., Watanabe, S., Morooka, T., & Shimizu, T. (2021). A multi-fidelity model for simulations and sensitivity analysis of piezoelectric inkjet printheads. *Micromachines*, *12*(9), 1038. https://doi.org/10.3390/mi12091038

Oktavianty, O., Haruyama, S., & Ishii, Y. (2022). Enhancing droplet quality of edible ink in single and multi-drop methods by optimization the waveform design of DoD inkjet printer. *Processes*, *10*(1), 91. https://doi.org/10.3390/pr10010091

Oktavianty, O., Kyotani, T., Haruyama, S., & Kaminishi, K. (2019). New actuation waveform design of DoD inkjet printer for single and multi-drop ejection method. *Additive Manufacturing*, 25, 522–531. https://doi.org/10.1016/j.addma.2018.12.008

Okuyama, K., & Yoshida, K. (2018). Dynamic behavior with rapid evaporation of an inkjet water droplet upon collision with a high-temperature solid above the limit of liquid superheat. *International Journal of Heat and Mass Transfer*, 116, 994–1002. https://doi.org/10.1016/j.ijheatmasstransfer.2017.09.093

Osher, S., & Tsai, R. (2003). Level set methods and their applications in image science. Communications in Mathematical Sciences, 1(4), 623–656. https://doi.org/10.4310/CMS.2003.v1.n4.a1 Paul, A., & Roy, S. S. (2022). Numerical simulation to predict printed width in EHD inkjet 3D printing process. *Materials Today: Proceedings*, 62, 373–379. https://doi.org/10.1016/j.matpr.2022.03.695

Peng, J., Huang, J., Wang, J., Meng, F., Gong, H., & Ping, B. (2022). The Driving Waveform Design Method of Power-Law Fluid Piezoelectric Printing Based on Iterative Learning Control. *Sensors*, 22(3), 935. https://doi.org/10.3390/s22030935

Peng, Z., Doroodchi, E., & Moghtaderi, B. (2020). Heat transfer modelling in Discrete Element Method (DEM)-based simulations of thermal processes: Theory and model development. *Progress in Energy and Combustion Science*, 79, 100847. https://doi.org/10.1016/j.pecs.2020.100847

Rojek, J., Nosewicz, S., & Thoeni, K. (2021). 3D formulation of the deformable discrete element method. *International Journal for Numerical Methods in Engineering*, *122*(14), 3335–3367. https://doi.org/10.1002/nme.6666

Rotter, S., Dosta, M., & Düster, A. (2023). Discrete element simulation of the breakage behavior of porous granules utilizing bond models. *Computational Particle Mechanics*, 1–15. https://doi.org/10.1007/s40571-023-00610-0

Rump, M., Sen, U., Jeurissen, R., Reinten, H., Versluis, M., Lohse, D., & Segers, T. (2023). Selective evaporation at the nozzle exit in piezoacoustic inkjet printing. *Physical Review Applied*, *19*(5), 054056. https://doi.org/10.1103/PhysRevApplied.19.054056

Sabu, S. (2022). A Review on Stitching Methods for the Inkjet Nozzles. In *The 2nd Advanced Manufacturing Student Conference* (AMSC22). Chemnitz, Germany 07–08 July 2022, Vol. 7, p. 227.

Sarmadi, M., Behrens, A. M., McHugh, K. J., Contreras, H. T., Tochka, Z. L., Lu, X., & Jaklenec, A. (2020). Modeling, design, and machine learning-based framework for optimal injectability of microparticle-based drug formulations. *Science advances*, *6*(28), eabb6594. https://doi.org/10.1126/sciadv.abb6594

Schenck, H., & Richardson, P. D. (1961). Theories of engineering experimentation. *Journal of Applied Mechanics*, 28(4), 638. https://doi.org/10.1115/1.3641806

Segura, L. J., Li, Z., Zhou, C., & Sun, H. (2023). Droplet evolution prediction in material jetting via tensor time series analysis. *Additive Manufacturing*, 66, 103461. https://doi.org/10.1016/j.addma.2023.103461

Seipel, S.; Yu, J., & Nierstrasz, V. A. (2020). Effect of physical parameters and temperature on the piezo-electric jetting behaviour of UV-curable photochromic inks. *Scientific Reports*, *10*(1), 18841. https://doi.org/10.1038/s41598-020-75449-z

Sethian, J. A., & Smereka, P. (2003). Level set methods for fluid interfaces. *Annual review of fluid mechanics*, *35*(1), 341–372.

https://doi.org/10.1146/annurev.fluid.35.101101.161105

Shah, M. A., Lee, D. G., & Hur, S. (2019). Design and characteristic analysis of a MEMS piezo-driven recirculating inkjet printhead using lumped element modeling. *Micromachines*, *10*(11), 757. https://doi.org/10.3390/mi10110757

Shah, M. A., Lee, D. G., Lee, B. Y., & Hur, S. (2021). Classifications and applications of inkjet printing technology: A review. *IEEE Access*, 9, 140079–140102. https://doi.org/10.1109/ACCESS.2021.3119219

Sidaravičius, D. J. (2012). Densitometric and Colourimetric Control of Printing Materials and Prints. Technika.

Sinha, U., Mynam, M., Nadimpalli, N. K. V., & Runkana, V. (2023). Droplet impingement on a solid surface: Parametrization and asymmetry of dynamic contact angle model. *Physics of Fluids*, *35*(6). https://doi.org/10.1063/5.0147849

Smith, P. J., & Shin, D. H. (2012). Inkjet-based micromanufacturing. John Wiley & Sons.

Smithers. (2020). The future of Inkjet Printing to 2025 (Report). https://www.smithers.com/services/market-reports/printing/future-of-inkjet-printing-to-2025

Solanki, S., Lee, S., Jebakumar, A., Lum, J., Hamidi-Haines, M., Denison, C., & Torniainen, E. (2022). Machine learning for predicting microfluidic droplet generation properties. *Computers & Fluids*, 247, 105651. https://doi.org/10.1016/j.compfluid.2022.105651

Son, Y., Kim, C., Yang, D. H., & Ahn, D. J. (2008). Spreading of an inkjet droplet on a solid surface with a controlled contact angle at low Weber and Reynolds numbers. *Langmuir*, 24, 2900–2907. https://doi.org/10.1021/la702504v.

Sun, S., Zhou, M., Lu, W., & Davarpanah, A. (2020). Application of symmetry law in numerical modeling of hydraulic fracturing by finite element method. *Symmetry*, *12*(7), 1122. https://doi.org/10.3390/sym12071122

Takagi, D., Lin, W., Matsumoto, T., Yaginuma, H., Hemmi, N., Hatada, S., & Seo, M. (2019). High-precision three-dimensional inkjet technology for live cell bioprinting. *International journal of bioprinting*, 5(2). https://doi.org/10.18063/ijb.v5i2.208

Tiyyagura, H. R., Majerič, P., Bračič, M., Anžel, I., & Rudolf, R. (2021). Gold inks for inkjet printing on photo paper: Complementary characterisation. *Nanomaterials*, *11*(3), 599. https://doi.org/10.3390/nano11030599

Tomas, J. (2000). Particle adhesion fundamentals and bulk powder consolidation. *KONA Powder and Particle Journal*, 18, 157–169. https://doi.org/10.14356/kona.2000022

Tomas, J. (2004). Fundamentals of cohesive powder consolidation and flow. *Granular Matter*, 6(2), 75–86. https://doi.org/10.1007/s10035-004-0167-9

Tomas, J. (2007). Adhesion of ultrafine particles—a micromechanical approach. *Chemical Engineering Science*, 62(7), 1997–2010. https://doi.org/10.1016/j.ces.2006.12.055

Tomas, J. (2009). Micromechanics of ultrafine particle adhesion—Contact models. In *AIP Conference Proceedings*, 1145 (1), (pp. 781–784). American Institute of Physics. https://doi.org/https://doi.org/10.1063/1.3180044

Tykhoniuk, R., Tomas, J., Luding, S., Kappl, M., Heimc, L., & Butt, H. J. (2007). Ultrafine cohesive powders: From interparticle contacts to continuum behaviour. *Chemical Engineering Science* 62 (pp. 2843–2864). https://doi.org/10.1016/j.ces.2007.02.027

Wang, H., & Hasegawa, Y. (2023). Multi-objective optimization of actuation waveform for high-precision drop-on-demand inkjet printing. *Physics of Fluids*, *35*(1). https://doi.org/10.1063/5.0122124

Wang, J., & Chiu, G. T. C. (2023). Control of on-demand nanoliter drop volume and jetting velocity in piezoelectric inkjet printing. *Mechatronics*, 94, 103031. https://doi.org/10.1016/j.mechatronics.2023.103031

Wang, J., & Chiu, G.T.C. (2020). Data-driven drop formation modeling in nanoliter dropon-demand inkjet printing. *Dynamic Systems and Control Conference*. American Society of Mechanical Engineers, 84287, pp. V002T28A002. https://doi.org/10.1115/DSCC2020-3292

Wang, J., Li, M., & Nguyen, T. (2023). Mechanisms of Droplet Formation and Deposition in Drop-On-Demand Needle-Valve Inkjets for Precision 3D Microprinting. *International Journal of Precision Engineering and Manufacturing-Green Technology*, 1–14. https://doi.org/10.1007/s40684-023-00515-6

Wang, X., Gong, L., Li, Y., & Yao, J. (2022). Developments and applications of the CFD-DEM method in particle-fluid numerical simulation in petroleum engineering: A review. *Applied Thermal Engineering*, 119865.

https://doi.org/10.1016/j.applthermaleng.2022.119865

Wang, Y., Chen, J., Yin, Z., & Li, Y. (2023). A high-adaptability nozzle-array printing system based on a set covering printing planning model for printed display manufacturing. *Scientific Reports*, *13*(1), 156. https://doi.org/10.1038/s41598-022-24135-3

Wang, Y., Liu, M., Zhang, Y., Liu, H., & Han, L. (2023). Recent methods of droplet microfluidics and their applications in spheroids and organoids. *Lab on a Chip*, 23(5), 1080–1096. https://doi.org/10.1039/D2LC00493C

Wang, Y., Yin, Z., Bao, F., & Shen, J. (2022). CFD-DEM Coupling Model for Deposition Process Analysis of Ultrafine Particles in a Micro Impinging Flow Field. *Micromachines*, *13*(7), 1110. https://doi.org/10.3390/mi13071110

West, T. G., & Yoo, J. (2023). Inkjet and Binder Jet Printing in Pharmaceuticals. In Banerjee, S. (Eds.), *Additive Manufacturing in Pharmaceuticals*. Springer. https://doi.org/10.1007/978-981-99-2404-2_6

Wijshoff, H. (2010). The dynamics of the piezo inkjet printhead operation. *Physics reports*, 491(4-5), 77–177. https://doi.org/10.1016/j.physrep.2010.03.003

Wu, G., Chen, S., Du, W., Zhai, S., Zeng, S., Yu, Y., & Zhou, W. (2022). Simulation on a three-dimensional collision of a moving droplet against a moving super-hydrophobic particle. *Powder Technology*, 405, 117558. https://doi.org/10.1016/j.powtec.2022.117558

Xiao, X., Li, G., Liu, T., & Gu, M. (2022). Experimental study of the jetting behavior of high-viscosity nanosilver inks in inkjet-based 3d printing. *Nanomaterials*, *12*(17), 3076. https://doi.org/10.3390/nano12173076 Yang, X., Lin, Y., Wu, T., Yan, Z., Chen, Z., Kuo, H. C., & Zhang, R. (2022). An overview on the principle of inkjet printing technique and its application in micro-display for augmented/virtual realities. *Opto-Electronic Advances*, *5*(6), 210123–1. https://doi.org/10.29026/oea.2022.210123

Yoo, H., & Kim, C. (2015). Experimental studies on formation, spreading and drying of inkjet drop of colloidal suspensions. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, 468, 234–245. https://doi.org/10.1016/j.colsurfa.2014.12.032

Yoshida, Y., Izumi, K., & Tokito, S. (2019). A push-mode piezo inkjet equivalent circuit model enhanced by diaphragm displacement measurements. *AIP Advances*, 9(2). https://doi.org/10.1063/1.5088384

Zhang, J., Peng, K., Xu, Z. K., Xiong, Y., Liu, J., Cai, C., & Huang, X. (2023). A comprehensive review on the behavior and evolution of oil droplets during oil/water separation by membranes. *Advances in Colloid and Interface Science*, 102971. https://doi.org/10.1016/j.cis.2023.102971

Zhang, Y., Hu, G., Liu, Y., Wang, J., Yang, G., & Li, D. (2022). Suppression and utilization of satellite droplets for inkjet printing: a review. *Processes*, *10*(5), 932. https://doi.org/10.3390/pr10050932

Zhao, D., Zhou, H., Wang, Y., Yin, J., & Huang, Y. (2021). Drop-on-demand (DOD) inkjet dynamics of printing viscoelastic conductive ink. *Additive Manufacturing*, 48, 102451. https://doi.org/10.1016/j.addma.2021.102451

Zhu, H. P., Zhou, Z. Y., Yang, R. Y., & Yu, A. B. (2007). Discrete particle simulation of particulate systems: theoretical developments. *Chemical Engineering Science*, *62*(13), 3378–3396. https://doi.org/10.1016/j.ces.2006.12.089

Zhu, L., Zhao, Z., Liu, C., Li, W., Zhang, Y., Zhang, Y., & Gao, X. (2023). CFD-DEM simulations of a fluidized bed with droplet injection: Effects on flow patterns and particle behavior. *Advanced Powder Technology*, *34*(1), 103897. https://doi.org/10.1016/j.apt.2022.103897

# **Author's Publications Collection**

## Papers in the Reviewed Scientific Journals*

Tofan, T., Kruggel-Emden, H., Turla, V., & Jasevičius, R. (2021). Numerical modeling of the motion and interaction of a droplet of an inkjet printing process with a flat surface. *Applied Sciences*, *11*(2), 527. https://doi.org/10.3390/app11020527

Tofan, T., Stonkus, R., & Jasevičius, R. (2021). Investigation of Colour Reproduction on Linen Fabrics when Printing with Mimaki TX400-1800D Inkjet with Pigment TP250 Dyes. *Coatings* 2021, *11*(3), 354. https://doi.org/10.3390/coatings11030354

Tofan, T., & Jasevičius, R. (2021). Modelling of the Motion and Interaction of a Droplet of an Inkjet Printing Process with Physically Treated Polymers Substrates. *Applied Sciences*, *11*(23), 11465. https://doi.org/10.3390/app112311465

Tofan, T., Borodinas, S., Kačianauskas, R., & Jasevičius, R. (2022). Modelling 3D Droplet Movement Using a Drop-on-Demand Inkjet Printhead Model. *Processes 2022*, *10*(8), 1467. https://doi.org/10.3390/pr10081467

^{*} The dissertation is defended on the basis of these articles. Full-text articles are available at the end of the chapter "Author's Publications Collection". Articles published with the consent of the publisher /-s.

### **Papers in Other Editions**

Tofan, T., Jasevičius, R., & Kruggel-Emden H. (2022). Numerical simulation of the movement of microparticles. In *AIP Conference Proceedings* 2425 (No. 1). AIP Publishing. https://doi.org/10.1063/5.0082405

Tofan, T., Hurtado, A. M., Kruggel-Emden, H., & Jasevičius, R. (2023). Numerical simulation of the inkjet droplet interaction with a solid surface. In *AIP Conference Proceedings* 2849 (No. 1). AIP Publishing. https://doi.org/10.1063/5.0163094

# Article 1. Tofan, T. et al. (2021). Numerical Modeling of the Motion and Interaction of a Droplet of Inkjet Printing Process with a Flat Surface (https://doi.org/10.3390/app11020527)





Articla

#### Numerical Modeling of the Motion and Interaction of a Droplet of an Inkjet Printing Process with a Flat Surface

Tim Tofan¹, Harald Kruggel-Emden², Vytautas Turla³ and Raimondas Jasevičius^{1,4,*}

- Department of Mechanical and Material Engineering, Vilnius Gediminas Technical University,
- J. Basanavicius str. 28, Vilnius 03224, Lithuania; tim.tofan@vilniustech.lt
- Chair of Mechanical Process Engineering and Solids Processing, Technical University of Berlin, Str. des 17. Juni 135, 10623 Berlin, Germany; kruggel-emden@tu-berlin.de
- Department of Mechatronics, Robotics and Digital Manufacturing, Vilnius Gediminas Technical University, J. Basanavicius str. 28, Vilnius 03224, Lithuania; vytautas.turla@vilniustech.lt
  - Institute of Mechanical Science, Vilnius Gediminas Technical University, J. Basanavicius str. 28, Vilnius 03224, Lithuania
- Correspondence: raimondas.jasevicius@vilniustech.lt

Abstract: The numerical simulation and analysis of the ejection of an ink droplet through a nozzle as well its motion through air until its contact with a surface and taking up of a stable form is performed. The fluid flow is modeled by the incompressible Navier-Stokes equations with added surface tension. The presented model can be solved using either a level set or a phase field method to track the fluid interface. Here, the level set method is used to determinate the interface between ink and air. The presented work concentrates on the demonstration how to check the suitability of ink for inkiet printhead nozzles, for instance, for the use in printers. The results such as velocity, change of size, and volume dependence on time of an ink droplet are presented. Recommendations for the use of specific inks are also given.



Citation: Tofan, T.; Kruggel-Emden H.; Turla, V.; Jasevičius, R. Numerical Modeling of the Motion and Interaction of a Droplet of an Inkjet

Printing Process with a Flat Surface.

Publisher's Note: MDPI stays neu-

tral with regard to jurisdictional clai-

ms in published maps and institutio-

Copyright: © 2021 by the authors. Li-

censee MDPI, Basel, Switzerland.

This article is an open access article

distributed under the terms and con-

ditions of the Creative Commons At-

tribution (CC BY) license (https://

creativecommons.org/licenses/by/

Appl. Sci. 2021, 11, 527. https://

doi.org/10.3390/app11020527

Received: 7 December 2020

Accepted: 5 January 2021

Published: 7 January 2021

nal affiliations

4.0/).

0

Keywords: drop-on-demand inkjet; printing nozzle; droplet; measurement of droplet trajectory; COMSOL

#### 1. Introduction

Nowadays, inkjet printing has become relevant besides application for brochures, flyers, and other paper-based products, also in areas such as shirt printing or personalized production while thereby satisfying increasing quality demands. In this way, inkjet printing is continuously enhancing its capabilities. According to Smithers Pira's [1] report, the inkjet printing market will grow at least till 2023, both by deepening its penetration into existing markets, and exploiting new emergent market opportunities. In doing so, there are three main markets where inkjet technology will increase in relevance: book printing, commercial printing, and packaging printing. With the printing market is expected to grow, there are many propositions to improve the productivity of inkjet processes. Nowadays, inkjet printing is worldly used in many manufacturing applications. It shows the same quality and speed as many other traditional printing methods such as flexography or lithography (Castreion-Pita et al. [2]).

Of relevance for inkjet printing is the drop-on-demand (DOD) inkjet technology. DOD technology is to eject small droplets of ink from a printing head nozzle onto a substrate only when required to form the image. Drops are generated by quick pressure increase inside a nozzle to eject the ink. The most popular technology to increase pressure is to use a piezoelectric element in every nozzle chamber. When a voltage is applied to the actuator, the piezoelectric element changes its shape to displace the chamber wall or roof into the chamber to generate the required pressure for the drop ejection, Hoath [3]. The main advantage in inkjet printing is the possibility to deposit different sized droplets one by one on many different materials with predetermined locations on the substrate without

Appl. Sci. 2021, 11, 527. https://doi.org/10.3390/app11020527

#### Appl. Sci. 2021, 11, 527

any contact. To perfectly fulfill its demand, inkjet printing is required to produce small quantity prints with low cost and high-resolution quality at tolerable requirements. Every year, inkjet printing is increasing in popularity and progressing in different using areas.

Inkjet printing technology has been extensively used due to its advantages regarding flexible droplet formation. There are various areas where inkjet technologies can be applied. As a consequence, there are many investigations on inkjet printing addressing things such as nozzle configurations (parameters), different jetting velocities, droplet sizes, and frequency of jets (Stringer and Derby [4], Wijshoff [5], Bos et al. [6]). Inkjet technology also can be adjusted by making microlenses for optical devices, Yuzo et al. [7]. Furthermore, inkjet technology can be used as a manufacturing technique for producing ceramic parts (Ainsley et al. [8]) or making collagen scaffolds (biomaterials) with a predefined internal morphology, Sachlos et al. [9].

It is found that a nozzle configuration affects droplet shape and velocity which is beneficial for the enhanced printing quality and high-throughput printing rate, Aqeel et al. [10].

In case of ink-jet printing technology which is non-contacting, material can be printed layer-on-layer to configure 3-D structures. This capability enabled the first use of ink-jet printing to build active microelectromechanical systems (MEMS), Fuller [11]. Despite such progress, for the moment, there are not many works to define how ink will perform over its lifetime and how ink structure changes over time. Such experiments are in progress at different universities and research institutes, Korvink et al. [12]. Nonetheless, the so far performed research places inkjet technology as the key focus for technology developers.

In the past, DOD printhead nozzle droplet ejection and the settling process were investigated with regard to the printing quality. The ability of modeling the jetting process and droplet settling parameters is important as it can be used as a tool for future ink and printing device developments. In the past, much research addressing the inkjet printing process was done by slow motion cameras (Martin, et al. [13]). Thereby pictures are examined, and droplet parameters can be calculated from 2D views. In addition, it is possible to address droplet motion by numerical simulations, Wu et al. [14]. Many authors (Bos et al. [6], Tan et al. [15]) have conducted research to compare differences between numerical simulations and slow motion camera observations. Obtained results are in agreement and based on the numerical simulations performed, of which we can base our inkjet droplet model.

It should be noted the current state of art, which may be related to the work presented here. Various ways of implementation of DOD technology and related investigations can be found in scientific literature. The development of drop-on-demand electrohydrodynamic jet (DOD E-Jet) printing to enhance print control and to achieve stable micro-structures on a flexible insulating substrate were analyzed by Abbas et al. [16]. The theoretical design of the inkjet process to improve delivery efficiency was investigated by comparing numerical investigations and physical experiments by Zhong et al. [17]. A high temperature drop-on-demand droplet generator for metallic melts was analyzed by Inani Moqadam et al. [18]. Microstructure adjustment of spherical micro-samples for high-throughput analysis using a drop-on-demand droplet generator were also analyzed by the latter authors [19]. Electro-hydrodynamic drop-on-demand printing of aqueous suspensions of drug nanoparticles were introduced by Elele et al. [20]. The effect of meniscus damping ratio on drop-on-demand electrohydrodynamic jetting was analyzed by Kim et al. [21].

The analysis of droplet motion is still relevant. A one-dimensional model for the droplet ejection process in inkjet devices was introduced by Jiang and Tan [22]. They showed that a 1D model can significantly reduce the computational time (usually less than one minute) yet with acceptable accuracy. The calculation of the critical point regarding the distribution of the droplets generated by various materials in the dimensionless plane considering piezoelectric print-heads (PPHs) were investigated by Wang et al. [23]. Ejection state prediction for a pneumatic micro-droplet generator by backpropagation (BP) neural networks was analyzed by Wang et al. [24]. The simulation and validation of the droplet generation process for revealing three design constraints in electrohydrodynamic

2 of 15

#### Appl. Sci. 2021, 11, 527

jet printing is given by Pan and Zeng [25]. Printed strain sensors based on an intermittent conductive pattern filled with resistive ink droplets were presented by Zymelka et al. [26]. Dynamics of electrowetting droplet motion in digital microfluidic systems was analyzed by Cui et al. [27].

The use of "Jet" technologies offers a variety of technical implementation options. A theoretical model for an electrostatic lens that is incorporated into an e-jet system to shape the electric field inside the printhead was introduced by Liu and Huang [28]. Controlling the voltage applied to electrodes located around the jet, its trajectory can be continuously adjusted by lateral accelerations which was investigated by Liashenko et al. [29]. Aerosol-jet printed nanocomposites for flexible and stretchable thermoelectric generators were analyzed by Ou [30]. A model and calculations referring to the shear rate of ink in an inkjet printer nozzle were given by Dybowska-Sarapuk et al. [31]. A review of the issues that come along with preparing and printing carbon nanotube ink was given by Tortorich et al. [32]. Inkjet printing of drug-loaded mesoporous silica nanoparticles was analyzed by Wickström et al. [33]. They suggest that inkjet printing technology could function as a flexible deposition method of pharmaceutical suspensions.

The printing can be applied even for biological structures. Mechanical properties of 3D-printed blood vessels were analyzed by Wang et al. [34]. Drop-on-demand (DOD) 3D bioprinting technology was introduced by Grottkau et al. [35].

Based on the good agreement of numerical and experimental investigations regarding the DOD printing process, our droplet simulation model has been developed with the modeling program COMSOL to investigate the different droplet parameters of the DOD printing process. This simulation model can be used to predict the behavior of the droplets in the printing process. The parameters of the droplet ejection, flight, and impact with the substrate will be analyzed to also elaborate on improved printing conditions.

The numerical simulations are initiated using a laminar-two-phase flow function setting and a level set interface. All stages from the main nozzle reservoir where the droplet is forming to the final interaction with the substrate are considered.

With the derived model, it is possible to configure inkjet nozzle properties such as the geometry of the nozzle, the inlet velocity, as well as parameters to study the influence on the size and the speed of the ejected droplet. With the current model, it becomes also possible to investigate how the printing process would perform under changes of the inkjet inks.

#### 2. Problem Formulation

There are many investigations available, focused on drop impact with different surfaces, different jetting parameters, and different Weber and Reynolds numbers (definitions provided later on), Son et al. [36], Bussmann et al. [37]. But there is growing interest on variable fluid drop impact with hydrophobic surfaces in reflecting many practical applications such as printing on polymers.

Researchers are thereby paying close attention to studying the parameters of the nozzle. Many works examine the properties of the nozzle (shape, dimensions, orifice parameters, piezoelectric excitation) that determine the size and velocity of the droplet formed and the circumstances of satellite droplets, Driessen [38]. Satellites are several times smaller droplets than the primary droplet. They can appear if unsuited parameters of ejection or unsuited inks are used. For high-quality printing, they should not appear.

The aim of this work is to provide a numerical model that can help evaluate the shape-changing droplet motion, various initial parameters, and effects/causes influencing this motion, which is an important key component in the inkjet process needed to predict difficult moments or even issues at an early state of the ink developing process. The typical process of DOD droplet formation consists of several stages. This study examines the vertical fall/movement of a droplet formed at the inkjet priority nozzle in air onto a substrate while changing its shape (Figure 1). Droplet status is changing at different

3 of 15

55



instances in time, which are represented by the considered points from A through F. These basic points help to define how a droplet forms and how parameters change over time.

(b)

Figure 1. Droplet fall/movement. (a) During the inkjet printing processes, the droplet is ejected from the inkjet nozzle and settles while changing its shape at different stages (A-F); (b) Highlighted inkjet droplet phases at different stages (A-F). Phases from the left: A—ejection and formation of the droplet; B—droplet pinch off from the nozzle; C—droplet sphere forming; D—droplet before the interaction with the surface; E—droplet spreading at the surface; F—droplet in final stable form.

At point A, the ejection and formation of the droplet comes along with the maximum droplet velocity, then pressure inside the nozzle reaches its maximum and the droplet is ejected from the nozzle. At point B, the droplet thread is pulled off from the printhead nozzle, at this point the droplet has its maximum length and final droplet mass and volume. Point C represents the droplet at almost spherical form when the droplet is formed. Before point C and D, the droplet does not change shape and loses velocity during its settling. Point D represent the instance where droplet interaction with the surface begins, and view E represents the droplet at maximum diameter while spreading. Final point F represents the droplet while oscillating to its final stable form.

In all six phases, different phenomena occur. It can be noticed that the changing forms of the droplet over time can be better explained by considering different instances in time. In the next sections, the different parameters and time sections will be discussed. An overview of the numerical experiment will be given while an insight into the methods used in the investigation is given along with the final parameters. In this research, a closer look is taken at the droplet shape, size, and volume formation and velocity turnover for three different ink parameters. Droplets are generated to perform particularly a vertical fall/movement in air on the substrate. The droplet formation, movement, and impact are modeled by applying a level set function and the Navier–Stokes equations by the commercial finite element package COMSOL Multiphysics. The modeling as part of COMSOL Multiphysics

4 of 15
allows similar to the experimental methods; we have used the plotting of flight path as well as the determination of droplet dimensions, velocity, and mass. Additionally, the drag and gravity forces can be also evaluated.

# 3. Methodology for the Simulation

To represent the fluid interface and convection with air as part of a laminar twophase flow, the level set method was considered (Olsson and Kreiss [39], Sun and Beckermann [40]). Inkjet inks are treated as Newtonian fluid. According to COMSOL Module User's Guide [41], the level set function defines the interface, where  $\phi = 0$  represents air and  $\phi = 1$  is ink. In a transition layer close to the interface,  $\phi$  goes smoothly from 0 to 1. The interface moves together with the fluid velocity, u, at the interplay. The following equation describes the convection of the reinitialized level set function, it thereby describes the interface between the 2 phases, ink and air:

$$\frac{\partial \Phi}{\partial t} + \boldsymbol{u} \cdot \nabla \Phi = \gamma \nabla \cdot \left( \varepsilon \nabla \Phi - \Phi (1 - \Phi) \frac{\nabla \Phi}{|\nabla \Phi|} \right). \tag{1}$$

Here,  $\varepsilon$  is corresponding with the thickness of the transition layer. For this model,  $\varepsilon = hc/2$ , where hc is the representative mesh size in the area passed by the falling droplet. As default, this interface thickness controlling parameter hc it generated automatically and for the current simulation is set to hc = 2.5 µm.

The parameter  $\gamma$  determines the amount of reinitialization or stabilization of the level set function, Olsson and Kreiss [39]. A suitable value for  $\gamma$  is the maximum magnitude occurring in the velocity field. As default, this reinitialization parameter  $\gamma$  is generated automatically and for the current simulation set to  $\gamma = 10 \text{ m/s}$ .

Beside defining the fluid interface, the level set function is used to smooth the density and viscosity across the interface. Density and dynamic viscosity depend on the interface coefficient  $\phi$ . Thereby changes in density and pressure during the calculations are considered, Olsson and Kreiss [39]. Density can be calculated as follows:

$$\rho = \rho_1 + (\rho_2 - \rho_1)\phi. \tag{2}$$

Here,  $\rho_1$  denotes the air density while  $\rho_2$  denotes the ink density. For the numerical simulation, the dynamic viscosity (shear viscosity)  $\mu$  must be taken into account (3). It describes the relationship between the shear rate and the shear stresses in the fluid. Dynamic viscosity is calculated by:

$$\mu = \mu_1 + (\mu_2 - \mu_1)\phi. \tag{3}$$

Here,  $\mu_1$ —is the air viscosity and  $\mu_2$ —is the ink viscosity. The transportation of mass and momentum are described by the incompressible Navier–Stokes equations. Both ink and air can be considered as incompressible if the fluid velocity is small compared to the speed of sound.

We investigate ink and air as fluid whose viscous stresses arise from its flow at every point. The Navier-Stokes equations describe the transfer of mass and impulses to an uncompressible fluid, including surface tension and gravity forces. The ink motion including mass conservation is calculated by the following differential equations (Sohr [42]):

$$\nabla \cdot \mathbf{u}) = \mathbf{0},\tag{4}$$

$$\rho\left(\frac{\partial \boldsymbol{u}}{\partial t} + \boldsymbol{u} \cdot \nabla \boldsymbol{u} - \boldsymbol{g}\right) - \nabla \cdot \left(\mu \left(\nabla \boldsymbol{u} + \nabla \boldsymbol{u}^{\mathrm{T}}\right)\right) + \nabla \boldsymbol{p} = \mathbf{F}_{\mathrm{st}}.$$
(5)

Here,  $\rho$  denotes the density,  $\mu$  is the dynamic viscosity, u represents the fluid velocity, p denotes pressure, and Fst is the surface tension force. To capture the effect of the surface tension, a volume force is added at the phase interface in the computational domain where the phase interface is at present. The force depends on the surface tension coefficient and the curvature of the phase interface.

The surface tension force is computed by:

$$\sigma_{st} = \sigma \delta \kappa \mathbf{n},$$
 (6)

where **n** is the unit vector in the normal direction,  $\sigma$  is the surface tension coefficient which varies according to the curve, and  $\delta$  equals a Dirac delta function that is nonzero only at the fluid interface,  $\kappa = -\nabla \cdot \mathbf{n}$  is the curvature. The unit vector in the normal direction is given by:

Ē

1

$$\mathbf{n} = \frac{\nabla \Phi}{|\nabla \Phi|},\tag{7}$$

while the delta function is approximated by:

$$\delta = 6|\phi(1 - \phi)||\nabla \phi|. \qquad (8)$$

To solve Equations (4) and (5), the adaptive mesh refinement functionality for inkjet nozzle modeling as part of COMSOL Multiphysics [41] is used to locally refine the mesh around the ink and air interface. This functionality will essentially divide the simulation into several time intervals, and locally refine the mesh in the region where the phase interface is present in each interval to increase calculation accuracy.

Applying this, the droplet mass can be calculated by:

$$m_d = \rho(V \cdot \phi).$$
 (9)

There are two dimensionless parameters which characterize droplets: the Reynolds number and the Weber number, Hoath [3]. The Reynolds number Re represents the ratio between inertial and viscous forces in a moving fluid and the Weber number, we commutate on the ratio of the inertia to the surface tension. These dimensionless numbers are defined by:

$$Re = \frac{\rho v_d d}{\mu}$$
 and  $We = \frac{\rho v_d^2 d}{\sigma}$ , (10)

where d is a length parameter—typically, the diameter of the jet, nozzle, or drop and  $v_d$  is the droplet velocity. In this research, the nozzle diameter will be used for calculating Re and We numbers.  $\sigma$  is the surface tension coefficient. The influence of velocity in these two dimensionless numbers can be removed by forming a further number, the Ohnesorge number Oh defined by:

$$Oh = \frac{\sqrt{We}}{Re} = \frac{\mu}{\sqrt{\sigma\rho d}}.$$
(11)

The value of the Ohnesorge number Oh, which reflects only on the physical properties of the liquid and the size scale of the jet or drop, is independent of the driving conditions (which control the velocity). *Oh* turns out to be closely related to the behavior of a jet emerging from a nozzle and, thus, to the conditions in DOD printing, Derby [43,44]. If the Ohnesorge number is too high ( $Oh \ge 1.0$ ), then viscous forces will prevent the separation of a drop, while if it is too low ( $Oh \le 0.1$ ), the jet will form a large number of satellite droplets. Satisfactory performance of a fluid in DOD inkjet printing requires an appropriate combination of physical properties, which will also depend on the droplet size and velocity (through the value of the Reynolds or Weber number).

Some authors use the symbol Z as inversed of the Ohnesorge number, Hoath [3]:

$$Z = \frac{1}{Oh}.$$
 (12)

Ink printability is considered to be appropriate for ink jet printing if Z in its original definition is between 1–10. Today's research shows that this range can be adjusted and there are variations possible as higher numbers than 10 can also show good printing

characteristics. In this sense, several exceptions were found and reported to the initial range suggested by Derby [43,44].

Ink parameters of the different inks considered in this investigation were aligned with those of ink manufacturers and are assignable to the good printability range (Magdassi [45]). Considered digital ink parameters in this investigation as well as those of air are shown in Table 1.

Table 1. Physical parameters of ink and air used in modeling the inkjet nozzle.

Substance	ρ, kg/m ³	$\mu$ , mN·s/m ²	σ, mN/m	Oh	Z
Air	1.225	$1.789 \cdot 10^{-2}$	-	-	~
Ink 1st	1050	7	72	0.16	6.09
Ink 2nd	1080	10	72	0.23	4.32
Ink 3rd	1110	14	72	0.32	3.13

Because of nozzle symmetry, an axisymmetric 2D model is used to simplify the calculations performed here. The true 3D inkjet nozzle geometry is shown in Figure 2. Thereby, the distance from the nozzle outlet to the substrate is 1 mm.



Figure 2. Geometric parameters of the inkjet nozzle.

Initially, the space between the inlet and the nozzle outlet is filled with ink. Then, ink is ejected through the nozzle during a period of 2.0 µs and it is consequently forced to flow out of the nozzle. When the ejection stops, a droplet volume still grows for a while and snaps off the nozzle. After that, the droplet is changing form and continues to travel until it hits the target. During this process, gravity g is considered, having a value 9.8066 m/s².

To control the droplet size and velocity during the ejection, a rectangular function is applied (Figure 3) for the inlet velocity that forces a pressure increase inside the modeled nozzle. It helps to set the length of the pulse and the value of the maximum nozzle inlet fluid velocity magnitude (maximum value 0.2 m/s which is a realistic value to increase pressure in the inkjet nozzle for inkjet printing). Both parameters can affect the resulting velocity of the ejected droplet, but the magnitude (depends on the time of the droplet formation) has a larger influence on the droplet velocity.



Figure 3. Rectangle function of the increasing pressure in the nozzle.

# 4. Obtained Results

The droplet status considered at different instances in time will help to demonstrate the change of parameters of an ejected droplet which are represented by the considered points from A through F (Table 2; Figures 1 and 2). Here,  $D_{d \cdot max}$  determinates the droplet max diameter in the horizontal direction, while  $D_h$  determinates the droplet height in vertical direction. These basic points help to define the droplet status over time t.

Table 2. Parameters of the considered inks at different points in time.

	Ink ₁					Ink ₂				Ink ₃			
	t μs	v _d ms	D _{d.max} µm	D _h µm	t µs	v _d ms	D _{d.max} µm	D _h µm	t μs	$v_d$ ms	D _{d.max} µm	D _h μm	
A	2	13.8	26.3	9.2	2	13.8	25.8	9.7	2	13.8	23.5	9.9	
в	11	8.51	20.4	110.9	13	7.88	20.2	115.3	14	7.28	20.0	118.7	
C	26	7.05	23.3	26.2	32	6.77	24.9	26.3	39	5.93	24.0	26.6	
D	143	4.48	22.2	21.7	150	4.28	22.2	21.8	169	3.60	21.9	22.0	
E	149	0.18	47.7	4.3	161	0.12	42.8	7.2	183	0.05	37.5	11.6	
F	174	0	36.0	11.6	172	0	36.1	12.1	181	0	36.3	11.9	

While analyzing the different ink parameters, firstly were calculated Reynolds, Weber, Ohnesorge, and Z numbers. Inks droplet parameter space with axes showing the Reynolds and Weber numbers are shown in Figure 4. Based on these numbers, all inks are suitable for inkjet printing, according to Derby [43,44] and Hoath [3]. But we can see some clear differences in droplet behavior, see Table 2.

Droplet propagation parameters were obtained by COMSOL software. The related graphics can help to understand the behavior and properties of the droplet during ejection, progression in air, and surface interaction, whereby no wettability is considered. Droplet velocity after ejection is not bigger than 10 m/s and compared to Wijshoff [46,47] varies uniformly. Firstly, the droplet velocity during ejection, during the flight, and during impact is outlined in Figure 5.

At point A, the droplets of the different inks reach the same maximum velocity at the maximum pressure in the nozzle chamber leading to the ejection of the droplet. The B point corresponds to the droplet thread pulled off from the printhead nozzle—here, different inks have different starting velocities for droplet formation—to assuming spherical form at point C where velocities do not change much.



Figure 4. Representation of the Reynolds and Weber numbers, showing the ink properties.



Figure 5. Representation of the droplets (Ink1, Ink2, Ink3) velocity turnover. Velocity versus time.

Point D represents the beginning of the droplet interaction with the surface, the different inks reach various velocities before impact with the surface. At point E, the spreading moment, inks with bigger density and viscosity take final sphere form faster and do not oscillate much. Lower density and viscosity inks rebound more, which is possible to observe by the increasing velocity after the impact. Ink₃ does not rebound and deposits least. Final point F represent the droplet in final-stable form which is reached at different times after the ejection:  $F_1$ -174  $\mu$ s (from  $D_1$  to  $F_1$ -25  $\mu$ s),  $F_2$ -172  $\mu$ s (from  $D_2$  to  $F_2$ -14  $\mu$ s),  $F_3$ -181  $\mu$ s (from  $D_3$  to  $F_3$ -2  $\mu$ s). Droplets from Ink $_1$  are not so viscous and after the impact, they rebound a few times up and down. That is why these inks reach stable phase later than Ink_2. Ink_3 does not oscillate and takes final form very fast after the impact with the surface.

In addition, we can address differences in the droplet forming parameters depending on time (Figure 6). Here, in Figure 6,  $D_{d,max}$  (Table 2) is the maximum droplet diameter (horizontal cross section);  $D_h$  (Table 2) is droplet height (vertical cross section).



Figure 6. Representation of the droplet size of  $Ink_1$ ,  $Ink_2$ ,  $Ink_3$  versus time.

If we compare the jets of the different inks regarding length and maximum diameter (horizontal cross section), few main differences can be noticed. Ink₁ with lowest density and viscosity parameters has the smallest jet length and reveals the biggest oscillations. The droplet maximum spreading diameters is  $E_1$  = 47.7 µm. The droplet sphere diameter during flight is ~22 µm. The minimum spreading diameter has Ink₃ which does not oscillate at all. The latter depends on droplet density and viscosity parameters—the droplet takes its final form then it touches the substrate.

It is important to know all these parameters if, for example, the use of UV curable inks is intended. Furthermore, after the droplet is generated, it is important to know the time period when the droplet reaches its final state on the surface. If we collate inks in this type of view, Ink₂ reveals the best suitable result. Droplet from Ink₂ does not oscillate much and has the smallest time period to reach a stable form on the surface. Droplets from Ink₁ reach the surface faster but, in case of too low viscosity and density parameters, oscillate more strongly before they take their final form on the surface. For UV curable inks, it is very important to undergo this step in the stable phase of the droplet.

In the next graph (Figure 7), droplet mass change is demonstrated.



Figure 7. Representation of the mass of the droplets (Ink1, Ink2, Ink3) versus time.

Droplet mass is integrated over the computational domain. At point A, the droplet reaches its maximum velocity and the droplet mass starts to increase during the first period till it reaches point B. At point B, the droplet thread is pulled off from the printing nozzle and mass is fully given. Droplets of different inks increase very similarly (Ink₁ fully formed

at 11 µs, Ink₂-13 µs, Ink₃-14 µs). When the liquid droplet is ejected, it needs a certain time and distance to reach the full spherical form and during the impact, it will lose its spherical shape and take final form on the surface. All these depend on the ejecting parameters, droplet velocity, nozzle configurations, and liquid characteristics. In order to examine the differences between ink droplets, their mass can be addressed, which are: Ink₁ droplet  $m_d = 14.4 \cdot 10^{-12}$  kg, Ink₂ droplet  $m_d = 14.8 \cdot 10^{-12}$  kg, Ink₃ droplet  $m_d = 15.2 \cdot 10^{-12}$  kg. From this graphic, we can see that droplet mass stays fixed after point B when the droplet thread is pulled off.

In the next graphic (Figure 8), droplet volume formation is demonstrated.



Figure 8. Representation of the volume versus time of droplets of Ink₁, Ink₂, and Ink₃.

Droplets reach their final volume at the same instance as already obtained for the mass. Inks with highest density and viscosity reach the largest volume:  $Ink_1$  droplet  $V_d$  = 4.3491 pL,  $Ink_2$  droplet  $V_d$  = 4.3493 pL, and  $Ink_3$  droplet  $V_d$  = 4.3495 pL.

# 5. Discussion

The considered initial physical parameters, presented in Table 1, are the physical parameters of ink and air used in modeling the inkjet nozzle. These data allowed the replication of the physical behavior of the droplet. In addition, there are known guidelines within which the printing process can be performed. Droplet formation limits were evaluated and selected in the numerical investigation. Comparing to Dijksman [48], the droplet formation (point A) has similar manner, here velocity is increasing, until the droplet starts to separate from the nozzle (point B). Various formed droplet structures can be expected (Zhong et al. [17]). Physical experiments show that this is the case when a droplet is moving with high speed (up to 20 m/s), while having a speed variance with a large span from 0 to 19.414 m/s, Zhong et al. [17]. In our simulation, we had a lower speed variance up to 14 m/s and satellites were not observed. Our droplet therefore could be classified as a single droplet (Class 1, Zhong et al. [17]). Buying COMSOL Multiphysics which allows modeling the change in speed, volume, and mass of the droplet.

The main aim of this research is to find the best modeling method for further investigations of different printing nozzle parameters, for printable fluid development and droplet ejection impulse parameters. This research should help to investigate printing technologies for broadening the new horizons of printing processes modeling. This work may be useful for further research to discover the best print head settings for printing with different inks or other chemical or biological fluids. It is important to mention that the work is

12 of 15

focused on further research by modernizing the printing inks and print head parameters as well as the droplet printing parameters. It is important to note that inks that may now be unsuitable for printing can adapt perfectly if the shape of the print head shape and the pulse of irritation are properly selected.

In addition to the basics of known experiments, data were sufficient to perform a numerical experiment. In the future, by introducing more parameters and deepening into the droplet behavior, a physical experiment may be necessary. As a first step in revealing the fundamental properties of the droplet shape, moving to different forms, different units of time are sufficient for characterization. The numerical fulfilment itself is also important, as an implementation study with COMSOL. Such study will be useful for scientists who would like to carry out/to repeat and improve this study as well. By using COMSOL, the numerical modeling of the motion of inkjet droplets has been achieved. Using the software, the length of the drop thread, the moment when the drop is pulled out of the thread, when the droplet forms a spherical shape, droplet mass, droplet volume, droplet conditions on impact, and the final state of the droplet was found. In addition, it is now possible to configure different nozzle parameters, different ejection pressures, and test different inks. In addition, future investigations will be dedicated to considering different types of materials, e.g., [31,49].

# 6. Conclusions

It is known that during inkjet printing processes, it is important to know how fast the droplet forms a sphere; which speed it will have before interacting with the surface and what form it will have in the stable phase. It is also important to predict the interaction of the droplet with the surface. Technically, all of this has been obtained and implemented in the COMSOL Multiphysics software in the context of an improved inkjet nozzle application. Droplet ejection, motion, and impact were analyzed using the COMSOL CPD module. Numerical experiments were performed on this basis. The presented results on the inkjet printing process can accurately predict characteristic moments or even issues at an early state of the ink developing process. By the numerical model, all important key aspects of the inkjet printing process were assessed. These can directly affect printing quality. Known effects in physical printing have been observed and presented in a numerical experiment, such as:

- The droplet reaches its maximum velocity at the ejection moment when the pressure inside the nozzle is at the highest level.
- When the thread of the drop is detached from the nozzle, its speed is much lower and during flight, the speed of the drop decreases further.
- After impact, it is observed, that the drop loses its spherical shape and takes on a different form on the surface.

After the numerical experiments, it was observed that further research is needed to analyze the behavior of different inks with categorically different parameter settings, different ejection settings, different nozzle parameters and droplet impact with different surface parameters to further improve the printing process.

Author Contributions: T.T., V.T., H.K.-E. and R.J. conceived the presented idea; processed and analyzed the data, wrote the paper. All authors have read and agreed to the published version of the manuscript.

Funding: The research was funded by the Vilnius Gediminas Technical University (VGTU).

Institutional Review Board Statement: The study was conducted according to the guidelines of theDeclaration of Helsinki, and approved by the Ethics Committee of UNIVERSITAT POLITECNICADE VALENCIA (date of approval: 23 October 2018).

Informed Consent Statement: Informed consent was obtained from all subjects involved in the study.

Data Availability Statement: The data presented in this study are available on request from the corresponding author. The data are not publicly available due to privacy.

# Conflicts of Interest: The authors declare no conflict of interest.

# Abbreviations

#### Roman

- d the diameter of the nozzle (mm);
- D_{d.max} maximum droplet diameter (μm);
- D_h droplet height (μm);
- DOD drop-on-demand;
- F_{st} surface tension force (N);
- gconst acceleration due to gravity (m/s²);
- *m_d* droplet mass (kg);
- n the unit vector in the normal direction;
- Oh dimensionless coefficient;
- p pressure (Pa);
- r droplet radius (mm);
- g gravitational constant (m/s²);
- Re dimensionless coefficient;
- t time (s);
- u fluid velocity (m/s);
- v_d droplet velocity (m/s);
- $V_d$  droplet volume (pL);
- We dimensionless coefficient;
- Z dimensionless number.
- Greek
- $\beta$  maximum spreading factor;
- γ parameter which determines the repetition of initiations;
- δ Dirac delta function;
- ε is the representative mesh size in the area passed by the falling droplet;
- k- is the curvature;
- $\eta$  is fluid viscosity (m/s);
- $\mu$  dynamic viscosity (N·s/m²);
- $\rho$  density (kg/m³);
- σ surface tension coefficient (mN/m);
- φ coefficient of level set interface between air and ink.

# References

- The Future of Inkjet Printing to 2023 Report; SMITHER PIRA: Akron, OH, USA, 2018; Available online: https://www.smithers. com/services/market-reports/printing/the-future-of-inkjet-printing-to-2023 (accessed on 18 June 2018).
- Castrejon-Pita, J.R.; Baxter, W.R.S.; Morgan, J.; Temple, S.; Martin, G.D.; Hutchings, I.M. Future, opportunities and challenges of inkjet technologies. At. Sprays 2013, 23, 541–565. [CrossRef]
- Hoath, S.D. Fundamentals of Inkjet Printing: The Science of Inkjet and Droplets. John Wiley & Sons: Hoboken, NJ, USA, 2016; p. 472. [CrossRef]
- 4. Stringer, J.; Derby, B. Limits to feature size and resolution in ink jet printing. J. Eur. Ceram. Soc. 2009, 29, 913–918. [CrossRef]
- 5. Wijshoff, H. The dynamics of the piezo inkjet printhead operation. *Phys. Rep.* 2010, 491, 77–177. [CrossRef]
- Bos, A.V.D.; Meulen, M.J.V.D.; Driessen, T.; van den Berg, M.; Reinten, H.; Wijshoff, H.; Versluis, M.; Lohse, D. Velocity profile inside piezoacoustic inkjet droplets in flight: Comparison between experiment and numerical simulation. *Phys. Rev. Appl.* 2014, 1, 014004, [CrossRef]
- Yuzo, I.; Shinji, K.; Yoshimitsu, A.; Yasuhiro, A. Inkjet fabrication of polymer microlens for optical-i/o chip packaging. *Jpn. J. Appl. Phys.* 2000, 39, 1490–1493. [CrossRef]
- Ainsley, C.; Reis, N.; Derby, B. Freeform fabrication by controlled droplet deposition of powder filled melts. J. Mater. Sci. 2002, 37, 3155–3161. [CrossRef]
- Sachlos, E.; Reis, N.; Ainsley, C.; Derby, B.; Czernuszka, J.T. Novel collagen scaffolds with predefined internal morphology made by solid freeform fabrication. *Biomaterials* 2003, 24, 1487–1497. [CrossRef]
- Aqeel, A.B.; Mohasan, M.; Lv, P.; Yang, Y.; Duan, H. Effects of nozzle and fluid properties on the drop formation dynamics in a drop-on-demand inkjet printing. *Appl. Math. Mech.* 2019, 40, 1239–1254. [CrossRef]
- Fuller, S.B.; Wilhelm, E.J.; Jacobson, J.M. Ink-jet printed nanoparticle microelectromechanical systems. J. Microelectromechanical Syst. 2002, 11, 54–60. [CrossRef]

- Korvink, J.G.; Smith, P.J.; Shin, D.H. Inkjet-Based Micromanufacturing; John Wiley & Sons: Hoboken, NJ, USA, 2012; p. 388. [CrossRef]
- Martin, G.D.; Hoath, S.D.; Hutchings, I.M. Inkjet Printing-the Physics of Manipulating Liquid Jets and Drops. J. Phys. Conf. Ser. 2008, 105, 012001. [CrossRef]
- Wu, H.C.; Lin, H.J.; Kuo, Y.C.; Hwang, W.S. Simulation of droplet ejection for a piezoelectric inkjet printing device. Mater. Trans. 2004, 45, 893–899. [CrossRef]
- Tan, H.; Torniainen, E.; Markel, D.P.; Browning, R.N. Numerical simulation of droplet ejection of thermal inkjet printheads. Int. J. Numer. Methods Fluids 2015, 77, 544–570. [CrossRef]
- Abbas, Z.; Wang, D.; Du, Z.; Quian, J.; Zhao, K.; Du, Z.; Wang, Z.; Cui, Y.; Zhang, X.; Liang, J. Numerical simulation of electrohydrodynamic jet and printing micro-structures on flexible substrate. *Microsyst. Technol.* 2020. [CrossRef]
- Zhong, Y.; Dong, X.; Yin, Z.; Fang, H. Theoretical design of inkjet process to improve delivery efficiency. J. Appl. Fluid Mech. 2020, 13, 275–286. [CrossRef]
- Imani Moqadam, S.; M\u00e4dler, L.; Ellendt, N. A high temperature drop-on-demand droplet generator for metallic melts. *Micromachines* 2019, 10, 477. [CrossRef]
- Imani Moqadam, S.; Mädler, L.; Ellendt, N. Microstructure adjustment of spherical micro-samples for high-throughput analysis using a drop-on-demand droplet generator. *Materials* 2019, 12, 3769. [CrossRef]
- Elele, E.; Shen, Y.; Boppana, R.; Afolabi, A.; Bilgili, E.; Khusid, B. Electro-hydrodynamic drop-on-demand printing of aqueous suspensions of drug nanoparticles. *Pharmaceutics* 2020, 12, 1034. [CrossRef]
- Kim, S.H.; Kang, H.; Kang, K.; Lee, S.H.; Cho, K.H.; Hwang, J.Y. Effect of meniscus damping ratio on drop-on-demand electrohydrodynamic jetting. *Appl. Sci.* 2018, *8*, 164. [CrossRef]
- 22. Jiang, H.; Tan, H. One dimensional model for droplet ejection process in inkjet devices. Fluids 2018, 3, 28. [CrossRef]
- Wang, J.; Huang, J.; Zhang, J. A method for calculating the critical velocity of microdroplets produced by circular nozzles. 3D Print. Addit. Manuf. 2020, 7, 338–346. [CrossRef]
- Wang, F.; Bao, W.; Wang, Y.; Wang, X.; Ren, K.; Wang, Z.; Li, J. Ejection state prediction for a pneumatic micro-droplet generator by BP neural networks. J. Adv. Mech. Des. Syst. Manuf. 2020, 14, JAMDSM0001. [CrossRef]
- Pan, Y.; Zeng, L. Simulation and validation of droplet generation process for revealing three design constraints in electrohydrodynamic jet printing. *Micromachines* 2019, 10, 94. [CrossRef]
- Zymelka, D.; Yamashita, T.; Sun, X.; Kobayashi, T. Printed strain sensors based on an intermittent conductive pattern filled with resistive ink droplets. Sensors 2020, 20, 4181. [CrossRef]
- Cui, W.; Zhang, M.; Duan, X.; Pang, W.; Zhang, D.; Zhang, H. Dynamics of electrowetting droplet motion in digital microfluidics systems: From dynamic saturation to device physics. *Micromachines* 2015, 6, 778–789. [CrossRef]
- Liu, Y.; Huang, Y. Theoretical and experimental studies of electrostatic focusing for electrohydrodynamic jet printing. J. Micromech. Microeng. 2019, 29, 065002. [CrossRef]
- Liashenko, I.; Rosell-Llompart, J.; Cabot, A. Ultrafast 3D printing with submicrometer features using electrostatic jet deflection. Nat. Commun. 2020, 11, 753. [CrossRef]
- Ou, C. Aerosol-Jet Printed Nanocomposites for Flexible and Stretchable Thermoelectric Generators. Ph.D. Thesis, University of Cambridge, Cambridge, UK, 2020. [CrossRef]
- Dybowska-Sarapuk, L.; Kielbasinski, K.; Arazna, A.; Futera, K.; Skalski, A.; Janczak, D.; Sloma, M.; Jakubowska, M. Efficient inkjet printing of graphene-based elements: Influence of dispersing agent on ink viscosity. *Nanomaterials* 2018, *8*, 602. [CrossRef] 32. Tortorich, R.P.; Choi, J.-W. Inkjet printing of carbon nanotubes. *Nanomaterials* 2013, *3*, 453–468.
- Wickström, H.; Hilger, E.; Nyman, J.O.; Desai, D.; Şen Karaman, D.; De Beer, T.; Sandler, N.; Rosenholm, J.M. Inkjet printing of drug-loaded mesoporous silica nanoparticles—A platform for drug development. *Molecules* 2017, 22, 2020. [CrossRef]
- Wang, J.; Krishnamoorthy, S.; Song, H.; Ma, C. Mechanical properties of 3D-printed blood vessels. DYNA Ing. Ind. 2020, 9, 541–545. [CrossRef]
- Grottkau, B.E.; Hui, Z.; Pang, Y. A novel 3D bioprinter using direct-volumetric drop-on-demand technology for fabricating micro-tissues and drug-delivery. Int. J. Mol. Sci. 2020, 21, 3482. [CrossRef] [PubMed]
- Son, Y.; Kim, C.; Yang, D.H.; Ahn, D.J. Spreading of an inkjet droplet on a solid surface with a controlled contact angle at low Weber and Reynolds numbers. *Langmuir* 2008, 24, 2900–2907. [CrossRef] [PubMed]
- Bussmann, M.; Chandra, S.; Mostaghimi, J. Modeling the splash of a droplet impacting a solid surface. *Phys. Fluids* 2000, 12, 3121–3132. [CrossRef]
- Driessen, T. Drop Formation from Axi-Symmetric Fluid Jets. Ph.D. Thesis, University of Twente, Enschede, The Netherlands, 2013.
- 39. Olsson, E.; Kreiss, G. A conservative level set method for two phase flow. J. Comput. Phys. 2005, 21, 225–246. [CrossRef]
- 40. Sun, Y.; Beckermann, C. Sharp interface tracking using the phase-field equation. J. Comput. Phys. 2007, 220, 626–653. [CrossRef]
- 41. CFD Module User's Guide; COMSOL: Burlington, MA, USA, 2018.
- Sohr, H. The Navier-Stokes Equations: An Elementary Functional Analytic Approach; Springer Science & Business Media: Berlin, Germany, 2012; p. 367. [CrossRef]
- Derby, B. Inkjet printing of functional and structural materials: Fluid property requirements, feature stability, and resolution. Annu. Rev. Mater. Res. 2010, 40, 395–414. [CrossRef]

- 44. Derby, B. Additive manufacture of ceramics components by inkjet printing. Engineering 2015, 1, 113–123. [CrossRef]
- 45.
- Magdassi, S. The Chemistry of Inkjel Hisky World Scientific: Singapore, 2010. [CrossRef] Wijshoff, H. Drop dynamics in the inkjet printing process. *Curr. Opin. Colloid Interface Sci.* **2018**, *36*, 20–27. [CrossRef] 46.
- 47. Wijshoff, H. Structure- and Fluid-Dynamics in Piezo Inkjet Printheads. Ph.D. Thesis, University of Twente, Enschede, The Netherlands, 2008.
- 48. Dijksman, J. Hydro-acoustics of piezoelectrically driven ink-jet print heads. Flow Turbul. Combust. 1999, 61, 211-237. [CrossRef]
- Systemati, J., Lydro-acoustics of piezoelectricary driven ink-jet print heads. Flow Turbul. Combust. 1999, 61, 211–237. [CrossRef]
  Venezia, E.; Viviani, M.; Presto, S.; Kumar, V.; Tomov, R.I. Inkjet Printing Functionalization of SOFC LSCF Cathodes. Nanomaterials 2019, 9, 654. [CrossRef] 49.

Article 2. Tofan, T. et al. (2021). Investigation of Colour Reproduction on Linen Fabrics when Printing with Mimaki TX400-1800D Inkjet with Pigment TP250 Dyes (https://doi.org/10.3390/coatings11030354)



Article



Investigation of Color Reproduction on Linen Fabrics when Printing with Mimaki TX400-1800D Inkjet with Pigment TP250 Dyes

Tim Tofan¹, Rimantas Stonkus² and Raimondas Jasevičius^{1,3,*}

- ¹ Department of Mechanical and Material Engineering, Vilnius Gediminas Technical University, J. Basanavicius str. 28, 03224 Vilnius, Lithuania; tim.tofan@vilniustech.lt
  - Department of Mechatronics, Robotics and Digital Manufacturing, Vilnius Gediminas Technical University,
- J. Basanavicius str. 28, 03224 Vilnius, Lithuania; rimantas.stonkus@vilniustech.lt
   Institute of Mechanical Science, Vilnius Gediminas Technical University, J. Basanavicius str. 28,
- 03224 Vilnius, Lithuania
- * Correspondence: raimondas.jasevicius@vilniustech.lt

Abstract: The aim of this research is to investigate related effect of dyeability to linen textiles related to different printing parameters. The study investigated the dange in color characteristics when printing on linen fabrics with an inkjet MIMAKI Tx400-1800D printiner with pigmented TP 250 inks. The dependence of color reproduction on linen fabrics on the number of print head passes, number of ink layers to be coated, linen fabric density, and different types of linen fabric was investigated. All this affects the quality of print and its mechanical properties. The change in color characteristics on different types of linen fabrics was determined experimentally. We determine at which print settings the most accurate color reproduction can be achieved on different linen fabrics. The difference between the highest and the lowest possible number of head passages was investigated. The possibilities of reproducing different linen fabric colors were determined.

Keywords: digital inkjet printing; linen fabrics; pigmented inks; color measurement

# 1. Introduction

These days, the demand for organic fabrics is growing rapidly. One of these fabrics is flax, which has been used since ancient times [1]. This natural fiber is back in fashion—with linen clothes, the body breathes and does not overheat, the skin is not irritated and the fabric does not become electric. Linen fabrics are extremely durable [2]. It is also necessary that the technology and inks used for printing also meet the ecological requirements [3].

Over time, many investigations have been performed on textile dyeability, yet there is still much room for process improvement. Many researchers discuss the various types of textile dyeing processes and the fabric changes after dyeing processes [4–7]. Additionally, the newest research of inkjet printing on linen textile were reviewed [3,8–10].

Many studies are related to the investigation of the mechanical properties of textiles [11–13]). There are many investigations on textile mechanical properties affected by the water absorption cycles [14,15]. Temperature also has an effect on mechanical properties on textile fibers [16,17].

One of the possible options is to use a Mimaki printer that is capable of printing with ecological textile inks, which are not harmful to health, do not cause allergies, and are easy to clean and dispose of. The main components of the dyes are pigment, binder and water. As the dyes adhere to the fabric only at high temperatures, they are suitable to print on natural fabrics, such as linen and cotton, which do not contain synthetic materials. One of the main advantages of this printer when printing with textile inks is that it guarantees an environmentally friendly production method [18]. The print is captured immediately after printing without the use of any additional chemistry. No toxic fumes or pollutants



Citation: Tofan, T.; Stonkus, R.; Jasevičius, R. Investigation of Color Reproduction on Linen Fabrics when Printing with Mimaki TX400-1800D Inkjet with Pigment TP250 Dyes. Contings 2021, 21, 354. https:// doi.org/10.3390/contings11000354

Academic Editor: Csaba Balázsi

Received: 27 February 2021 Accepted: 18 March 2021 Published: 19 March 2021

Publisher's Note: MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.



Copyright: © 2021 by the authors. Licensee MDP/, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 40/). are emitted into the environment, so this method of printing on linen is environmentally ecofriendly. In addition, no additional water is used during printing and ink fixing.

Linen comes in different colors: natural, bleached or melange. Melange linen has two different types of threads used to warp and weft directions. In this experiment, the melange linen has bleached thread in a weft direction and natural linen thread in a warp direction. Various units are used to refer to the measurement of a fiber; in this research, for measuring the linear mass density of fibers, tex (g/km) will be used.

Due to the different fabric shades and characteristics, it is more difficult to obtain the corresponding image color on linen fabrics than, for example, on paper [19]. The customer's desire to achieve maximum color matching to the digital standard is limited [20].

When printing, it is very important to choose the most suitable number of ink layers and the number of passes of the print head, as this will significantly differ the print quality and print speed. So, it is very important to know the quantitative and qualitative printing parameters. All this directly affects the company's economic performance. In the case of large orders, the change in the print speed will significantly affect the order execution speed, the number of ink layers, the ink consumption and the mechanical properties of the print.

The aim of this study was to investigate the change in color characteristics by different printing settings when printing on linen fabrics with an inkjet MIMAKI Tx400-1800D printer with pigmented TP 250 inks.

# 2. Methodology of Investigation

Ten linen fabrics from three different types were selected for the study: bleached, melange and natural linen [21]. The linen fabric was made of different threads in warp and weft directions. Warp threads are the yarns held in the loom during the weaving of the fabric and weft threads are the yarn that is passed through the weft yarns during the weaving of the fabric. The fabrics of each type differ in color, fabric density grammage, thickness and density of the threads (Table 1).

To determine the change in color characteristics of the prints on the linen fabrics, depending on the fabric structure and color, in different printing modes, the generated Barbieri Rasterlink color reproduction scales were printed on all fabrics (Figure 1).

e.,												Ĭ				
6						111										
		_														
																L
																┡
	Ц															L
				-			-						-		-	
		-	-	_	_			-	-	-				-	-	Ļ
					_	-	_						_			

Figure 1. Printed Color scale for testing [22].

The Mimaki TX400-1800D printing machine (Mimaki, Tomi-city, Nagano, Japan) has Ricoh GEN4 12 printheads. It can print 7–35 pL, depending on the ink characteristics; suitable ink viscosity should be 10–12 mPa-s. The printhead printing width is 32.4 mm. The number of nozzles is 384 (2 × 192 channels) and the nozzle spacing (within a row) is 0.1693 mm/row. This printhead is suitable to print with different inks, such as UV, solvent, Aqueous, pigment and other.

Textile pigment TP 250 dyes (cyan, magenta, yellow, black, light cyan, light magenta, orange, blue) were used for the experiments. Mimaki textile pigment dyes contain a binder and a binding agent. The colors fix to the fibers by heating the textile to 180 °C. Printed fabrics do not lose their breathability and water absorbability. 69

Investigated: cyan-C, magenta-M, yellow-Y, red-R, green-G, blue-B, black-K, CMY color reproduction only at 100% color tone coverage. The Mimaki TX400 8c Tp250 Liverpool v1 profile provided by the printer manufacturer was used for the research.

After the printing dyes were cured with 180 °C temperature in a roll-to-roll heating machine, measurements of color characteristics were performed with a BARBIERI SpectroPad spectrophotometer [22]. All textile color measurements were made according to BS EN ISO 105-J01:2000 standards 24 h after the textile finally cools down after printing [23].

	Types of	Sample	Grams per Square	Thickness of	Thread (tex)	Threads q-t	y per 10 cm
No.	Linen Fabric	Article	Metre (g/m ² )	Warp	Weft	Warp	Weft
1	Bleached	OBR 840	140	38	38	199	150
2	Bleached	OBR 0114	170	42	42	193	194
3	Bleached	OBR 491	190	56	56	176	142
4	Bleached	OBR 1542	245	87	87	156	118
5	Melange	OBR 052	150	38	38	199	182
6	Melange	OBR 831	190	56	56	176	142
7	Melange	OBR 482	280	110	110	142	102
8	Natural	OBR 166	125	30	30	226	174
9	Natural	OBR 051	150	38	38	199	184
10	Natural	OBR 1041	240	83	83	160	112

Table 1.	Linen fa	abrics s	selected	for	investigation.
the second second second					

To change the characteristics of colors CIE  $L^*a^*b^*$  values, and to determine the difference in color reproduction,  $\Delta E$  as the digital values of CMYKRGB color  $L^*a^*b^*$  were taken as reference in Table 2.

Color	$L^*$	a*	<i>b</i> *
С	62	-44	-50
М	52	81	-7
Υ	95	-6	95
R	52	74	54
G	57	-74	30
В	25	25	-55
K	12	2	0
CMY	23	1	-2

Table 2. Digital reference values of CMYKRGB colors L*a*b*.

The color difference is calculated according to the following equation:

$$\Delta E = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2}$$
(1)

where *L** is lightness value (0 for black and 100 for white); *a**—position between red and green value (negative–green, positive–red); *b**—position between blue and yellow value (negative–blue, positive–yellow) [24], Figure 2.



Figure 2. CIE L*a*b* color system.

Difference in  $\Delta L^*$ ,  $\Delta a^*$  and  $\Delta b^*$  was calculated using the following equation:

$$\Delta L^* = \sqrt{\left(L_1^* - L_2^*\right)^2}$$
⁽²⁾

$$\Delta a^* = \sqrt{\left(a_1^* - a_2^*\right)^2}$$
(3)

$$\Delta b^* = \sqrt{\left(b_1^* - b_2^*\right)^2} \tag{4}$$

where, depending on the comparison,  $L_1^*$ ,  $a_1^*$ ,  $b_1^*$  represents primary fabric value and  $L_2^*$ ,  $a_2^*$ ,  $b_2^*$  represents the second coat of dyes or dyed fabric value. Mean value was calculated by using the following equation [25]:

$$\overline{x} = \frac{\sum x}{n}$$
(5)

Standard deviations were determined using the following equation [25]:

$$s = \sqrt{\frac{\sum (x - \overline{x})^2}{n - 1}};$$
(6)

Standard error of the mean was calculated using the following equation [25]:

$$SE_{\overline{x}} = \frac{s}{\sqrt{n}}$$
(7)

The description of the differences of the  $\Delta E$  value is presented in Table 3.

Table 3.  $\Delta E$  values and their corresponding color differences [26].

$\Delta E$	Meaning		
0–1	Normally invisible difference		
1–2	Very small difference, only obvious to trained eye		
2-3.5	Medium difference, also obvious to untrained eye		
3.5–5	Obvious difference		
>5	Very obvious difference		

# 3. Results of Research on Color Properties

The shades of linen fabrics are very different (Figure 3). This has a fairly significant effect on the reproduction of the print colors by coating the fabric with different colors of paint. Linen textile woven fabric structure is illustrated in Figure 4. Therefore, it is worth

comparing how the shade of linen differs from the reference white  $(L^* = 100, a^* = 0, b^* = 0)$  color (Figure 5).

1 2 3 4 9 2 9 9

Figure 3. Linen fabrics selected for investigation.



Figure 4. Woven fabric structure characteristics.



**Figure 5.** Color difference  $\Delta E$  of linen fabric surface without print with the reference white color.

Comparing the color characteristics of linen fabrics with the reference white color, certain regularities were found (Figure 5). The lowest  $\Delta E$  values are obtained by comparing bleached linen fabrics and the highest are obtained by comparing natural linen fabrics. The shade of linen fabric surfaces has a significant effect on the reproduction of print colors.

Bleached linen OBR 840 was selected to determine the change in color characteristics depending on the number of paint layers and the number of passages. 1 to 5 layers of paint coating were printed on this fabric in different 4 and 16 pass modes. From the three possible pass modes (4, 8, and 16), the minimum and maximum numbers of passes were deliberately chosen for better difference detection. The results of color reproduction are presented in Figures 6 and 7.



Figure 6. Different colors changes of  $\Delta E$  value depending on dye layers when printing in 4 passes.



Figure 7. Different colors changes of  $\Delta E$  value depending on dye layers when printing in 16 passes.

There is a significant difference between layers 1 and 5 (from 3% to 55%), while there is a relatively small difference between layers 3 and 4 ( $\leq$ 5%), and between layers 4 and 5 ( $\leq$ 4%).

Comparing the 4 and 16 pass print modes, the same patterns of  $\Delta E$  color characteristic changes remain. Comparing the difference in color reproduction from the number of passages, it was observed that the color difference is insignificant ( $\Delta E \leq 3$ ), except for the green color.

When printing in the selected four-pass printing mode, it was analyzed and found to have the greatest influence on the change in color characteristics of  $\Delta E$ . The change in  $L^*a^*b^*$  values at different paint layers is shown in Figures 8–10.

It was observed that the value of lightness  $(L^*)$  changes the most in the ovverall color (Figure 8). It was found that, by changing the number of layers of ink to be coated, the coan color lightness changes the most, changing from 48.99 for one-layer printing to 68.29 for five-layer printing; the brightness of green color lightness also differs quite strongly; the color darkens by as much as 26.5% when printed with five layers compared to one layer. Yellow is the least sensitive to the change in the number of paint layers (change is 6%). The most sensitive to the change in the number of paint layers are cyan (28% change) 73

and green (26% change). Comparing the change in brightness of all the colors lightness analyzed in the diagram, it is clear that it changes the most when printing with two and three layers of ink, while when printing with an additional four and five layers, the change in lightness is insignificant (average change, 2–4%, respectively). It can also be seen that the differences in blue, black and CMY colors between layers 2, 3, 4 and 5 are very slight, and the difference between magenta, red and green is very small between layers 3, 4, 5.

It was observed that the values of a * have practically no effect on determining the color difference of  $\Delta E$  (Figure 9). The difference between the first and fifth coats of paint is  $\leq$ 3, less than 5%.

When analyzing the  $b^*$  values (Figure 10), it was observed that the yellow color value changes the most: the difference between the first layer was 50.60, and for the five layers it was 76.84, 34%. This may have been influenced by the color of the fabric, as the coating of one coat of paint is not sufficient to represent the color. Other values are almost the same; the difference between one and five coats of paint does not exceed 3.



Figure 8. Different colors changes of  $\Delta L^*$  value depending on ink layer when printing in 4 passes.



**Figure 9.** Different colors changes of  $\Delta a^*$  value depending on ink layer when printing in 4 passes.



Figure 10. Different colors changes of  $\Delta b^*$  depending on ink layer when printing in 4 passes.

Therefore, it can be concluded that most of the  $\Delta E$  color difference is due to lightness (*L**). It also shows that the difference between paint layers 3, 4 and 5 is not very significant and practically invisible. This again shows that printing with more than three coats of toner is not relevant.

The change in color characteristics of prints on linen fabrics with four passes and coating with two coats of dye was further analyzed. This is the most commonly used print setting, at which the printer reproduces colors well and prints fast enough.

All three different types (bleached, melange, natural) of linen fabrics compared to two coats of paint are compared. The results of the study are presented in Figures 11–13.

In the study of the application of the two dye layers on bleached fabrics (Figure 11), it was observed that the thickest OBR 1542 fabric has the smallest change in  $\Delta E$  color characteristics. It was also observed that the thinnest linen OBR 840 reproduces red, green, blue, black and brown. Additionally, when printing with two coatings, the color reproduction is very good on OBR 0114, and the change in color characteristics is very similar to OBR 1542. The color characteristics  $\Delta E$  values were found to be very similar for all bleached fabrics; the difference between them is not large ( $\leq 4$ ).



Figure 11. Different colors changes of  $\Delta E$  on bleached linen fabrics when printing in 4 passes with 2 ink layers.

```
Coatings 2021, 11, 354
```





Figure 12. Different colors changes of  $\Delta E$  on melange linen fabrics when printing in 4 passes with 2 ink layers.



**Figure 13.** Different colors changes of  $\Delta E$  on natural linen fabrics when printing in 4 passes with 2 ink layers.

When studying the reproduction of color characteristics on melange fabrics (Figure 12) in the presence of two layers of paint, it was observed that the colors are best reproduced on the thickest linen fabric OBR 482. Slightly worse colors are reproduced on the thinnest OBR 052 fabric. The color difference between different tissues is not large ( $\leq$ 4). The color yellow is reproduced practically equally on all fabrics.

In the study of color reproduction on natural fabrics in the presence of two coats of paint (Figure 13), much larger changes in color characteristics were observed between different fabrics. The largest difference was observed in yellow. It was also observed that the colors are much better reproduced on the OBR 051 fabric, slightly worse on the OBR 106 fabric, and the largest difference was obtained on the thickest linen fabric of the OBR 1041, except for the dark colors.

Figure 14 shows the ranges of reproducible print colors on different linen fabrics in the  $a^*$  and  $b^*$  plane without evaluating the color lightness  $L^*$ .





Figure 14. Color reproduction on different linen fabric in a*b* when printing in 4 passes with 2 ink layers.

The range of digital reference colors (black line) show that it is possible to reach the limits of maximum rich colors and the range of colors is reproduced in the print on paper (white line). Limited color saturation reproduction on all types of linen fabrics is clearly visible. The most affected are cyan and blue (B) color saturation. Color reproduction in prints on bleached linen fabrics is closest to color reproduction on paper. There is a very limited reproduction of color saturation in prints on natural linen fabrics. This is due to the fact that the fabrics of natural linen are quite dark, and the color of the dye is "suppressed".

Other studies on inkjet printing can also be acknowledged. There are many different articles with experiments measuring textile color using digital inkjet printing technology [27]. Textile digital inkjet printing allows for better quality control, faster prepress, reduction in the use of material and better repeatable color prints on textile substrates [27]. Experiments with a digital inkjet printer on different pretreated textiles were also performed here to find the best method of pretreatment. It was emphasized that simulated colors should be compared with original colors under daylight illuminance. To obtain more accurate results, we took into account this assumption. As well as this, a dye's physical properties have a very important effect on the print quality [28]. Here, the inkjet printing of cotton with natural dyes with different physical and rheological properties (pH, conductivity, surface tension, and viscosity) of the inks was investigated. Parameters were measured over a period of 90 days for the evaluation of ink stability and suitability in the digital linkjet printing. We took this into account for the evaluation of ink stability and suitability.

It could also be mentioned that, in our work, textile pretreatment before printing and different dyes characteristics are investigated. We looked at the research from a different angle, and our work is focused on the investigation of different printer parameters (number of printing drops and the thickness of the ink layer) to affect color reproduction on different natural linen textiles.

# 4. Conclusions

 Color reproduction when printing both 4 and 16 passes is very similar. The difference between the values of ΔE of the change in color characteristics between the different passages does not exceed 3;

11 of 12

- The number of applied paint layers has the greatest influence on the change in lightness (L*);
- 3. The best color reproduction is achieved with two coats of dye;
- 4. When printing with 1 layer of ink, the color reproduction is the worst;
- Printing in different passes setting does not increase color reproduction, but printing in a smaller passes number increases printing speed a lot;
- At layers 4–5 of paint, the change of color characteristic ΔE increases and colors are reproduced less effectively than at dye layers 2–3;
- In prints on bleached linen fabrics, the colors are best reproduced on the thickest linen fabric and the worst on the thinnest;
- On melange linen fabrics, the worst colors are reproduced on the thinnest linen fabric and the color change in its surface without printing is the largest;
- In the study of natural and melange linen fabrics, the color change in the surface without pressure has a significant influence on color reproduction.

Author Contributions: T.T., R.S., and R.J. conceived the presented idea, processed and analyzed the data and wrote the paper. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by Vilnius Gediminas Technical University.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Informed consent was obtained from all subjects involved in the study.

Data Availability Statement: The data presented in this study are available on request from the corresponding author. The data are not publicly available due to privacy.

Conflicts of Interest: The authors declare no conflict of interest.

# References

- Strand, E.A.; Frei, K.M.; Gleba, M.; Mannering, U.; Nosch, M.L.; Skals, I. Old textiles-new possibilities. Eur. J. Archaeol. 2010, 13, 149–173. [CrossRef]
- Buchert, J.; Pere, J.; Johansson, L.S.; Campbell, J.M. Analysis of the surface chemistry of linen and cotton fabrics. Text. Res. J. 2001, 71, 626–629. [CrossRef]
- El-Hennawi, H.M.; Shahin, A.A.; Rekaby, M.; Ragheb, A.A. Ink jet printing of bio-treated linen, polyester fabrics and their blend. Carbohydr. Polym. 2015, 118, 235–241. [CrossRef] [PubMed]
- Baaka, N.; Ticha, M.B.; Haddar, W.; Amorim, M.T.P.; Mhenni, M.F. Upgrading of UV protection properties of several textile fabrics by their dyeing with grape pomace colorants. *Fibers Polym.* 2018, 19, 307–312. [CrossRef]
- Bernava, A.; Reihmane, S. Influence of modification methods on colour properties of a linen fabric dyed with direct dyes. Proc. Est. Acad. Sci. 2018, 67, 131–137. [CrossRef]
- Burkinshaw, S.M.; Salihu, G. The role of auxiliaries in the immersion dyeing of textile fibres: Part 1 an overview. Dye. Pigment. 2019, 161, 519–530. [CrossRef]
- Jabar, J.M.; Ogunmokun, A.I.; Taleat, T.A.A. Color and fastness properties of mordanted Bridelia ferruginea B dyed cellulosic fabric. Fash. Text. 2020, 7, 1–13. [CrossRef]
- Özgüney, A.T.; Bozacı, E.; Demir, A.; Özdoğan, E. Inkjet printing of linen fabrics pretreated with atmospheric plasma and various print pastes. AATCC J. Res. 2017, 4, 22–27. [CrossRef]
- Rekaby, M.; Abd-El Thalouth, J.I.; Abd El-Salam, S.H. Improving reactive ink jet printing via cationization of cellulosic linen fabric. Carbohydr. Polym. 2013, 98, 1371–1376. [CrossRef]
- Liang, Y.; Liu, X.; Fang, K.; An, F.; Li, C.; Liu, H.; Qiao, X.; Zhang, S. Construction of new surface on linen fabric by hydroxyethyl cellulose for improving inkjet printing performance of reactive dyes. Prog. Org. Cont. 2021, 154, 106179. [CrossRef]
- Tan, P.; Tong, L.; Steven, G.P. Modelling for predicting the mechanical properties of textile composites—A review. Compos. Part A Appl. Sci. Manuf. 1997, 28, 903–922. [CrossRef]
- Kan, C.W.; Yuen, C.W.M.; Lam, Y.L. Effect of enzyme treatment and dyeing on the mechanical properties of linen. *Coloration Technol.* 2009, 125, 269–276. [CrossRef]
- Miller, S.A. Natural fiber textile reinforced bio-based composites: Mechanical properties, creep, and environmental impacts. J. Clean. Prod. 2018, 198, 612–623. [CrossRef]
- Kim, H.J. Effect of water absorption fatigue on mechanical properties of sisal textile-reinforced composites. Int. J. Fatigue 2006, 28, 1307–1314. [CrossRef]

- Ding, Y.; Shamey, R.; Chapman, L.P.; Freeman, H.S. Pretreatment effects on pigment-based textile inkjet printing-colour gamut and crockfastness properties. *Coloration Technol.* 2019, 135, 77–86. [CrossRef]
- Hufenbach, W.; Gude, M.; Böhm, R.; Zscheyge, M. The effect of temperature on mechanical properties and failure behaviour of hybrid yarn textile-reinforced thermoplastics. *Mater. Des.* 2011, 32, 4278–4288. [CrossRef]
- Nguyen, T.H.; Vu, X.H.; Larbi, A.S.; Ferrier, E. Experimental study of the effect of simultaneous mechanical and high-temperature loadings on the behaviour of textile-reinforced concrete (TRC). Constr. Build. Mater. 2016, 125, 253–270. [CrossRef]
- 18. Sharma, A. Eco-friendly Textiles: A boost to sustainability. Asian J. Home Sci. 2013, 8, 768-771.
- 19. Javoršek, D.; Javoršek, A. Colour management in digital textile printing. Coloration Technol. 2011, 127, 235-239. [CrossRef]
- 20. Osaki, K. High quality color reproduction on jacquard silk textile from digital color images. AUTEX Res. J. 2003, 3, 173-179.
- 21. ORSHA Lnen Mill. Available online: https://linenmill.by/product-category/tkani/bazovye-tkani/ (accessed on 16 January 2021).
- BARBIERI Electronic. Portable Spectrophotometer for Professional Digital Printing. Available online: http://www.barbierielectronic. com/en/digital-imaging/1-0.html (accessed on 16 January 2021).
- British Standards Document BS EN ISO 105-J01 Textiles. Test for Colour Fastness. General Principles for Measurement of Surface Colour. Available online: https://doi.org/10.3403/02058086U (accessed on 28 February 2021).
- 24. Sidaravičius, D.J. Densitometric and Colorimetric Control of Printing Materials and Prints; Technika: Vilnius, Lithuania, 2012.
- Crowle, G. Effects of Cotton Structure and Surface Interactions on Dye Uptake. Ph.D. Thesis, Deakin University, Geelong, Australia, 2017.
- Electronics for Imaging, Inc. Delta E, Delta H, Delta T: What Does It Mean? Available online: https://help.efi.com/fieryxf/ KnowledgeBase/color/Delta%20E_H_T.pdf (accessed on 26 February 2021).
- Javoršek, D.; Weingerl, P.; Starešinič, M. Digital Textile Printing Using Color Management. In Textiles: History, Properties and Performance and Applications; Mondal, M.I.H., Ed.; Nova Science Publishers, Inc.: New York, NY, USA, 2014; pp. 53–80.
- Savvidis, G.; Karanikas, E.; Nikolaidis, N.; Eleftheriadis, I.; Tsatsaroni, E. Ink-jet printing of cotton with natural dyes. *Coloration Technol.* 2014, 130, 200–204. [CrossRef]

# Article 3. Tofan, T. et al. (2021). Modelling of the Motion and Interaction of a Droplet of an Inkjet Printing Process with Physically Treated Polymers Substrates (https://doi.org/10.3390/app112311465)



Article



Modelling of the Motion and Interaction of a Droplet of an Inkjet Printing Process with Physically Treated Polymers Substrates

Tim Tofan¹ and Raimondas Jasevičius^{1,2,*}

- ¹ Department of Mechanics and Materials Engineering, Vilnius Gediminas Technical University,
- J. Basanavicius Street 28, 03224 Vilnius, Lithuania; tim.tofan@vilniustech.lt
- Institute of Mechanical Science, Vilnius Gediminas Technical University, J. Basanavicius Street 28, 03224 Vilnius, Lithuania
- * Correspondence: raimondas.jasevicius@vilniustech.lt

Abstract: This study examines the effect of energetic surface treatment on the adhesion strength of high-density polyethylene (HDPE), polypropylene (PP) and polyethylene terephthalate (PET) substrates. The purpose of this work is to determine the surface wetability of polymers suitable for food contact. These plastics have been treated with various pre-treatment methods to improve surface tension and good adhesion for inkjet printing and avoid any visual changes. It is important to determine the adhesion of the ink to the polymer surface to improve post-consumer recycling. Digital inks have been tested on various treated plastics to analyse coating properties and adhesion forces in accordance with DIN ISO 2409 standards. The impact of the inkjet droplet on the treated and non-treated surface was also investigated using the COMSOL computer simulation software.

Keywords: drop-on-demand inkjet; surface energy; droplet; measurement of droplet contact angle



Citation: Tofan, T.; Jasevičius, R. Modelling of the Motion and Interaction of a Droplet of an Inkjet Printing Process with Physically Tracted Polymers Substrates. *Appl.* Sci. 2021, 11, 11465. https://doi.org/ 10.3390/app112311465

Academic Editors: Fonseca Elza and Hernâni Miguel Reis Lopes

Received: 11 November 2021 Accepted: 1 December 2021 Published: 3 December 2021

Publisher's Note: MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affillations.



Copyright: © 2021 by the authors. Licensee MDP/, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 40/). 1. Introduction

The growth of the inkjet printing market poses many different challenges. Printing on various polymers can cause adhesion problems due to their low surface energy and adhesion strength. To solve these issues, there are many treatments available to improve surface energy and enhance adhesion. They can all be divided into three main types: mechanical treatment, affecting mainly the surface topography (e.g., grit blasting, peel ply); chemical treatment using chemicals (e.g., solvent cleaning, primers, ultrasonic cleaning); and energetic treatments aimed primarily at surface free energy (e.g., plasma, corona, flame, UV/ozone), according to Petrie and Edward [1].

It was observed that UV/ozone, corona discharge and low-pressure plasma treatment methods did not significantly change the surface roughness of many polymers, Oosterom et al. [2]. It is very important to preserve the previous view of the surface for printing on them. A solid surface is defined as a material that is rigid and resistant to stress. In inkjet printing, the roughness of the substrate significantly affects the diameter of the printed droplets, according to Novaković et al. [3]. The influence of the surface roughness factor impact was considered by Lim et al. [4]. A solid surface can be characterized by its surface free energy and surface roughness which affect the adhesion strength.

One of the main surfaces treatment methods for polymers is an economically affordable flame. Changes caused by the surface of the polypropylene homopolymer after flame treatment were investigated by Sutherland et al. [5]. Adhesion between flame treated polypropylene and paint film was assessed using a composite butt test and the measured bond strength was found to be much higher than that obtained by solvent wiping or chlorinated polyolefin primers. The most important flame treatment properties can be profitably used to improve the wettability and adhesion properties of polyolefin surfaces without altering the optical internal parts of the surface, Farris et al. [6]. Brewis and Briggs observed the adhesion of polyethylene and polypropylene [7]. They observed that these polymers with poor adhesive properties have a complex subject that requires an understanding of the excellent performance after some pre-treatment and the changes that result from this pre-treatment. Another example is atmospheric plasma treatment, which is relatively inexpensive and easier to integrate and operate on production lines. Plasma is produced by excitation of gas with electrical energy to make it highly reactive. This method is very effective in activating polymeric surfaces, Hegemann et al. [8], Sandanuwan et al. [9]. The effect of the treatment can last from 48 h to 7 days, as stated by Sundrival et al. [10]. Atmospheric plasma treatment improved adhesion strength in different ways. The results show that in order to fully understand the mechanism of activation of this atmospheric pressure plasma flow system, it is necessary to take into account not only the reactions of the plasma surface, but also the post-plasma processes with the ambient gas atmosphere, see Lommatzsch et al. [11] and Sundriyal et al. [12]. A vacuum plasma system with different frequency power was investigated to modify polyimide substrates to improve the surface treatment efficiency with respect to adhesive strength, Lee et al. [13].

Previous studies have shown that various pre-treatment methods have improved the adhesion strength of polyethylene (PE), polyethylene terephthalate (PET), polypropylene (PP), and high-density polyethylene (HDPE). In this work, research is related to increasing surface energy for inkjet printing. However, the effectiveness of the presented experiment still depends on the properties of materials, time, ink parameters, processing, and research conditions.

# 2. Problem Formulation

There are many studies available that focus on surface preparation, treatments, and other parameters modifications. In this study, we will analyse the adhesion problems of ink printing on different types of polymers. Energy treatment methods such as corona discharge, plasma discharge or gas flame treatment at atmospheric pressure were also reviewed.

These surface treatments remove organic contaminants, trigger oxidation processes, and increase the polarity and wettability of polymeric surfaces to increase adhesion to printing inks, Ebnesajjad and Ebnesajjad [14]. Here, a plasma is created by exciting a gas with electrical energy to make it highly reactive. The quality of inkjet printing mainly depends on the control of the printheads, the behaviour of the droplets in flight and the distribution of droplets on the target surface, as well as its wettability, Liu [15].

Inkjet printing technology is applied to the fabrication of thick electro-adhesive devices with a main polyimide dielectric layer, Berdozzi et al. [16]. Inkjet printing is becoming one of the most efficient, inexpensive, and flexible techniques to fabricate thin-film devices for flexible electronics applications, such as micro-supercapacitors, Sundriyal et al. [17,18]. Mention may be made of the study of polymers in inkjet printing from various perspectives. The advantage of inkjet printing in achieving transverse conductivity in Poly(3,4ethylenedioxythiophene)/Poly(styrenesulfonate) (PEDOT/PSS) compared to spin coating is given by Wilson et al. [19]. It was observed (Wilson et al. [20]) that both in-plane conductivity and contact resistance increased with the concentration of dimethyl sulfoxide (DMSO) in the printing ink. It has been demonstrated that inkjet printed thin films offer comparable and even better surface and electrical properties to layers deposited by the more commonly used spin coating technique, Wilson et al. [21].

In the analysis of different polymers, surface treatment for ink counting is very important to improve the sufficient surface adhesion strength. It should be noted that polymers generally have a very low surface energy (20-22 mN/m). For various printing methods, it is necessary to achieve a stable surface energy of 45 mN/m to 71 mN/m on the surface, Eckert [22]. A typical method for measuring the static and dynamic surface tension of inks, as well as the properties of the substrate surface, is the analysis of the contact (wetting) angle of droplet.

To test the adhesion of the printed layer, it is best to use ASTM D3359 standard Cross-Cut test methods [23] to measure the adhesion using tape. The printed substrate samples are scratched with a special cutting tool and a special certified tape is used. These test methods cover procedures for evaluating the adhesion of coating films to metallic substrates by applying and removing pressure-sensitive tape on cuts made in the film. Numerical simulations with COMSOL can help you see the shape of the droplets on a variety of treated and non-treated surfaces.

There are many problems with inkjet droplet adhesion with HDPE, PP and PET polymers. These polymers have very low surface free energy values, so it is important to increase this to an acceptable level for coating. It is important to keep the visual characteristics of the surface and the acceptability of food contact. This study is designed to analyse various surface treatment methods for inkjet printing.

The aim of the research is to analyse different polymer surface treatments and their influence on surfaces, investigate how surface energy and surface tension are measured and describe how the surface of the polymers must be treated to ensure that ink is properly fixed. The focus of the study is to demonstrate the differences in the interaction of inkjet droplets with the surface.

# 3. Methodology

The terms of surface energy (SE) and surface tension (ST) correspond physically. The SE is commonly used to describe energy for solid surfaces and ST for liquid surfaces. ST is defined as the work required to increase the surface area isothermally and reversibly by a unit amount, Ebnesajiad and Ebnesajiad [14]. However, the ST of solids is also incorrectly indicated. SE uses the unit of energy per area in mJ/m² (millijoule per square meter), with the equivalent unit mN/m (millinewton per meter) commonly used for ST, Ebnesajiad and Ebnesajiad [14]. The formula symbol is  $\sigma$  (less often  $\gamma$ ), which, respectively, represents the tension at the solid/air, liquid/air, and solid/liquid interfaces.

There are various ways to analyse surface energy: Wu's equation, Owens–Wendt, Van Oss–Good–Chaudhury equation, Lifshitz–van der Waals and Lewis acid-base, Girifalco [24]. As a first step, we use Young's equation, which is the standard for simulation in COMSOL:

$$\sigma_s = \sigma_{sl} + \sigma_l \cdot \cos\theta \tag{1}$$

Here,  $\sigma_s$  is the surface free energy of a solid,  $\sigma_l$  is the surface free energy of a liquid,  $\sigma_{sl}$  is the interfacial free energy between a solid and a liquid,  $\theta$  is the contact angle of a solid and a liquid. The following illustration (Figure 1) shows the droplet height *h*, droplet diameter *d*, and the contact (wetting) angle resulting from the equilibrium of forces during solid wetting according to Young's Equation (1) in the stable phase:



Figure 1. A drop on a surface with an indication of the contact angle and surface tension for three media, respectively (according to Kinlokh [25]).

In a simple system such as that shown in Figure 1, the adhesion strength can be calculated through the work of adhesion  $W_a$ , Dupré [26]:

$$W_a = \sigma_s + \sigma_l - \sigma_{sl} \tag{2}$$

The interaction of the droplet with the surface was simulated using the COMSOL Multiphysics software [27]. The droplet height and base were taken from the programming results, and the contact angle was obtained from Bonadiman et al. [28]:

$$\theta = 2 \tan^{-1} \left(\frac{2h}{d}\right) \tag{3}$$

where *h* is the droplet height, *d* is the droplet diameter, and  $\theta$  is the contact (wetting) angle. Estimation of the surface energy of a solid by the contact angle (Good and Girifalco [29])

$$\cos\theta = \frac{\gamma_s - \gamma_{sl}}{\gamma_l}.\tag{4}$$

For displaying the interface and transferring ink in air on a smooth surface, we considered the Level Set method. Typically, inkjet ink is a Newtonian fluid that does not change viscosity when force is applied. The interface between the inkjet droplet and another substance is described by  $\phi$ , where  $\phi = 1$  represents ink and  $\phi = 0$  is another substance. At the interface conversion level,  $\phi$  varies from 0 to 1. The conversion layer goes along with the jet droplet velocity u during interaction. The following COMSOL Equation (5) describes the convection of a reinitialized level set function, taking into account the interface between the two phases, ink and air:

$$\frac{\partial \Phi}{\partial t} + \mathbf{u} \cdot \nabla \Phi + \gamma \left[ \left( \nabla \left( \Phi (1 - \Phi) \frac{\nabla \Phi}{|\nabla \Phi|} \right) \right) - \varepsilon \nabla \cdot \nabla \Phi \right] = 0 \tag{5}$$

where  $\varepsilon$  is the thickness of the transition layer, and it can be defined as  $\varepsilon = hc/2$  (hc is the representative mesh size in the area passed by the falling droplet in the simulation, set to  $hc = 2.5 \ \mu m$ ).  $\gamma$  is the amount of reinitialization or stabilization of the level set function and the current simulation is set to  $\gamma = 10 \ m/s$ .

Density and dynamic viscosity depend on the interfacial coefficient  $\phi$ :

$$\rho = \rho_1 + (\rho_2 - \rho_1)\phi \tag{6}$$

where,  $\rho_1$ ,  $\rho_2$  denote air and ink density, respectively. For numerical simulation, the dynamic viscosity (shear viscosity)  $\mu$  describes the relationship between the shear rate and shear stresses in a fluid:

$$\mu = \mu_1 + (\mu_2 - \mu_1)\phi \tag{7}$$

here  $\mu$ ,  $\mu_2$  are the viscosity of air and ink, respectively. The movement of ink, including the conservation of mass, can be described (Sohr [30]):

$$(\nabla \cdot \mathbf{u}) = \mathbf{0},\tag{8}$$

$$\left(\frac{\partial \boldsymbol{u}}{\partial t} + \boldsymbol{u} \cdot \nabla \boldsymbol{u} - \boldsymbol{g}\right) - \nabla \cdot \left(\boldsymbol{\mu} \left(\nabla \boldsymbol{u} + \nabla \boldsymbol{u}^{\mathsf{T}}\right)\right) + \nabla \boldsymbol{p} = \mathbf{F}_{\mathsf{st}} \tag{9}$$

here  $F_{st}$  is the surface tension force, *p* is the pressure,  $\rho$  is the density,  $\mu$  is the dynamic viscosity, and  $\mu$  is the droplet velocity. The force can be calculated taking into account the surface tension coefficient and the curvature of the interface.

1

$$\mathbf{F}_{st} = \sigma \delta \kappa \mathbf{n},$$
 (10)

where  $\sigma$  is the surface tension coefficient, which varies depending on the curve, and  $\delta$  is the Dirac delta function, which is nonzero only at the liquid interface, and n is the unit vector in the normal direction:

$$n = \frac{\nabla \Phi}{|\nabla \Phi|}$$
(11)

5 of 11

The delta function is approximated by the following equation:

$$\delta = 6|\phi(1-\phi)||\nabla\phi| \tag{12}$$

The adaptive mesh refinement functionality for simulation inkjet nozzles in COMSOL Multiphysics [27] is used to locally refine the mesh around the ink-air interface. For numerical simulation in COMSOL, the inkjet ink parameters are presented in Table 1. These are the optimal inkjet ink settings.

Table 1. Inkjet inks parameters.

Substance	ρ kg/m ³	$\mu \text{ mN} \cdot \text{s/m}^2$	σmN/m	Oh
Ink	1080	10	72	0.23

Here, in Table 1, the additional dimensionless parameter Oh represents the printability of the ink.

# 3.1. Characterization of the Surface of Polymers

During a physical experiment, surface energy was investigated using formamide test inks (Series A) called "Surface Test Inks C28 to C72" from Plasmatreat [31]. These inks were used to determine the surface energy of plasma-treated and non-treated PP, HDPE, and PET surfaces. To determine the surface energy of the surface, we start with the lowest ST ink C28 and move on to the higher C72 test ink, which was quickly applied on the surface using the integrated bottle brush. If the edges of the brush stroke remained stable for 2 s, the surface was easily wetted. The surface energy of the substrate is then at least equal to the value of the test ink. Thus, we gradually approached the values of the surface energy of the substrates. These test inks were used to determine changes in surface energy. The experiments were carried out at a room temperature of 230 °C, when the humidity was 55%.

These polymers have been used in the food industry and have different glossy and matte surface finishes. It is important to emphasize that on a matte surface, the surface tension parameters should be higher. These parameters were taken from the mould manufacturer's measurement parameters when measuring the surface roughness of the final product. All samples were taken in black colour, which is more commonly found in the food industry. Samples are 100% virgin, without any post consumed material. For the test, we used standard injected plates with different mould surface roughness. The results of surface measurements are presented in Table 2.

Table 2. Surface characteristics of PP, HDPE, PET materials before treatment.

Material	Manufacturer	Material Thickness (mm)	Surface Roughness (µm)	Surface Tension (mN/m)
1. PP (Matte Surface)	MOPLEN HP500N	1 mm	2.8	32
2. PP (Glossy Surface)	MOPLEN HP500N	1 mm	0.05	28
3. HDPE (Matte Surface)	TIPELIN 3110J	1 mm	2.5	32
4. PET (Glossy surface)	NEOPET80	1 mm	0.05	38

3.2. Cross-Cut and Tape Test

The Cross-Cut and Tape test (DIN ISO 2409) are performed direct after printing and after 24 h. The parts are placed on a conveyor, which transfers them in set steps through the device (process parameters are summarized in Table 3 below). Generally, the Cross-Cut quality classification values such as GT0 and GT1 are acceptable results according to DIN ISO 2409. If the results are lower than the GT2 value, the quality is unacceptable.

Cross-Cut result

Percent area removed

Class

	Table 3. Classification	n of Cross-Cut and ta	ape test (DIN ISO 24	09) results.	
sification Value	GT0	GT1	GT2	GT3	GT4
	+++++++-		-UTUI-		-1.11

First, the samples were printed without any pre-treatment. Second, the polymer surfaces were cleaned with IPA (isopropyl alcohol) and printed immediately after the surface dried. This shows pretty good adhesion results exactly after printing but lost all the benefits after 24 h. The third method of treatment was treatment with atmospheric pressure plasma. It was applied using the RD2005 air plasma system (Figure 2) manufactured by Plasmatreat [31].

15-35

Figure 2. Activation with the RD2005 open air plasma.

The process was carried out in air at an inlet pressure of 4 bar, a flow rate of 51 lpm and a power of 600 W per nozzle. To minimize these plasma-induced degradation and aging effects, the treatment speed and the distance between the sample and the nozzle have been carefully adjusted. Preliminary tests showed that no significant physical changes on the surface were observed at a distance of 9 mm to the surface and a speed of 5 mm/s. The samples were printed using a Mimaki UJF-3042 printing machine with LUS120 LED-UV lamp curing ink. The samples were printed immediately after plasma treatment to minimize the aging effect (Figure 3). For good adhesion to UV cured ink, the plasma treatment parameters are selected according to the surface tension.

Figure 3. Inkjet Printing process of the treated/non-treated parts.

6 of 11

35-650

After printing, a Cross-Cut and Tape test (DIN ISO 2409) was performed (Figure 4). It was made direct after printing and after 24 h.



Figure 4. Cross-Cut tool illustration.

# 4. Experimental Results and Discussion

The results of the Cross-Cut and Tape test (DIN ISO 2409) are given in Table 4. The change in free energy of the surface was measured with Plasmatreat test inks. Contact angles were simulated using the COMSOL Multiphysics simulation program.

Test No.	Sample No.	SE (mN/m)	Contact Angle (°)	Cross-Cut Test Direct after Printing	Cross-Cut Test after 24 h.	Samples Treatment Method
	1	32	82.3	GT1	GT4	-
	2	28	83.8	GT3	GT4	<u></u>
1	3	32	78.1	GT4	GT4	-
	4	38	79.7	GT3	GT4	-
	1	32	-	GT1	GT4	Cleaned with IPA
2	2	28	(23)	GT2	GT4	Cleaned with IPA
2	3	32	-	GT2	GT4	Cleaned with IPA
	4	38	127	GT2	GT4	Cleaned with IPA
	1	70	52.4	GT0	GT0	Plasma treated
2	2	58	55.3	GT0	GT0	Plasma treated
3	3	72	48.7	GT0	GT0	Plasma treated
	4	72	49.1	GT0	GT0	Plasma treated

Table 4. Experiment results.

To better understand the differences in CrossCut testing with a PET sample between the classification values GT2 and GT4, Figure 5 is shown.



Figure 5. Cross-Cut testing of PET material. Comparison of IPA cleaned PET polymer (a) Cross-Cut test direct after printing (GT2); (b) Cross-Cut test 24 h. after printing (GT4); (c) Cross-Cut dimensions.

The purpose of the test is to activate the surfaces of different materials to allow adhesion using UV-ink printing with an inkjet printer. The Cross-Cut test shows which

material can achieve stable adhesion with a classification value GT0. Excellent adhesion can result when samples are treated with open-air plasma and printed with UV ink, a process that gives all samples a long stable GT0 value.

Using COMSOL, the shape of the inkjet droplet (Figure 6) is obtained on non-treated and treated PET material surfaces.



Figure 6. Droplet shape on non-treated (a) and treated (b) poly (ethylene terephthalate) surfaces.

Using COMSOL (applying Equations (8) and (9)), the change in droplet velocity at impact on the non-treated and treated surface is illustrated in Figure 7. Here, droplet oscillations were observed during impact and the obtained velocity were higher on the surface of the non-treated poly (ethylene terephthalate).



Figure 7. The change in inkjet droplet velocity on non-treated and treated poly (ethylene terephthalate) surfaces.

It can be noted that different surfaces play a role in achieving different droplet velocities during impact. At the end of the contact process, when the velocity approaches zero, the droplet takes on its final shape on the surface. It is important to mention that, depending on various surface parameters, droplets on the surface take on different shapes. The droplet diameter can be twice as large and cover four times the area if the treated poly (ethylene terephthalate) surface is considered.

During the impact, the droplet velocity changes and decreases to zero. Before reaching a stable phase, several processes of oscillation damping occur on the surface, and their behaviour depends on the properties of the surface. It should be noted that the behaviour of droplet oscillations upon impact depends not only on the characteristics of the surface, but also on the properties of the fluid. The results show that on the non-treated surface of polyethylene terephthalate the level (velocity) of droplet oscillations is twice as high as on the treated surface. In addition, the droplet on the treated surface takes on its final shape faster and is more stable during the printing process.

9 of 11



The change in droplet diameter at impact on a non-treated and treated surface is illustrated in Figure 8.

Figure 8. The change in inkjet droplet diameter on the non-treated and treated poly (ethylene terephthalate) surfaces.

After the impact of a droplet on the non-treated surface, the contact diameter increases to 48  $\mu$ m and after oscillation decreases to a stable shape and size with a diameter of 32  $\mu$ m. In the case of a treated surface, the droplet slowly expands to a diameter of 64  $\mu$ m and remains still. It should be noted that the decrease in the contact area of the droplet on the non-treated surface of polyethylene terephthalate can be predicted by considering surface energies as well as interfacial energy.

# 5. Conclusions

It is known that in the process of inkjet printing on various polymers, it is important to know the surface energy of the polymer in order to better understand the adhesion force. The atmospheric pressure plasma treatment method was examined to reach stable adhesion to polymers with a GT0 classification value in Cross-Cut tests. By applying numerical simulations with COMSOL, many variations in droplet impact parameters have been found on treated and non-treated polymer surfaces, such as changes in droplet velocity and diameter during impact. The behaviour of oscillations of the droplets during impact depends on the properties of the fluid, jetting parameters and the characteristics of the surface energies.

Numerical simulations could help improve print quality and speed and improve the quality of inkjet inks.

In future studies, this research information can be extended to other materials, combined surface treatments and mixed adhesives. With regard to stricter plastics regulations, it is very important to explore the possibility of recycling printed polymers to use them for post-consumer recycling. It has been observed that the adhesion between the ink and the polymers should be appropriate. In addition, the ranking of applied treatments and adhesives can be quantified using statistical tools such as the multidimensional scaling analysis that unveils the most important relationships and conclusions.

Author Contributions: Conceptualization, methodology, investigation, writing—original draft preparation T.T. and R.J. All authors have read and agreed to the published version of the manuscript.

Funding: The research was funded by the Vilnius Gediminas Technical University (VGTU).

Institutional Review Board Statement: The study was conducted according to the guidelines of the Declaration of Helsinki, and approved by the Ethics Committee of UNIVERSITAT POLITÈCNICADE VALÈNCIA (date of approval: 23 October 2018).

Informed Consent Statement: Informed consent was obtained from all subjects involved in the study.

Data Availability Statement: The data presented in this study are available on request from the corresponding author. The data are not publicly available due to privacy.

Conflicts of Interest: The authors declare no conflict of interest.

# Nomenclature

DOD	drop-on-demand;
GT	classification value of the Cross-Cut quality;
HDPE	high-density polyethylene;
IPA	isopropyl alcohol;
PET	polyethylene terephthalate;
PP	polypropylene;
SE	surface energy;
ST	surface tension;
Roman	
d	the diameter of the droplet (mm);
Fst	surface tension force (N);
8	gravitational constant (m/s ² );
Sconst	acceleration due to gravity (m/s ² );
n	the unit vector in the normal direction;
Oh	dimensionless coefficient that represents the printability of the ink;
р	pressure (Pa);
r	droplet radius (mm);
t	time (s);
и	droplet (fluid) velocity (m/s);
Wa	work of the adhesion (J/m ² );
Greek	
β	maximum spreading factor;
Y	parameter which determines the repetition of initiations;
δ	Dirac delta function;
ε	is the representative mesh size in the area passed by the falling droplet;
η	is fluid viscosity (m/s);
θ	droplet contact angle with surface (°);
k	is the curvature;
μ	dynamic viscosity (N·s/m²);
ρ	density (kg/m³);
$\sigma$	surface tension coefficient (mN/m);
φ	coefficient of level set interface between air and ink.

### References

- 1. Petrie, E.M. Handbook of Adhesives and Sealants; McGraw-Hill Education: New York, NY, USA, 2007; p. 800.
- Oosterom, R.; Ahmed, T.J.; Poulis, J.A.; Bersee, H.E.N. Adhesion performance of UHMWPE after different surface modification techniques. Med. Eng. Phys. 2006, 28, 323–330. [CrossRef]
- Novaković, D.; Stančić, M.; Karlović, I.; Kašiković, N.; Vukmirović, V.; Milošević, R. Influence of surface roughness on print quality on digitally printed self adhesive foils. J. Print Media Technol. Res. 2013, 2, 67–76.
- Lim, Y.Y.; Goh, Y.M.; Liu, C. Surface treatments for inkjet printing onto a PTFE-based substrate for high frequency applications. Ind. Eng. Chem. Res. 2013, 52, 11564–11574. [CrossRef]
- Sutherland, I.; Brewis, D.M.; Health, R.J.; Sheng, E. Modification of polypropylene surfaces by flame treatment. Surf. Interface Anal. 1991, 17, 507–510. [CrossRef]
- Farris, S.; Pozzoli, S.; Biagioni, P.; Duó, L.; Mancinelli, S.; Piergiovanni, L. The fundamentals of flame treatment for the surface activation of polyolefin polymers—A review. *Polymer* 2010, *51*, 3591–3605. [CrossRef]
- 7. Brewis, D.M.; Briggs, D. Adhesion to polyethylene and polypropylene. Polymer 1981, 22, 7-16. [CrossRef]

89

- Hegemann, D.; Brunner, H.; Oehr, C. Plasma treatment of polymers for surface and adhesion improvement. Nucl. Instr. Meth. Phys. Res. Sect. B: Beam Interact. Mater. Atoms 2003, 208, 281–286. [CrossRef]
- Sandanuwan, T.; Hendeniya, N.; Attygalle, D.; Amarasinghe, D.A.S.; Weragoda, S.C.; Samarasekara, A.M.P.B. Atmospheric cold plasma to improve printability of polyethylene terephthalate. In Proceedings of the Moratuwa Engineering Research Conference (MERCon), Moratuwa, Sri Lanka, 27–29 July 2021; pp. 654–658. [CrossRef]
- Sundriyal, P.; Sahu, M.; Prakash, O.; Bhattacharya, S. Long-term surface modification of PEEK polymer using plasma and PEG silane treatment. Surf. Interfaces 2021, 25, 101253. [CrossRef]
- Lommatzsch, U.; Pasedag, D.; Baalmann, A.; Ellinghorst, G.; Wagner, H.E. Atmospheric pressure plasma jet treatment of polyethylene surfaces for adhesion improvement. *Plasma Process. Polym.* 2007, 4, S1041–S1045. [CrossRef]
- Sundriyal, P.; Pandey, M.; Bhattacharya, S. Plasma-assisted surface alteration of industrial polymers for improved adhesive bonding. Int. J. Adhes. Adhes. 2020, 101, 102626. [CrossRef]
- Lee, C.C.; Shih, R.C.; Huang, P.C.; Tseng, S.F. Adhesion enhancement of conductive graphene/PI substrates through a vacuum plasma system. Surf. Cont. Technol. 2020, 388, 125601. [CrossRef]
- Ebnesajjad, S.; Ebnesajjad, C. Surface Treatment of Materials for Adhesive Bonding, 2nd ed.; Elsevier: Amsterdam, The Netherlands; William Andrew: MA, USA, 2014; p. 360. [CrossRef]
- Liu, Q.; Lo, J.H.Y.; Li, Y.; Liu, Y.; Zhao, J.; Xu, L. The role of drop shape in impact and splash. Nat. Commun. 2021, 12, 3068. [CrossRef]
- Berdozzi, N.; Chen, Y.; Luzi, L.; Fontana, M.; Fassi, I.; Tosatti, L.M.; Vertechy, R. Rapid fabrication of electro-adhesive devices with inkjet printed electrodes. *IEEE Robot. Autom. Lett.* 2020, 5, 2770–2776. [CrossRef]
- Sundriyal, P.; Bhattacharya, S. Inkjet-printed electrodes on A4 paper substrates for low-cost, disposable, and flexible asymmetric supercapacitors. ACS Appl. Mater. Interfaces 2017, 9, 38507–238521. [CrossRef]
- Sundriyal, P.; Bhattacharya, S. Scalable micro-fabrication of flexible, solid-state, inexpensive, and high-performance planar micro-supercapacitors through inkjet printing. ACS Appl. Energy Mater. 2019, 2, 1876–1890. [CrossRef]
- Wilson, P.; Lekakou, C.; Watts, J.F. System design and process optimization for the inkjet printing of PEDOT:Poly(styrenesulfonate). J. Micro Nano-Manuf. 2014, 2, 011004. [CrossRef]
- Wilson, P.; Lekakou, C.; Watts, J.F. In-plane conduction characterisation and charge transport model of DMSO co-doped, inkjet printed Poly (3, 4-ethylenedioxythiophene): Polystyrene sulfonate (PEDOT: PSS). Org. Electron. 2013, 14, 3277–3285. [CrossRef]
- Wilson, P.; Lekakou, C.; Watts, J.F. A comparative assessment of surface microstructure and electrical conductivity dependence on co-solvent addition in spin coated and inkjet printed poly (3, 4-ethylenedioxythiophene): Polystyrene sulphonate (PEDOT: PSS). Org. Electron. 2012, 13, 409–418. [CrossRef]
- Eckert, W. Surface Pretreatment with Corona, Plasma, Flaming or Silicate Coating: Which Method is Best? Power Point Presentation by Dr. Werner Eckert; Arcotec GmbH: Mönsheim, Germany, 2019; p. 20.
- ASTM D3359-17. Standard Test Methods for Rating Adhesion by Tape Test; ASTM International: West Conshohocken, PA, USA, 2017. Available online: https://www.astm.org/SUBSCRIPTION/REDLINE_PAGES/D3359.htm (accessed on 11 November 2021).
- Bangera, A.E.; Appaiah, K. A conditional justification for the determination of surface energy of solids using contact angle methods. *Mater. Chem. Phys.* 2019, 234, 168–171. [CrossRef]
- 25. Kinloch, A.J. The science of adhesion. J. Mater. Sci. 1980, 15, 2141-2166. [CrossRef]
- Dupré, M.A. Theories Mecanique de la Chaleur; Gauthier Villars: Paris, France, 1869; p. 369. Available online: http://books.google.com/books?id=qD4JAAAAIAAJ&coe=UTF-8 (accessed on 11 November 2021).
- CFD Module User's Guide; COMSOL: Burlington, MA, USA, 2018. Available online: https://doc.comsol.com/5.4/doc/com. comsol.help.cfd/CFDModuleUsersGuide.pdf (accessed on 11 November 2021).
- Bonadiman, R.; Marques, M.; Freitas, G.; Reinikainen, T. Evaluation of printing parameters and substrate treatment over the quality of printed silver traces. In Proceedings of the 2nd Electronics System-Integration Technology Conference, London, UK, 1–4 September 2008; pp. 1343–1348. [CrossRef]
- Good, R.J.; Girifalco, L.A. A theory for estimation of surface and interfacial energies. III. Estimation of surface energies of solids from contact angle data. J. Phys. Chem. 1960, 64, 561–565. [CrossRef]
- Sohr, H. The Navier-Stokes Equations: An Elementary Functional Analytic Approach; Springer Science & Business Media: Berlin, Germany, 2012; p. 367. [CrossRef]
- 31. Plasmatreat USA, Inc. Illinois, USA. Available online: https://www.plasmatreat.com (accessed on 11 November 2021).

# Article 4. Tofan, T. et al. (2022). Modeling 3D Droplet Movement Using a Drop-on-Demand Inkjet Printhead Model (https://doi.org/10.3390/pr10081467)



Article



# Modeling 3D Droplet Movement Using a Drop-on-Demand Inkjet Printhead Model

Tim Tofan¹, Sergejus Borodinas^{2,3}, Rimantas Kačianauskas^{2,3}, and Raimondas Jasevičius^{1,3,*}

- ¹ Department of Mechanics and Materials Engineering, Vilnius Gediminas Technical University, J. Basanavicius Street 28, 03224 Vilnius, Lithuania; tim.tofan@vilniustech.lt
- ² Department of Applied Mechanics, Vilnius Gediminas Technical University, Saulétekio al. 11,
- 10223 Vilnius, Liffuania; sergejus.borodinas@vilniustech.lt (S.B.); rimantas.kacianauskas@vilniustech.lt (R.K.) Institute of Mechanical Science, Vilnius Gediminas Technical University, J. Basanavicius Street 28,
- 03224 Vilnius, Lithuania
- * Correspondence: raimondas.jasevicius@vilniustech.lt

Abstract: This article presents a numerical simulation of a printhead model for drop-on-demand (DoD) inkjet printers. A three-dimensional droplet model is provided for the numerical study of inks, ejection parameters, droplet movement, and the analysis of droplet impacts on the surface. This work is devoted to the analysis of different droplet ejection settings during the printing process, when the behavior of the droplet directly affects the accuracy of the printing process itself. A numerical model was also developed to investigate the effect of various settings on droplet stability, including printhead size and nozzle orifice, motion parameters (pulse strength and droplet ejection amplitude) and fluid properties. The results reflect the behavior of the ink droplet over time. The behavior of the drop was tested at different waveform ejection parameters and a mass turnover was observed.

Keywords: drop-on-demand; inkjet printhead nozzle; droplet; printing process; 3D modeling



Citation: Tofan, T.; Borodinas, S.; Kačianauskas, R.; Jasevičius, R. Modeling 3D Droplet Movement Using a Drop-on-Demand Inkjet Printhead Model. *Processes* 2022, *10*, 1467. https://doi.org/10.3390/ pr10881467

Academic Editor: Yanzhen Zhang

Received: 10 July 2022 Accepted: 25 July 2022 Published: 27 July 2022

Publisher's Note: MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.



Copyright © 2022 by the authors. Licensee MDPJ, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 40/).

# 1. Introduction

Nowadays, inkjet printing technology has become more widely adopted by manufacturers due to its simplicity and flexibility. There is increasing demand for paper-based products, including in such areas as T-shirt printing, microchip production or personalized production, while meeting increasing quality requirements.

An image analysis study of ink-paper interactions was conducted by Fiadeiro et al. [1]. It is important to identify variations in the interaction of ink and paper, manifested in the machine and cross directions of the paper (Mendes et al. [2]). The influence of paper properties on the print quality of an inkjet printer is described by Bandyopadhyay [3]. Skowronski [4] analyzed the complexity of the interaction between ink and paper in inkjet printing and their influence on the properties of printed paper [4]. The initial coating of the paper remains important. The interaction between offset ink and coated paper is described by Ström [5]. Liu and Derby [6] analyzed the droplet formation of inkjet printers using different printing ink properties and actuation strength (voltage). A physical experiment was carried out to determine the possibility of jection of well-shaped drops without the formation of satellite drops. The influence of voltage parameters on the piezoelectric ejection process of ink droplets with different rheological properties was investigated and analyzed by lio et al. [7].

The droplet ejection process in inkjet printers is modeled by a single dimension presented by Jiang and Tan [8]. Droplet stability after ejection was analyzed by Zhong et al. [9]. To control the formation of droplets, Wang and Chiu [10] investigated data-driven models of droplet formation. To improve the print quality by increasing the dots per inch (DPI) of the piezoelectric inkjet printhead, the width and length of the nozzle chamber become smaller. In this situation, the formation of droplets with different waveform times was

Processes 2022, 10, 1467. https://doi.org/10.3390/pr10081467

Processes 2022, 10, 1467

2 of 12

performed by Wei et al. [11]. The printing quality is directly related to the print excitation parameters (Qiu-min et al. [12]). They mentioned that the piezoelectric (PZT) driving system is an important part in inkjet printing equipment. The main function is to eject droplets of an ideal shape and volume at the correct velocity and without satellites to reach the surface at the intended location. A machine learning approach for predicting the maximum spreading factor of droplets upon impact on surfaces with various wettability's were investigated by Tembely et al. [13]. They found that machine learning algorithms can more accurately predict the experimental results of the maximum spreading factor. Increasing the quality of DoD inkjet drops was analyzed by Oktavianty et al. [14]. They mentioned that it is necessary to establish a flexible waveform design for different fluid compositions. The influence of excitation on dynamic characteristics of piezoelectric microjets was investigated by Li et al. [15]. They demonstrated that the direct coupling method proves to be an effective and feasible method to quantitatively simulate the fluid dynamic characteristics of the micro-jet. An evaluation of the relationship between interdigital geometry quality and inkjet parameters was analyzed by Bertolucci et al. [16]. They developed a methodology for assessing the relationships between geometrical quantities and printing parameters. The effect of ink supply pressure on the piezoelectric inkjet was investigated by Kim et al. [17]. They mentioned that decreasing the ink supply pressure can be expected to improve the stability and productivity of inkjet printing. Simulations and sensitivity analysis of piezoelectric inkjet printheads were provided by Nguyen et al. [18]. They developed a lumped element model (LEM) for an inkjet head to simulate its droplet generation in inkjet printing. The inkjet printing of PEDOT:PSS-based conductive patterns for 3D forming applications was investigated by Basak et al. [19]. They developed a polymer PEDOT: PSS-based ink formulation that was inkjet printable through a 21 µm orifice. The effect of process parameters on the performance of drop-on-demand 3D inkjet printing was analyzed by Elkaseer et al. [20]. They developed an optimization-oriented simulation tool of droplet behavior during the drop-on-demand 3D inkjet printing process.

In this paper, we perform numerical simulation using the COMSOL multiphysics finite element method and analyze the ejection of droplet through a nozzle into the air, the shape turnover of droplet, and the distribution of satellites during the fall. Inkjet printing inks are calculated as Newtonian fluid. The flow of ink is modeled by the Navier-Stokes equations for an incompressible fluid. The simulation model presented is solved using the level set method to determine the ink-air interface.

# 2. Problem Formulation

There are many inkjet printers that use inkjet printheads that move in one direction, making the printers easier to use. The print material is moving in the opposite direction of the printhead.

It can also be noted that there are many articles (Introduction section) on similar topics related to inkjet printing, and these topics remain relevant, leading not only to improvements in the printing process itself, but also to such fundamental things as the fundamental behavior of a liquid drop during its formation, movement into air and interaction with a solid surface.

Our study is characterized by the principles of numerical simulation and aspects of their optimization; when using finite elements, we study the chamber of the nozzle of the inkjet printhead, the ejection parameters, the fall of the droplet on the surface and the impact on it. In addition, similar inkjet printing processes are investigated by Elkaseer et al. [20] and Wilson et al. [21].

Elkaseer et al. [20] are investigating inkjet printing technology as an alternative manufacturing method for producing functional parts from multi-material, such as micro-schemes.

They simulated and investigated the printing process, which takes into account the effects of droplet volume, droplet center-to-center distance, jetted droplets coverage percentage, ink-solid substrate contact angle, and overlapping droplets coalescence performance, as well as the number of printed layers. The simulation tool was created using the MATLAB
#### Processes 2022, 10, 1467

programming language. The simulation results were experimentally validated, and the results showed good agreement with the maximum deviation for the horizontal features. The method they developed was used to find out the effect of processed TIFF image resolution and droplet diameter on the dimensional accuracy of inkjet 3D printed parts [20].

For our further research, it is useful to use a geometric-based approach to simulate the coalescence of droplets on a substrate, taking into account the resolution and percentage of coverage of the printed image file and the droplet diameter.

Wilson et al. [21] studied inkjet process optimization for a laboratory inkjet printing system designed to print with polymer inks, with a focus on transparent and electrically conductive polymers. In this study, the authors investigated the speed of droplets from a nozzle as a function of the input voltage waveform and dwelling time difference. With the correct ejection parameters, the authors obtained a continuous line from droplets on the polymer surface. This study [21] presents a new model incorporating the effects of inertia, surface tension, and ink viscoelasticity. Due to the increased viscosity of the ink at low rates, the ink becomes more shear thinning. These studies can be used for our further research on the similar effect of inks on the inkjet process.

It should be noted that when using COMSOL Multiphysics to conduct a numerical experiment, it is necessary to determine the detailed paint parameters. The corresponding geometric parameters of the head are also important. Several of the referenced publications [20,21] do not use COMSOL Multiphysics, so comparing the results may be one of the future tasks.

#### 3. Methodology

Based on standard PZT printhead models, a single inkjet printhead nozzle chamber was modeled using COMSOL Multiphysics software. The geometry of the threedimensional numerical model of the inkjet printhead nozzle is shown in Figure 1.



Figure 1. Inkjet printhead nozzle geometry.

A droplet is formed by jetting a fixed amount of ink into an air-filled cylindrical printing chamber with a length L = 1 mm and a radius R = 0.1 mm. The radius of the jetting channel is equal to  $r_1 = 0.1$  mm. The initial circular shape and the cross-sectional radius  $r_2 = 12 \ \mu$ m of the droplet are predefined by the channel shape and size.

The waveforms are generated by the top inlet of the nozzle by applying the fluid intake velocity and various waveform dwelling times. Different waveforms dwelling times are shown at Figure 2.

93

#### Processes 2022, 10, 1467



Figure 2. The inlet velocity for inkjet printhead nozzle (determined by waveform functions).

Here, the printhead was actuated by an optimized waveform with different dwelling times  $t_1=1~\mu s, t_2=3~\mu s$ , and  $t_3=12~\mu s$ . The considered droplet ejection times were set to demonstrate differences in droplet behavior. Typically, PZT inkjet printheads use different dwelling times to control droplet size on the surface. To better understand the droplet behavior and impact differences, we chose the different dwelling times mentioned above. As the results will show, these are the time intervals when the droplet formed on the printed surface has a surface close to the shape of a hemisphere, with a dwelling time of 1 $\mu s$ , and the spherical surface of the droplet is completely lost with a dwelling time of 12 $\mu s$ ; an additional intermediate dwelling time interval was chosen to be 3 $\mu s$ . The ink inlet velocity was  $V_{in}=0.5~\frac{m}{3}$ . When the pressure waveform appears, the liquid begins to push out the ink through the printhead nozzle, forming the jet. Then the jet pinches off from the nozzle, the main droplet begins to form, and peripheral satellites may appear.

Numerical simulation based on the continuity equation and the N-S equation reconcile two-phase flows of incompressible substances. The continuity equation for an incompressible fluid is calculated using the following equation:

$$\nabla \cdot \mathbf{u} = \frac{\partial \mathbf{u}_{\mathbf{x}}}{\partial \mathbf{x}} + \frac{\partial \mathbf{u}_{\mathbf{y}}}{\partial \mathbf{y}} + \frac{\partial \mathbf{u}_{\mathbf{z}}}{\partial \mathbf{z}} = 0 \tag{1}$$

Here,  $\nabla$  is the vector differential operator, and **u** is the fluid velocity. From the definitions of calculus, we know that the gradient operator ( $\nabla$ ) is calculated by the equation

$$\nabla = \frac{\partial}{\partial x} \mathbf{i} + \frac{\partial}{\partial y} \mathbf{j} + \frac{\partial}{\partial z} \mathbf{k}$$
(2)

where i, j, k are the unit vectors in the directions of the x, y and z coordinates.

The N-S equation describes the momentum conservation equation for a viscous incompressible fluid in motion. This equation is written as follows:

$$\rho(\mathbf{u} \cdot \nabla)\mathbf{u} = \mu \left(\nabla \left(\nabla \cdot \mathbf{u} + (\nabla \cdot \mathbf{u}^{\mathrm{T}}\right)\right) - \nabla p + \rho \mathbf{g}$$
(3)

Here,  $\rho$  is the substance density;  $\mu$  is the substance viscosity; p is the pressure; and  ${\bf g}$  is the acceleration due to gravity.

The following equation describes the interface between the ink and air phases. The convection of the reinitialized level set function is written as the following:

$$\frac{\partial \Phi}{\partial t} + \mathbf{u} \cdot \nabla \Phi = \gamma \nabla \cdot \left( \epsilon \nabla \Phi - \Phi (1 - \Phi) \frac{\nabla \Phi}{|\nabla \Phi|} \right) \tag{4}$$

Here, the coefficient  $\varphi$  represents the level set interface between ink and air. Accordingly,  $\varphi=0$  represents the air environment, and  $\varphi=1$  represents ink. In the transition layer close to the interface,  $\varphi$  smoothly changes from 0 to 1. The fluid velocity,  $u_r$  represents the motion of interface during the environments interplay. The parameter  $\gamma$  determines the amount of stabilization of the level set function. A suitable value for  $\gamma$  is the maximum magnitude occurring in the velocity field.

By default, this reinitialization parameter  $\gamma$  for the current simulation is set to  $\gamma = 10 \frac{m}{s}$ . Here,  $\epsilon$  refers to the thickness of the transition layer. For this simulation,  $\epsilon$  is taken as half of the representative mesh size in the area that the falling droplet passes through. For this simulation, the interface thickness is controlled by the default and set to 1.25  $\mu$ m.

By defining the interface of different substrates, the level set function is used to stabilize the density and viscosity calculations through the interface. In numerical simulation, it is necessary to take into account the dynamic viscosity (shear viscosity)  $\mu$ . It describes the interface between the shear rate and shear stresses in the substrate. The substrate density and dynamic viscosity in different finite elements depend on the interface coefficient  $\phi$ . Density and dynamic viscosity are written as follows:

$$\rho = \rho_1 + (\rho_2 - \rho_1)\phi \tag{5}$$

$$\mu = \mu_1 + (\mu_2 - \mu_1)\phi$$
 (6)

Here,  $\rho_1$  indicates the air density and  $\rho_2$  indicates the ink density;  $\mu_1$  indicates the air viscosity and  $\mu_2$  indicates the ink viscosity.

In numerical simulation, ink and air are investigated as a substrate, from the flow of which viscous stresses arise at each point. The N–S equations describe the mass and momentum transfer of an incompressible fluid, including the surface tension and gravity forces. The motion of the droplet, taking into account the conservation of mass, is calculated from the following differential equations:

$$(\nabla \cdot \mathbf{u}) = 0 \tag{7}$$

$$\rho\left(\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} - \mathbf{g}\right) - \nabla \cdot \left(\mu \left(\nabla \mathbf{u} + \nabla \mathbf{u}^{T}\right)\right) + \nabla p = \mathbf{F}_{st}$$
(8)

The surface tension force is calculated by the equation

$$\mathbf{F}_{st} = \sigma \delta \kappa \mathbf{n}$$
 (9)

Here,  $\delta$  is the Dirac delta function;  $\sigma$  is the surface tension coefficient; k is the curvature; and n is the unit vector (normal direction). The unit vector is calculated from the following equation:

$$\mathbf{n} = \frac{\nabla \Phi}{|\nabla \Phi|} \tag{10}$$

The delta function is approximated by the following equation:

r

$$\Delta = 6|\phi(1-\phi)||\nabla\phi| \tag{11}$$

To calculate the droplet motion, including mass conservation, the adaptive mesh refinement function is used to locally improve the mesh at the ink-air interface. For this simulation, the finite elements sequence and turnover are shown at Figure 3.



Figure 3. Representation of an adaptive mesh refinement sequence.

This functionality will essentially divide the simulation progress into several time intervals and locally refine the mesh in the region, where the phase interface is present in each interval to improve the accuracy of the calculation. To begin with, we have about 220,000 finite elements, and during the ejection, the mesh of the droplet changes 12 times. At the second mesh refinement, the number of finite elements reaches 800,000. The average quality of the elements decreases with an increase in the number of finite elements.

The final droplet mass can be calculated using the following COMSOL Multiphysics numerical simulation equation:

$$\mathbf{m}_{\mathbf{d}} = \rho(\mathbf{V} \cdot \boldsymbol{\Phi}) \tag{12}$$

There are several dimensionless parameters that are very important to the inkjet printhead working process. The Reynolds number Re determines the ratio of a fluid's inertia to its viscosity, while the Weber number We determines the ratio of inertia to its surface tension (Seipel et al. [22]). These dimensionless numbers are calculated using the following equations:

$$Re = \frac{\rho dv_d}{\mu}$$
(13)

$$We = \frac{\rho dv_d^2}{\sigma}$$
(14)

Here, v_d is the droplet velocity and d is the length of the nozzle diameter.

Several effects are observed during the motion of the ink droplet. A typical picture of a droplet, a fragment of the mesh and a contour plot of the velocity magnitude accompanied with the Cell Reynold's number are shown in Figure 4.



Figure 4. Illustration of the shape of a droplet in motion with a mesh and distribution of velocities.

The influence of droplet velocity in the inkjet printing process can be eliminated by forming the Ohnesorge number Oh. There is a solution based on the N–S equations for expressing droplet ejection limitations considering the interfacial, viscous and inertial properties of the fluid (Seipel et al. [22]):

$$Z = \frac{\sqrt{\sigma \rho d}}{\mu} = \frac{1}{Oh} = \frac{Re}{\sqrt{We}}$$
(15)

The Oh value can only be calculated from the physical properties of the inks and the nozzle size scale. It is independent of the waveform extraction impulse that controls the velocity. Oh closely affects the behavior of an expanding droplet from a printhead nozzle.

The initial ink printability specification is required for ejection of stable droplets and should be in the acceptable range of 1 < Z < 10, (Fromm [23]; Derby and Reis [24]). If the number Z > 10, then during the printing process, satellite droplets will form, which degrading the print quality. If the number Z < 1, then the viscous forces of the fluid will prevent the expansion of the droplet.

Proper operation of the printing process in a DoD inkjet printhead requires a propriate combination of physical parameters, which will also depend on the droplet size and velocity (via the Reynolds or Weber number value). The results entail empirical data that are embedded in numerical simulation calculations based on the dimensionless numbers We, Re and Z.

#### 4. Experiment and Results

The physical parameters of ink and air used in numerical simulation of the inkjet printhead nozzle are presented in Table 1.

Table 1. Physical parameters of the substances used in the numerical modeling of the inkjet printhead nozzle.

Substance	ρ kg/m ³	$\mu$ mN·s/m ²	σ mN/m	Oh	Z
Air	1.225	$1.789 \times 10^{-2}$	-	-	-
Ink	1110	14	72	0.32	3.13

97

Processes 2022, 10, 1467



The droplet ejection progress was taken from numerical simulation using COMSOL Multiphysics software. The momentum strength of the droplet ejection is predicted from the different velocity at the fluid inlet. For these simulations, the inlet velocity was set to 0.5 m/s. The presented graphs with different waveform dwelling times are shown in Figure 5.



(b)

Figure 5. Cont.



(c)

Figure 5. Representation of the movement of ejected droplet with different ejection parameters. Waveform dwelling time: 1 µs (a); 3 µs (b); 12 µs (c). Inlet velocity  $V_{in} = 0.5 \frac{m}{s}$ .

When the dwelling time is 1 µs, the final mass of the droplet is  $2 \times 10^{-13}$  kg. The droplet forms a geometrical surface close to a hemisphere after the pitch off the nozzle and reaches the surface approximately 40 µs from the start of actuation. After increasing the dwelling period to 3 µs, the mass of the droplet increases to  $3 \times 10^{-12}$  kg. Larger droplets also form a geometrical surface close to a hemisphere and reach the surface faster by 35 µs. After increasing the formatting time to 12 µs, the droplet is ejected with a large tail and does not form a hemisphere when falling. After the impact with a surface, the droplet splashes and does not form the final round shape on the surface. The final mass before droplet impact with a surface and splashing is  $1.4 \times 10^{-10}$  kg.

The final mass of the droplet depends on the different dwelling time period. The mass turnover of droplets is shown in Figure 6. Here, us and ng stand for microseconds ( $\mu$ s) and nanograms (ng), respectively.

For this research, we investigate the droplet mass formatting with different ejection times. During the droplet formatting periods, the droplet mass increases in accordance with the effect of the inlet velocity. After the waveform decreases, the droplet continues to move forward toward the surface and pitch off from the nozzle.

If the inlet velocity was too low, the droplet would shrink to the nozzle and not move forward to the surface, and the kinetic energy of the droplet would be too low. After the droplet reaches the surface, the droplet splashes and we can see a sudden change in the graph. For a large formatting time of 12  $\mu$ s, we see that a large mass grows in after 35–40  $\mu$ s. This is caused by splashing and the droplet jet replacing the main droplet.

99

10 of 12

```
Processes 2022, 10, 1467
```



Figure 6. Representation of the droplet mass turnover. Mass versus formatting time.

#### 5. Conclusions

At this research, we investigate the droplet ejection turnover over time. The main aim is to understand the behavior of the droplet at different waveform ejection parameters. Mass turnover is also observed. It is important to investigate the trajectory of the fall of the droplet and the mass of the droplet to understand the final place and diameter of the droplet on the surface. These aspects affect the final printing quality. Theoretical studies of three-dimensional numerical simulation help to evaluate such a component as the waveform dwelling time. The different dwelling times indicate a direct effect on the impact of the droplet on the surface. The masses of the ejected droplet before impact with the surface at different dwelling periods are as follows:  $1 \ \mu s - 2.5 \times 10^3 \ \mu g$ ;  $3 \ \mu s - 16 \times 10^3 \ \mu g$ ; and  $12 \ \mu s - 65 \times 10^3 \ \mu g$ . This numerical simulation is an important alternative to quality improvement analysis; further research should be focused on the impact of droplets on different surfaces.

Author Contributions: Conceptualization, methodology, investigation, writing—original draft preparation T.T., S.B., R.K. and R.J. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Institutional Review Board Statement: The study was conducted according to the guidelines of the Declaration of Helsinki and approved by the Ethics Committee of UNIVERSITAT POLITÈCNICADE VALÈNCIA (date of approval: 23 October 2018).

Informed Consent Statement: Informed consent was obtained from all subjects involved in the study. Data Availability Statement: The data presented in this study are available on request from the corresponding author. The data are not publicly available due to privacy.

Data Availability Statement: The data presented in this article is available upon request from the corresponding author.

Conflicts of Interest: The authors declare no conflict of interest.

#### Nomenclature

DoD	drop-on-demand;
N-S	Navier-Stokes;
PZT	piezoelectric;
TIFF	tagged image file format.
Roman	
d	is the diameter of the nozzle (mm);
Fst	is the surface tension force (N);
g	is the acceleration due to gravity $(m/s^2)$ ;
Ľ	is the air length between nozzle and surface (mm);
md	is the droplet mass (kg);
n	is the unit vector in the normal direction;
Oh	is the Ohnesorge number, a dimensionless coefficient;
p	is the pressure (Pa);
R	is the air chamber radius (mm);
r ₁	is the inlet channel radius (mm);
r ₂	is the jetting nozzle radius (μm);
Re	is the Reynolds number, a dimensionless coefficient;
t	is time (s);
Vd	is the droplet volume (pL);
Vin	is the inlet ink velocity (m/s);
We	is the Weber number, a dimensionless coefficient;
Z	is the ink printability range, a dimensionless number.
Greek	
β	is the maximum spreading factor;
γ	is the parameter which determines the repetition of initiations;
Δ	is the delta function;
$\nabla$	is the vector differential operator;
δ	is the Dirac delta function;
ε	is the representative mesh size in the area passed by the falling droplet;
η	is fluid viscosity (m/s);
к	is the curvature;
μ	is the dynamic viscosity (N·s/m ² );
ρ	is the density (kg/m ³ );
σ	is the surface tension coefficient (mN/m);
φ	is the coefficient of level set interface between air and ink;
Vd	is the droplet velocity (m/s).

#### References

- Fiadeiro, P.T.; Mendes, A.O.; Ramos, A.M.M.; Sousa, S.C.L. Study of the ink-paper interaction by image analysis: Surface and bulk inspection. In Proceedings of the 8th Iberoamerican Optics Meeting and 11th Latin American Meeting on Optics, Lasers, and Applications, Porto, Portugal, 22-26 July 2013. [CrossRef]
- Mendes, A.O.; Fiadeiro, P.T.; Ramos, A.M.M.; de Sousa, S.C.L. Development of an optical system for analysis of the ink-paper interaction. Mach. Vis. Appl. 2013, 24, 1733–1750. [CrossRef]
- Bandyopadhyay, S. Effect of paper properties on print quality of ink jet printer. In Proceedings of the IS&T NIP17: International Conference on Digital Printing Technologies, Fort Lauderdale, FL, USA, 30 September-5 October 2001; pp. 141–143.
- Skowronski, J.W.A. Clarification of the Complexity of Ink–Paper Interactions in Inkjet Printing and their Impact on Printed Paper Properties. Zeszyty Naukowe. Rozprawy Naukowe/Politechnika Łódzka 2009, 374, 110.
- Ström, G. Review: Interaction between offset ink and coated paper-a review of the present understanding. In Advances in Paper Science and Technology, Transactions of the XIIIth Fundamental Research Symposium, Cambridge, 2005; I'Anson, S.J., Ed.; FRC: Manchester, UK, 2018; pp. 1101–1137. [CrossRef]
- Liu, Y.; Derby, B. Experimental study of the parameters for stable drop-on-demand inkjet performance. Phys. Fluids 2019, 31, 032004. [CrossRef]
- Jiao, T.; Lian, Q.; Zhao, T.; Wang, H. Influence of ink properties and voltage parameters on piezoelectric inkjet droplet formation. Appl. Phys. 2021, 127, 11. [CrossRef]
- 8. Jiang, H.; Tan, H. One dimensional model for droplet ejection process in inkjet devices. Fluids 2018, 3, 28. [CrossRef]

Processes 2022, 10, 1467

- Zhong, Y.; Fang, H.; Ma, Q.; Dong, X. Analysis of droplet stability after ejection from an inkjet nozzle. J. Fluid Mech. 2018, 845, 378–391. [CrossRef]
- Wang, J.; Chiu, G.T.C. Data-driven drop formation modeling in nanoliter drop-on-demand inkjet printing. In Proceedings of the ASME 2020 Dynamic Systems and Control Conference, Online, 5–7 October 2020; Volume 84287, p. V002T28A002. [CrossRef]
- Wei, H.; Xiao, X.; Yin, Z.; Yi, M.; Zou, H. A waveform design method for high DPI piezoelectric inkjet print-head based on numerical simulation. *Microsyst. Technol.* 2017, 23, 5365–5373. [CrossRef]
- Qiu-Min, W.U.; Xin-Yu, C.U.I.; Lei, X.U.; Xin, S.U. Research on driving waveform and residual oscillation in piezo inkjet. Res. Sq. 2022. [CrossRef]
- Tembely, M.; Vadillo, D.C.; Dolatabadi, A.; Soucemarianadin, A. A machine learning approach for predicting the maximum spreading factor of droplets upon impact on surfaces with various wettabilities. *Processes* 2022, 10, 1141. [CrossRef]
- Oktavianty, O.; Haruyama, S.; Ishii, Y. Enhancing droplet quality of edible ink in single and multi-drop methods by optimization the waveform design of DoD inkjet printer. *Processes* 2022, 10, 91. [CrossRef]
- Li, K.; Liu, J.-K.; Chen, W.-S.; Zhang, L. Influences of excitation on dynamic characteristics of piezoelectric micro-jets. *Micromachines* 2017, 8, 213. [CrossRef] [PubMed]
- Bertolucci, F.; Berdozzi, N.; Rebaioli, L.; Patil, T.; Vertechy, R.; Fassi, I. Assessing the relationships between interdigital geometry quality and inkjet printing parameters. *Micromachines* 2022, 13, 57. [CrossRef] [PubMed]
- Kim, S.; Choi, J.H.; Sohn, D.K.; Ko, H.S. The effect of ink supply pressure on piezoelectric inkjet. *Micromachines* 2022, 13, 615. [CrossRef] [PubMed]
- Nguyen, V.-T.; Leong, J.Y.C.; Watanabe, S.; Morooka, T.; Shimizu, T. A multi-fidelity model for simulations and sensitivity analysis of piezoelectric inkjet printheads. *Micromachine* 2021, 12, 1038. [CrossRef] [PubMed]
- Basak, I.; Nowicki, G.; Ruttens, B.; Desta, D.; Prooth, J.; Jose, M.; Nagels, S.; Boyen, H.G.; D'Haen, J.; Buntinx, M.; et al. Inkjet printing of PEDOT:PSS based conductive patterns for 3D forming applications. *Polymers* 2020, 12, 2915. [CrossRef] [PubMed]
- Elkaseer, A.; Schneider, S.; Deng, Y.; Scholz, S.G. Effect of process parameters on the performance of drop-on-demand 3D inkjet printing: Geometrical-based modeling and experimental validation. *Polymers* 2022, 14, 2557. [CrossRef] [PubMed]
- Wilson, P.; Lekakou, C.; Watts, J.F. System design and process optimization for the inkjet printing of PEDOT: Poly (styrenesulfonate). J. Micro Nano-Manuf. 2014, 2. [CrossRef]
- Seipel, S.; Yu, J.; Nierstrasz, V.A. Effect of physical parameters and temperature on the piezo-electric jetting behaviour of UV-curable photochromic inks. Sci. Rep. 2020, 10, 18841. [CrossRef] [PubMed]
- 23. Fromm, J.E. Numerical calculation of the fluid dynamics of drop-on-demand jets. IBM J. Res. Dev. 1984, 28, 322-333. [CrossRef]
- 24. Derby, B.; Reis, N. Inkjet printing of highly loaded particulate suspensions. MRS Bull. 2003, 28, 815–818. [CrossRef]

## Summary in Lithuanian

## Įvadas

#### **Problemos formulavimas**

Įvairių medžiagų sąveikos elgseną svarbu suprasti ir mechanikos mokslo atžvilgiu. Ne išimtis – mikrodalelės. Mikrodalelių apdirbimas yra svarbus farmacijos, maisto, chemijos, spausdinimo ir kitoms pramonės šakoms. Geresnis dalelių elgsenos supratimas ir galimybė ją imituoti skaitmeniniu būdu gali padėti geriau suprasti tiek moksliniuose darbuose nagrinėjamus, tiek pramonėje vykstančius procesus.

Tyrime nagrinėjama mikrodalelė yra lašelis, kuris judėdamas ir sąveikaudamas gali deformuotis bei prilipti prie nagrinėjamo paviršiaus. Taip pat svarbu suprasti lašelių elgesį, kai jie praranda formą, sąveikaudami su paviršiumi. Norint kiekybiškai įvertinti dalelių elgesį, reikia eksperimentinių įrodymų. Spausdinimo pramonėje lašelis atlieka svarbų vaidmenį ir minėtos problemos nėra išimtis.

Remiantis Smithers (2020) ataskaita, rašalinis spausdinimo būdas bus plėtojamas greičiau nei kitos spausdinimo formos dėl mažėjančių tiražų esamose rinkose ir lankstesnių spaudos pritaikymo galimybių. Plečiantis spausdinimo rinkai, didėja paklausa detaliai ištirti spausdinimo procesų produktyvumą ir kokybę. Šiais laikais rašalinio spausdinimo technologija visame pasaulyje naudojama daugelyje gamybos sričių, tokių kaip mikroschemų gamyba, kompozicinių struktūrų kūrimas, lazerinių optikų gamyba ir daugelis kitų (Castrejon-Pita ir kt., 2013; Takagi ir kt., 2019; Huang ir kt., 2020; Maisch ir kt., 2021; West ir Yoo, 2023). Pasaulyje atlikta daug mokslinių tyrimų, skirtų tirti įvairias rašalinių spausdinimo galvučių struktūras, skirtingus lašelio išspausdinimo parametrus, lašelių judėjimą oru, esant skirtingiems išoriniams veiksniams (poveikiams), bei lašelio sąveikas su įvairiais paviršiais (Son ir kt., 2008; Bussmann ir kt., 2000; Kang ir kt., 2020; Yang ir kt., 2022; Li ir kt., 2022; Xiao ir kt., 2022; Mau ir Seitz, 2023; Wang ir Chiu, 2023).

Mokslininkai yra susikoncentravę į naujausius spausdinimo galvutės tyrimus ir eksperimentus, siekia pritaikyti atradimus pramonės srityse (Okuyama ir Yoshida, 2018; Aqeel ir kt., 2019; Shah ir kt., 2019; Yoshida ir kt., 2019; Sabu, 2022; Wang ir kt., 2023). Daugelyje darbų nagrinėjamos spausdinimo galvučių savybės, tokios kaip dažų rezervuaro parametrai; išspausdinimo angų savybės; lašelių sužadinimo parametrai, lemiantys susidarančių lašelių greitį ir parametrus, taip pat palydovinių lašelių susidarymo sąlygos (Jiang ir Tan, 2018; Driessen, 2018; Li ir kt., 2019; Khan ir kt., 2021; Rump ir kt., 2023). Šiais laikais didėja susidomėjimas kintamu skysčių rašalinių lašelių formavimu ir judėjimo valdymu, atspindinčiu daugybę praktinių pritaikymų, tokių kaip aukštos kokybės spausdinimas neprarandant greičio (Wang ir Chiu, 2020; Kamis ir kt., 2021; Peng ir kt., 2022; Wang ir kt., 2023).

Skaitmeninis eksperimentinis modeliavimas galėtų padėti įvertinti lašelių išstūmimą, formą, kintantį lašelių judesį, įvairius pradinius parametrus ir poveikį, turintį įtakos lašelių judėjimui (tai yra pagrindiniai rašalinio proceso komponentai). Šioje disertacijoje daugiausia dėmesio skirta eksperimentiniam ir teoriniam rašalinių lašelių susidarymo ir judėjimo tyrimui. Disertacijoje pateikiami skaitmeninio modeliavimo aspektai ir jų kūrimo sąlygų charakteristikos.

#### Darbo aktualumas

Mažėjant tiražams, skaitmeninis rašalinis spausdinimo būdas pasaulyje sparčiai populiarėja, atsiranda poreikis spausdinti ant skirtingų medžiagų. Siekiant atitikti aukštus spaudos kokybės ir spausdinimo greičio reikalavimus, būtina užtikrinti tinkamus dažų lašelių formavimosi, judėjimo bei sąveikos parametrus. Išnagrinėjus šiuos parametrus galima nustatyti, koks spausdinimo greitis gali būti parenkamas neprarandant spausdinimo kokybės.

Skaitmeninio rašalinio spausdinimo būdo parametrų tobulinimo aspektai ir jų skaitinis tyrimas yra šio darbo naujumas. Aktualumas paremtas spausdinimo proceso rekomendacijomis bei skaitiniais tyrimais, siejamais su spausdinimo kokybe.

#### Tyrimo objektas

Tyrimo objektas yra rašalo lašelis, kuris naudojamas skaitmeninio rašalinio spausdinimo procese.

#### Darbo tikslas

Ištirti skaitmeninės rašalinės spaudos metu susidariusio lašelio dinamiką skirtingais jo formavimosi etapais, apimant lašelio sąveiką su paviršiumi.

#### Darbo uždaviniai

Atsižvelgiant į žinomos mokslinės literatūros apžvalgą disertacijos leidinių rinkinyje, kai modeliuojama ir analizuojama rašalo lašelio judėjimo elgsena skaitmeninio rašalinio spausdinimo procese, buvo suformuluoti sekantys uždaviniai:

- Taikant fizikinius ir skaitinius tyrimo metodus ištirti rašalinio spausdinimo lašelio susidarymo ir judėjimo sąlygas; nustatyti parametrus, pagal kuriuos rašalo lašelis suformuojamas, ir įvertinti skirtingų veiksnių įtaką rašaliniam spausdinimo procesui; išanalizuoti lašelių judėjimo modeliavimo metodus.
- 2. Ištirti skaitiniu būdu rašalinės spausdinimo galvutės modeliavimo galimybes, įtraukiant lašelių išstūmimo parametrus, lašelių susidarymą ir judėjimą, kai lašelis palieka purkštuką, kad būtų galima išnagrinėti lašelių greitį, tūrį, lašelių formos kitimą ir palydovų susidarymą, taip pat skirtingų paviršiaus parametrų įtaką lašelio sąveikai su paviršiumi.
- Nustatyti rašalų fizikinių parametrų įtaką lašelio formavimuisi ir judėjimui link paviršiaus. Fiziškai išnagrinėti spausdinimo parametrų įtaką spalvinėms charakteristikoms ir ištirti lašelių sąveikos su skirtingais paviršiais aplinkybes pagal paviršiaus charakteristikas.
- 4. Sukurti skaitinį modelį, siekiant ištirti rašalinės spausdinimo galvutės lašelių susidarymą ir judėjimą oru, apimant skirtingų rašalų ir išstūmimo parametrus, tokius kaip lašelio generavimo laikas ir rašalo padavimo greitis. Pasiūlyti sprendimus rašalinėms spausdinimo galvutėms kurti ir spausdinimo kokybei gerinti neprarandant našumo.

## Tyrimo metodika

Nagrinėjant darbo objektą buvo taikyti eksperimentiniai ir skaitiniai tyrimo metodai. Skaitmeninės rašalinės spaudos generuojamas dažų lašelis modeliuojamas baigtiniais elementais naudojant *COMSOL Multiphysics* programinę įrangą. Spalvos charakteristikų atgaminimas ant skirtingų paviršių esant skirtingiems spaudos parametrams buvo matuojamas spektrofotometru ir apdorojamas *Barbieri Rasterlink* programine įranga. Dažomo paviršiaus energijos pokyčiams nustatyti naudojami *Plasmatreat* dažikliai su skirtingais paviršiaus įtempiais. Dažyto paviršiaus adhezinės savybės buvo tikrinamos pagal DIN ISO 2409 standartą. Gauti rezultatai analizuojami pagal skirtingų parametrų pokyčius priklausomai nuo pradinių parametrų.

## Darbo mokslinis naujumas

Remiantis disertacijos publikacijų rinkinyje pateikta žinoma moksline literatūros apžvalga, rengiant disertaciją nustatytas šis skaitmeninės rašalinės spaudos lašelio formavimosi ir judėjimo modeliavimo naujumas:

- Skaitiškai ištirtas skirtingų spausdinimo rašalų poveikis spausdinimo procesui. Rezultatai rodo, kad, priklausomai nuo rašalų fizikinių savybių, keičiasi lašelių sąveikos parametrai ir sąveikos trukmė. Nustatytas rašalinio spausdinimo procese naudojamo lašelio formavimasis pradiniais etapais, fiksuojami lašelio formos ir judėjimo greičio pokyčiai vertikalioje ašyje.
- 2. Fiziškai nustatytas skirtingų spausdinimo parametrų poveikis spalvinių charakteristikų atkūrimui ant skirtingų savybių paviršių, įvertinant atspaudo spektrines savybes CIE L*a*b* skalėje.
- Nustatyti fizikiniai polimerinių medžiagų parametrų pokyčiai esant skirtingiems apdorojimo būdams, įvertinant atspaudo adhezines savybes pagal DIN ISO 2409

standartą. Skaitiškai sumodeliuota rašalinio spaudos lašelio sąveika su skirtingą energiją turinčiais paviršiais, fiksuojant lašelio vilgymo kampus ir judėjimo greičius.

4. Naudojant COMSOL modeliavimo CFD modulio programinę įrangą buvo realizuoti rašalinės galvutės lašelių išstūmimo, formavimosi, judėjimo ir sąveikos procesai. Naudojant sudarytą modelį, galima ištirti lašelio sriegio ilgį ir momentą, kada jis ištraukiamas iš purkštuko, lašelio formą, lašelio masę, lašelio tūrį, lašelio elgseną smūgio į paviršių metu ir galutinę jo būseną. Be to, taikant skaitinį modelį, tapo įmanoma konfigūruoti skirtingus purkštukų parametrus, skirtingus išstūmimo slėgius ir išbandyti skirtingų tipų rašalus.

## Darbo rezultatų praktinė reikšmė

Siekiant nustatyti, išanalizuoti, suprasti rašalinių lašelių formavimo ir judėjimo metu įvairiomis darbo sąlygomis vykstančius procesus, disertacijoje sukurti įvairūs uždaviniai, kurių sprendimas apima eksperimentinius ir teorinius tyrimus. Buvo gauti keli svarbūs parametrai, tokie kaip rašalo masė, lašelių dydis ir rašalo lašelio forma, taip pat buvo ištirti skirtingi spausdinimo procesui įtaką darantys veiksniai, tokie kaip rašalo charakteristikos, spausdinimo galvutės išstūmimo parametrai, purkštuko geometrija. Siūlomą skaitinį rašalinio spausdinimo galvutės modelį būtų galima lengvai įgyvendinti naudojant *COMSOL Multiphysics CFD* programinę įrangą. Disertacijoje taikomas trimatis 3D CFD skaičiavimams sudarytas modelis galėtų padėti tiriant rašalo lašelio susidarymą, rašalinių spausdinimo galvučių projektavimą ir naujų skaitmeninių spausdintuvų kūrimą. Pateiktas skaitmeninis modelis ir metodai padeda įvertinti įvairius parametrus, tokius kaip lašelio išstūmimo iš spausdinimo galvutės laikas, lašelio masė ir skersmuo, jo kritimo greitis ir formos pasikeitimas.

#### Ginamieji teiginiai

- Skaitinis spausdinimo galvutės modelis leidžia ištirti parametrų (u; D_{d.max}; D_h; m_d) įtaką rašalo lašelio formavimuisi kiekviename laiko žingsnyje.
- 2. Reguliuojant rašalinės spausdinimo galvutės išpurškimo parametrus galima gauti vienodą lašelių dengiamąjį plotą ant paviršiaus su skirtingų tipų rašalais.
- Fizikinės rašalo savybės (μ; ρ; σ; Oh; Z) turi lemiamos įtakos modeliuojamo lašelio elgesiui spausdinimo metu, ypač kai lašelis sąveikauja su skirtingų tipų paviršiais. Tokiu būdu, keičiant fizines rašalo savybes, buvo ištirtas poveikis lašelio formavimuisi ir judėjimui.

## Darbo rezultatų aprobavimas

Disertacijos rezultatai buvo publikuoti 6 moksliniuose leidiniuose: 4 straipsniai buvo išspausdinti cituojamumo rodiklį turinčiuose žurnaluose, įtrauktuose į "*Clarivate Analytics Web of Science*" duomenų bazę, 2 – konferencijų medžiagoje.

Darbo rezultatai pristatyti 3 tarptautinėse konferencijose:

18-oji Skaitinės analizės ir taikomosios matematikos tarptautinė konferencija (*International Conference of Numerical Analysis and Applied Mathematics*, ICNAAM 2020), Rodas, Graikija.

- 19-oji Skaitinės analizės ir taikomosios matematikos tarptautinė konferencija (*International Conference of Numerical Analysis and Applied Mathematics*, ICNAAM 2021), Rodas, Graikija.
- 10-oji Kietųjų dalelių judėjimo ir valdymo tarptautinė konferencija (*International Conference on Conveying and Handling of Particulate Solids*, CHOPS 2022), 2022 m. liepos 5–9, Salernas, Italija.

#### Disertacijos struktūra

Disertaciją sudaro įvadas, analitinė literatūros apžvalga, tyrimo metodai, tyrimo rezultatai ir išvados, literatūros sąrašas, autoriaus publikacijų disertacijos tema sąrašas, pridedami keturi autoriaus moksliniai straipsniai ir santrauka lietuvių kalba.

Pirmajame skyriuje apžvelgiama rašalinio spausdinimo struktūra, tyrimo principai ir formuojamo lašelio, esant skirtingiems jo parametrams, tyrimai.

Antrajame skyriuje apžvelgiama teorinė, skaitinė ir eksperimentinė tyrimų metodika, skirta nustatyti rašalo lašelio formavimąsi, išspausto lašelio judėjimą ir lašelio smūgio į skirtingus paviršius elgseną modeliuojant.

Trečiajame skyriuje apibendrinti 1–4 publikacijų eksperimentinių ir skaitmeninių tyrimų rezultatai, atsižvelgiama į bendrą šių darbų sampratą.

Bendra disertacijos apimtis yra 118 puslapių, joje pateikiamos 34 lygtys, 15 paveikslų, 3 lentelės. Rengiant disertaciją buvo panaudoti 104 literatūros šaltiniai.

## 1. Spausdinimo galvučių tipų ir lašelio judėjimui modeliuoti skirtas programas aprašančios literatūros apžvalga

Pirmajame skyriuje pateikiama bendra rašalinių spausdintuvų technologijos veikimo apžvalga; aptarti rašalo parametrams keliami reikalavimai ir lašelių formavimosi pagrindai; aptarti konstituciniai skaitinio modeliavimo metodai; aptarti matematiniai dalelių judėjimo modeliai; įgyvendintas diskrečiųjų elementų modeliavimas: baigtinių elementų metodu ištirtas kietosios mikrodalelės judėjimas lašelio pavidalu ir taikyta *COMSOL* skaičiuojamoji skysčių dinamika. Remiantis publikuotais darbais, pateikiami įvairūs galimi modeliavimo metodai. Nurodomos mikroobjekto modeliavimo galimybės, taikomos kietam ir skystam objektui. Abu atvejai aprašyti skyriuje. Pagrindinis dėmesys skiriamas rašalinių spausdintuvų technologijai, kuri yra pagrindinis šio darbo akcentas. Skyriuje ne tik supažindinama su šia technologija, bet ir supažindinama su galimai taikomomis spausdinimo galvutėmis ir kritimo testo metodais bei taikomomis programomis.

Remiantis literatūros tyrimu, buvo padarytos šios išvados:

- Jei taikomas diskrečiujų elementų metodas, svarbi sąlyga, kad kietoji dalelė gali išlaikyti sferinę formą smūgio metu (standi dalelė su minkštu kontaktu). Tačiau judesio ir sąveikos metu skystas lašelis nuolat keičia formą. Dėl šios priežasties vienos dalelės judesiui kaip lašeliui turi būti taikoma skaitinė baigtinių elementų modeliavimo ir skaičiavimo skysčio dinamikos programinė įranga.
- Stebint besikeičiančią lašelio formą laikui bėgant, lašelių judėjimą ir sąveiką galima geriau paaiškinti, atsižvelgiant į atskirus laiko tarpsnius. Lašelio judėjimą

siūloma suskirstyti į 6 etapus: lašelio išmetimas, lašelio siūlo nuplėšimas nuo antgalio, lašelio sferos formavimasis, lašelis prieš sąveiką, lašelių sąveika (skleidimas) substrato paviršiuje ir lašelio stabilumo fazė.

- Lašelių išmetimo ir judėjimo spausdinimo metu svarbu stebėti lašelių formą ir greičio apykaitą kritimo fazėje. Antra svarbi dalis yra lašelių smūgis į paviršių. Šiuos parametrus galima gauti ir ištirti taikant rašalinio purkštuko modelio skaitmeninį modeliavimą.
- 4. Skaitinio modelio, skirto lašelių susidarymui ir lašelių judėjimui tirti, sukūrimas gali būti naudingas, siekiant pagerinti rašalinių spausdinimo galvučių struktūrą ir rašalų kūrimą. Naudojant skaitinį rašalinės spausdinimo galvutės modelį galima stebėti: lašelių siūlų ilgio kaitą; lašelių parametrų kitimą atitrūkstant nuo purkštuko; lašelių formos, masės ir tūrio apykaitą; lašelių poveikio sąlygas; stabilios būklės ant paviršiaus lašelių charakteristikas. Be to, taikant skaitinį modeliavimą, galima patikimai ištirti skirtingus rašalus, skirtingus išstūmimo nustatymus ir konfigūruoti skirtingus purkštukų parametrus.

## 2. Rašalinio spausdinimo proceso tyrimo metodika

Antrajame skyriuje pateikiama daug teorinių, skaitinių ir eksperimentinių tyrimų, siekiant nustatyti svarbiausius parametrus, turinčius įtakos spausdinamo lašelio formavimuisi, lašelio judėjimui ir lašelio sąveikai su skirtingais paviršiais. Aptariamas ir paaiškinamas duomenų rinkimas ir šiame tyrime naudojamas skaitinio modeliavimo galimybių taikymas. Taip pat pateikiami rašalinių spausdinimo galvučių darbo principai ir skaitinio modeliavimo galimybės, atsižvelgiant į spausdinamų dažų technines charakteristikas ir fizikinius parametrus.

Šiame skyriuje pateikta skaitinių ir eksperimentinių tyrimų metodologija apima šias išvadas:

- COMSOL skaitinis modelis naudojant baigtinių elementų metodą gali būti taikomas tiriant įvairius fizinius efektus, pastebėtus spausdinimo procese, pavyzdžiui, maksimalus lašelio greitis išstūmimo metu, kai slėgis spausdinimo galvutėje yra didžiausias ir lašelio siūlas atsiskiria nuo rašalo purkštuko; lašelio greičio kitimas (angl. *turnover*) jam krintant; lašelio smūgis į paviršių, įvertinant patį procesą.
- Spalvų reprodukcijos matavimo tyrimai apima eksperimentinių priemonių formavimą ir būtinų CIE L*a*b* duomenų rinkimą bei apdorojimą, naudojant matematinius ir statistinius metodus.
- 3. Atsižvelgiant į bandymo procedūrą DIN ISO 2409, turi būti įvertintas rašalo sluoksnio pakartotinis pasipriešinimas jo atsiskyrimui nuo pagrindo, kai reikia šį sluoksnį supjaustyti dešiniojo kampo (angl. *right-angle*) gardele (angl. *lattice pattern*). Šios procedūros metu tiesiogiai neišmatuojama sukibimo jėga, tačiau ji padeda empiriškai nustatyti rašalinio atspausdinto sluoksnio sukibimą su paviršiumi.

# 3. Rašalo lašelių formavimo ir judėjimo modeliavimo tyrimo rezultatai ir išvados

Trečiajame skyriuje pateikiami 4-ių straipsnių fizikiniai ir skaitiniai tyrimo rezultatai. Straipsnių išvados tiesiogiai susietos su baigiamajame darbe suformuotais uždaviniais. Nagrinėjant rašalinio lašelio susidarymą bei judėjimą, išanalizuoti rašalų parametrai ( $\mu$ ;  $\rho$ ;  $\sigma$ ; *Oh*; *Z*), spausdinimo galvutės išpurškimo parametrai (t;  $V_{in}$ ), spausdinamo paviršiaus parametrai (SE;  $\theta$ ), turintys įtakos spausdinimo kokybei ir spaudos greičiui. 1 straipsnyje pateikiami skaitinio modeliavimo rezultatai, įvertinant skirtingų rašalų parametrų įtaką rašalinės spaudos procesui. 2 straipsnyje pateikiamas fizikinis tyrimas rašalinės spaudos spalvoms atgaminti pagal skirtingus spausdinimo parametrus. 3 straipsnyje pateikiamas fizikinis ir skaitinis tyrimas, rašalinės spaudos generuojamų lašelių sąveikos rezultatai su skirtingų charakteristikų polimeriniais paviršiais. 4 straipsnyje nagrinėjama skirtingų spausdinimo galvutės išstūmimo parametrų įtaka rašaliniam spausdinimo procesui. Bendrosiose išvadose apibendrinami visi tyrimo rezultatai ir pateikiamos svarbiausios baigiamojo darbo išvados.

#### 1 straipsnio tyrimo rezultatai

Rašalinio skaitmeninio 1 spausdinimo metu labai svarbu stebėti lašelių formos ir greičio kitimą; lašelio sąveiką su paviršiumi bei stabilią lašelio formą ant paviršiaus po sąveikos. Techniškai visas šias charakteristikas galima atsekti ir užfiksuoti skaitiškai modeliuojant rašalinio purkštuko darbą. Lašelių išstūmimas, formavimasis, judėjimas ir sąveika su paviršiumi buvo tiriami naudojant *COMSOL CFD* modulį ir Navjė–Stokso lygtį. Pateiktas modelis padeda numatyti proceso parametrus ar net ankstyvas problemas, kuriant spausdinimo rašalus ar naujas rašalines spausdinimo galvutes. Kuriant skaitinį modelį, buvo įvertintas kiekvienas svarbus rašalinio spausdinimo proceso žingsnis.

1 straipsnyje aprašomas skaitinis rašalinės spausdinimo galvutės modelis, kurį naudojant palyginama skirtingų rūšių rašalų įtaka spausdinimo procesui. Skirtingų rašalų parametrai pateikiami S3.1 lentelėje.

Medžiaga	$\rho, kg/m^3$	$\mu$ , mN·s/m ²	σ, mN/m	Oh	Ζ
Oras	1,225	$1,789 \cdot 10^{-2}$	—	-	_
Rašalas1	1050	7	72	0,16	6,09
Rašalas ₂	1080	10	72	0,23	4,32
Rašalas ₃	1110	14	72	0,32	3,13

S3.1 lentelė. Skirtingų rašalų savybės (Tofan, 2021)

Skirtingų rašalų lašelių greičio ir matmenų parametrai pateikiami spausdinimo proceso laiko etapais (nuo A iki F). Lašelių formatavimo etapai suskirstyti tokiais žingsniais: pirmasis žingsnis (A) parodo didžiausią lašelių greitį, kai rašalas išstumiamas iš spausdinimo galvutės; antrasis žingsnis (B) yra tada, kai lašelio siūlas atsikabina nuo spausdinimo galvutės, šiuo metu lašelio ilgis yra didžiausias; trečiasis žingsnis (C) fiksuojamas, kai lašelis suformuoja sferą; ketvirtasis etapas (D) atvaizduoja lašelį prieš sąveiką su paviršiumi; penktasis žingsnis (E) fiksuoja didžiausią lašelio skersmenį ant paviršiaus osciliacijos metu; paskutinis žingsnis (F) atvaizduoja lašelį galutinėje stabilioje formoje po osciliacijos (virpesių) procesų. Pagrindiniai spausdinimo proceso etapai pateikiami S3.2 lentelėje.

Rašalası			Rašalas2			Rašalas₃						
Etapai	t µs	u m/s	D _{d.max} μm	κ D _h μm	t µs	u m/s	D _{d.max} µm	D _h µm	t µs	u m/s	D _{d.ma} μm	μm D _h
Α	2	13,8	26,3	9,2	2	13,8	25,8	9,7	2	13,8	23,5	9,9
В	11	8,51	20,4	110,9	13	7,88	20,2	115,3	14	7,28	20,0	118,7
С	26	7,05	23,3	26,2	32	6,77	24,9	26,3	39	5,93	24,0	26,6
D	143	4,48	22,2	21,7	150	4,28	22,2	21,8	169	3,60	21,9	22,0
Е	149	0,18	47,7	4,3	161	0,12	42,8	7,2	183	0,05	37,5	11,6
F	174	0	36,0	11,6	172	0	36,1	12,1	181	0	36,3	11,9

**S3.2 lentelė.** Generuojamo spaudos lašelio parametrai skirtingais etapais pagal rašalų savybes (Tofan, 2021)

Spausdinimo metu svarbu stebėti visus šiuos etapus, kol lašelis užsifiksuoja ant spausdinamo paviršiaus. Jei palyginsime visų trijų rašalų spausdinimo procesus, rašalas₂ atvaizduoja geriausius rezultatus, rašalo₂ lašelis turi mažiausią osciliacijos rodiklį ir per trumpiausią laikotarpį paviršiuje pasiekia stabilią fazę. Rašalo₁ lašeliai greičiau pasiekia paviršių, tačiau esant mažiausiems klampumo ir tankio parametrams, jie stipriau osciliuoja paviršiuje, kol įgauna galutinę formą. Rašalai, kurie sukietinami UV šviesoje, turi būti sukietinti stabilioje lašelio fazėje.

Greičio pokytis yra svarbus veiksnys, siekiant užtikrinti spausdinimo kokybę, nes šis pokytis turi tiesioginę įtaką rašalų pasiskirstymui ant paviršiaus. Skirtingų rašalų lašelių greičio pokytis išpurškimo, kritimo ir sąveikos metu pavaizduotas S3.1 pav.



**S3.1 pav.** Skirtingų dažų lašelių greičio pokytis spausdinimo metu (Tofan, 2021)

Lašelio didžiausiasis greitis yra žingsnyje A, kai slėgio banga purkštuko viduje pasiekia aukščiausią lygį. Toliau lašelio greitis mažėja, kai lašelis išsitempia ir galiausiai atitrūksta nuo spausdinimo purkštuko žingsnyje B. Žingsnyje C lašelis suformuoja sferą ir tęsia judėjimą link paviršiaus. Lašeliui krentant, greitis nuolat mažėja, kol lašelis pasiekia paviršių žingsnyje D ir prasideda sąveikos procesas su paviršiumi. Žingsnyje E lašelio skersmuo ant paviršiaus yra didžiausias, o žingsnyje F parodomas stabilios būsenos lašelis, kai greitis lygus nuliui. Taip pat galime fiksuoti lašelių parametrų skirtumus, priklausomai nuo laiko žingsnių. Toliau S3.2 pav. parodoma parametrų  $D_{d.max}$  (lašelio skersmuo pagal horizontalų skerspjūvį) ir  $D_h$  (yra lašelio ilgis pagal vertikalų skerspjūvį) kaita laiko atžvilgiu.



**S3.2 pav.** Skirtingų dažų suformuotų lašelių parametrų pokytis spausdinimo metu (Tofan, 2021)

Nustatyta, kad pagal rašalų fizikines savybes ( $\mu$ ;  $\rho$ ) galima tikėtis skirtingų lašelių elgesio ir sąveikos rezultatų esant vienodoms rašalų išpurškimo sąlygoms. Lyginant skirtingus rašalų tipus nustatyta, kad lašelio smūgio į paviršių metu mažesnio klampumo ir tankio dažų svyravimo (osciliacijos) amplitudė spausdinimo paviršiuje yra didžiausia. Didžiausias lašelio skersmuo paviršiuje yra rašalo₁ žingsnyje E₁ = 47,7 µm. Skirtingų lašelių suformuotų sferų skersmuo kritimo metu yra apie 22 µm. Po sąveikos su pagrindu lašelis praranda sferinę formą ir svyruoja paviršiuje, kol galiausiai įgauna stabilią formą. Mažiausias lašelio paskleidimo skersmuo yra naudojant rašalą₃, kurio tankis ir klampumas yra didžiausi. Šio rašalo lašelis beveik nesvyruoja paviršiuje ir įgauna galutinę formą iškart po sąveikos, kai taikomi mažiausi svyravimo parametrai.

Modeliuojant lašelio judėjimą, išlieka klausimas ne tik dėl dinaminio elgesio, bet ir dėl spalvos charakteristikų. Atlikus skaitinius eksperimentus, buvo pastebėta, kad, norint išanalizuoti rašalų spalvų atkūrimo savybes ant paviršiaus naudojant įvairius spausdinimo parametrus, reikalingi tolesni fizikiniai tyrimai.

#### 2 straipsnio tyrimo rezultatai

Siekiant išgauti geriausius spausdinimo parametrus spalvoms atkurti ant paviršių, buvo atlikti eksperimentai su skaitmenine rašaline spauda ant skirtingų lino tekstilės paviršių. Norint gauti tikslesnius rezultatus, buvo nuspręsta, kad atkurtų spalvų reikšmės natūralios dienos šviesoje būtų lyginamos su skaitmeninių spalvų reikšmėmis.

Fiziniam eksperimentui atlikti buvo parinkta 10 lino audinių su skirtingais atspalviais: balintas, melanžinis (natūralus siūlas, maišytas su balintu lino siūlu) ir natūralios spalvos linas. Audinio paviršiaus spalva, struktūra ir gramatūra turi didelę įtaką spalvų atkūrimui spausdinant skirtingų spalvų dažais.

Spausdintų spalvų  $L^*a^*b^*$  parametrai buvo lyginami su etaloninių skaitmeninių spalvų parametrais.

Balinto lino audiniai (OBR 840, OBR 0114, OBR 491, OBR 1542) pasižymi panašiomis spalvų atkūrimo savybėmis, kaip ir baltas popieriaus lapas, o natūralių lininių audinių (OBR 166, OBR 051, OBR 1041) spalvų atkūrimas yra labai ribotas. Spausdinant ant balinto lininio audinio, spalvos geriausiai atkuriamos ant storiausio balinto lininio audinio (OBR 1542), o prasčiausiai – ant ploniausio natūralaus lino audinio (OBR 1041). Audinio spalva daro didelę įtaką rašalinio spausdintuvo spalvų atkūrimui. Spalvų atkūrimas ant lininio audinio spausdinant 4 kartus ir 2 rašalo sluoksniais pavaizduotas S3.3 pav.



**S3.3 pav.** Spalvų charakteristikos ant lino audinių dvimatėje erdvėje a*b*, spausdinant 4 praėjimais ir dengiant 2 sluoksniais rašalo (Tofan, 2021)

Siekiant įvertinti skaitmeninių rašalų stabilumą, spalviniai parametrai buvo matuojami praėjus 90 dienų po spausdinimo proceso. Matuojamos 6 spalvų charakteristikos: Y – geltona; R – raudona; M – purpurinė; B – mėlyna; C – žydra; G – žalia. Skaitmeninių etaloninių spalvų a*b* reikšmės pažymėtos juoda linija S4.3 pav., o spalvų a*b* reikšmės, išmatuotos atspausdinus ant popieriaus, pažymėtos balta linija. Ribotas spalvų atgaminimas ant visų rūšių lino audinių aiškiai pasiskirsto į 3 grupes pagal audinio spalvas. Sunkiausia atgaminti C ir B spalvų sodrumą. Spalvų atkūrimas spausdinant ant balinto lininio audinio yra artimiausias spalvų atkūrimui ant popieriaus, o spausdinant ant natūralaus lino audinio yra labai ribotos spalvų sodrumo atkūrimo galimybės.

Didžiausias dėmesys buvo skiriamas tiriant spausdinimo sluoksnių skaičiaus (nuo 1-o iki 5-ių) ir spausdinimo praėjimų skaičiaus (4, 8, 16 kartų) įtaką spalvų atkūrimui ant skirtingų lino audinių paviršių. Spausdinant skirtingais praėjimų skaičiais spalvų  $L^*a^*b^*$  reikšmės nekinta ir spalvų skirtumai nepastebėti, tačiau praėjimo skaičius turi didelę įtaką bendram spaudos greičiui. Spalvų skirtumas tarp skirtingų spaudinių yra  $\Delta E \leq 3$ .

Siekiant detaliau išnagrinėti spalvinių charakteristikų pokytį pagal skirtingus rašalo sluoksnius ir spausdinimo praėjimus, pasirinktas balintas lininis audinys (OBR 840). Spausdinant skirtingais rašalų sluoksniais, didžiausias yra šviesumo ( $L^*$ ) parametro pokytis. Didžiausias skirtumas tarp spausdintų sluoksnių fiksuojamas tarp 1-o ir 5-o sluoksnio ( $\Delta E$  nuo 3 % iki 55%), o skirtumas tarp 3-čio ir 4-to sluoksnių ( $\Delta E \le 5\%$ ) ir tarp 4-to ir 5-to sluoksnio ( $\Delta E \le 4\%$ ) yra nereikšmingas. Spausdinant 1-u rašalo sluoksniu, spalvų atkūrimas yra prasčiausias, spalvoms trūksta ryškumo. Geriausias spalvų atkūrimas pasiekiamas spausdinant 2-iem sluoksniais. Spausdinant 4-iais ir 5-iais rašalo sluoksniais, spalvos charakteristikos  $\Delta E$  pokytis didėja ir spalvų atkūrimas yra mažiau efektyvus, nei 2-iejų ir 3-jų rašalo sluoksnių.

Palyginus 4 ir 16 praėjimų spausdinimo nustatymus, išliko tie patys  $\Delta E$  spalvos charakteristikų pokyčiai. Lyginant spalvų atkūrimo skirtumą nuo praėjimų skaičiaus, spalvų kitimas yra nereikšmingas ( $\Delta E \leq 3$ ), išskyrus žalią spalvą ( $\Delta E \leq 14$ ).

Atlikus fizinius eksperimentus esant skirtingiems spausdinimo nustatymams ir įvertinus jų įtaką spalvų perteikimui, pastebėta, kad reikalingi tyrimai, kuriuose būtų išanalizuota skirtingų spausdinių paviršiaus įtaka spausdinių kokybei. Būtina ištirti paviršiaus energijos įtaką lašelio dengiamam plotui ir vilgymo kampui ant paviršiaus. Kiti tyrimai turėtų apimti lašelių susidarymą, judėjimo greitį ir sąveikos su įvairiais paviršiais aplinkybes, siekiant nustatyti, kaip greitai lašeliai pasiekia stabilią fazę, lašelių dengiamąjį plotą ir lašelių drėkinimo kampus ant spausdinimo paviršiaus. Ankstesni tyrimai, atsižvelgiant į pagrindinę tyrimų kryptį, t. y. spaudos kokybės užtikrinimą, reikalauja papildomo tyrimo, orientuoto į lašelių sąveikos su paviršiumi atvejus. Šis tyrimas papildys dvi ankstesnes publikacijas ir jame bus išnagrinėta paviršiaus energijos įtaka lašelio sąveikai su paviršiumi. Tai tiesiogiai paveiks spausdinimo kokybės parametrus.

#### 3 straipsnio tyrimo rezultatai

Pirmiausia, norint geriau išanalizuoti lašelių sąveiką su paviršiumi, svarbu sudaryti tinkamas sąlygas lašeliams nusodinti. Pavyzdžiui, kontroliuojant lašelių nusodinimą ant paviršiaus, galima padidinti spausdinimo greitį, neprarandant spausdinimo kokybės. Lašelių nusodinimas ant spausdinimo paviršiaus rašalinio skaitmeninio spausdinimo procese apibūdinamas kaip penkių nuoseklių etapų seka: sąveika, sklaida, atsipalaidavimas, drėkinimas ir pusiausvyra.

Fizinis eksperimentas atliktas ant juodos spalvos 4 skirtingų polimerinių medžiagų. Visos šios žaliavos naudojamos maisto pramonėje ir gali kontaktuoti su maisto produktais. Medžiagų parametrai nurodyti S3.3 lentelėje.

Bandiniai	Žaliavos gamintojas	Medžiagos storis (mm)	Paviršiaus šiurkštumas (µm)	Paviršiaus energija (mN/m)	
1. PP (polipropilenas)	MOPLEN HP500N	1	2,8	32	
2. PP (polipropilenas)	MOPLEN HP500N	1	0,05	28	
3. HDPE (didelio tan- kio polietilenas)	TIPELIN 3110J	1	2,5	32	
4. PET (polietileno tereftalatas)	NEOPET80	1	0,05	38	

S3.3 lentelė. Spausdinamų medžiagų parametrai (Tofan, 2021)

Siekiant įvertinti dažų dangos atsparumą mechaniniam poveikiui, taikomas DIN ISO 2409 tyrimo metodas. Tyrimo rezultatai atvaizduoti S3.4 lentelėje. Paviršiaus energijai nustatyti naudojami *Plasmatreat* bandomieji rašalai esant skirtingoms įtempimo energijoms. *Plasmatreat* testavimo rašalai pagaminti pagal DIN 53364 ir ISO 8296 standartus. Lašelio ir paviršiaus sąveikos kampai buvo modeliuojami, naudojant *COMSOL Multiphysics* programinę įrangą.

Tyrimai	Bandiniai	Paviršiaus energija (mN/m)	Vilgymo kampas (°)	DIN ISO 2409 tyrimas iškarto po spausdinimo proceso	DIN ISO 2409 tyrimas praėjus 24 val. po spausdinimo proceso
	1	32	82,3	GT1	GT4
1	2	28	83,8	GT3	GT4
1	3	32	78,1	GT4	GT4
	4	38	79,7	GT3	GT4
2	1	32	-	GT1	GT4
	2	28	-	GT2	GT4
	3	32	-	GT2	GT4
	4	38	-	GT2	GT4
3	1	70	52,4	GT0	GT0
	2	58	55,3	GT0	GT0
	3	72	48,7	GT0	GT0
	4	72	49,1	GT0	GT0

S3.4 lentelė. Tyrimų rezultatai (Tofan, 2021)

Eksperimento metu siekiama nustatyti spausdinimo dažų ir skirtingų medžiagų paviršių sukibimo priklausomybę nuo skirtingų apdorojimo būdų: 1 tyrimas atliktas be jokių paviršių apdorojimų, 2 – nuvalius paviršius su izopropilo spiritu, 3 – apdorojant paviršius plazminiu įrenginiu. Taikant DIN ISO 2409 tyrimo metodą, galima nustatyti, kuri medžiaga gali pasiekti gerą sukibimą su paviršiumi pagal klasifikavimo vertes GT0-GT4. Klasifikacijos vertė GT0 rodo puikų dažų sukibimą su paviršiumi, kai nėra pažeidžiamas rašalo sluoksnis, o vertė GT4 – kai pašalinama daugiau nei 35 proc. atspausdinto sluoksnio. Visų bandinių rezultatai yra skirtingi, matuojant dažų sukibimą pagal DIN ISO 2409 standartą iškart po spausdinimo, tačiau galutinis matavimas atliekamas po 24 valandų. Puikūs sukibimo rezultatai fiksuojami, kai mėginiai prieš spausdinimą buvo apdoroti RD2005 plazminiu įrenginiu. Šis apdorojimas užtikrino stabilų GT0 klasifikacijos vertės išlaikymą visiems mėginiams.

Taikant skaitinį modeliavimą, buvo ištirta lašelių greičio kaita spausdinant ant neapdorotų ir apdorotų paviršių, S3.4 pav.



**S3.4 pav.** Lašelių greičio pokytis sąveikos metu (žingsnyje D) su neapdorotu ir apdorotu plazminiu įrenginiu polietileno tereftalato paviršiumi (Tofan, 2021)

Lašeliui kontaktuojant su skirtingais paviršiais, greitis fiksuojamas kas 1 µs. Prieš pasiekiant stabilią paviršiaus fazę, pastebėti keli virpesių slopinimo procesai, o lašelių elgesys priklauso nuo paviršiaus energijos. Svarbu pažymėti, kad lašelių osciliacija sąveikos metu taip pat priklauso nuo skysčio įtempties energijos, tačiau šiam skaitiniam eksperimentui buvo naudojami vienodi spausdinimo rašalai, turintys tas pačias savybes. Rezultatai rodo, kad ant neapdoroto paviršiaus lašelio osciliacijos svyravimų greitis yra du kartus didesnis, lyginant su apdorotu paviršiumi. Neabejotinai skirtingos paviršiaus energijos lemia skirtingą lašelio sąveiką su paviršiais. Kai lašelio judėjimo greitis sumažėja iki nulio, lašelis paviršiuje pasiekia stabilią būseną ir įgauna galutinę formą, S3.5 pav.



S3.5 pav. Lašelių forma ir vilgymo kampai ant neapdoroto ir apdoroto plazminiu įrenginiu polietileno tereftalato paviršiaus (Tofan, 2021)

Atliktas skaitinis modeliavimas naudojant *COMSOL* programinę įrangą. Pasitelkus modeliavimą buvo nustatytas rašalo lašelių greitis ir skersmens pokytis sąveikos metu su apdorotais ir neapdorotais polimerų paviršiais. Dažų susigėrimo koeficientas nėra taikomas, nes spausdinama ant vienalyčių polimerinių medžiagų. Apdorojimo metodai, kai padidinama paviršiaus energija be jokių matomų fizinių pokyčių, turi įtakos galutinei rašalo

lašelių formai ir sumažina sąveikos osciliacijos poveikį. Spausdinant rašaliniu būdu ant skirtingų paviršių, norint geriau suprasti dažų ir polimero sukibimo savybes, svarbu nustatyti paviršiaus energijos vertes. Lašeliai ant paviršiaus turi skirtingus drėkinimo (kontakto) kampus pagal skirtingus paviršiaus parametrus. Lašelių dengiamasis plotas ant paviršiaus gali skirtis keturis kartus, o lašelio aukštis – du kartus (atsižvelgiama į paviršiaus energijos vertę). Lašeliui atsitrenkus į neapdorotą polietileno tereftalato paviršių, lašelio skersmuo svyravimo metu padidėja iki 48 μm, o po to sumažėja iki 32 μm stabilumo būsenoje. Spausdinant ant apdoroto paviršiaus, lašelio skersmuo tolygiai didėja ir išlieka galutinės formos, jo skersmuo yra 64 μm. Lašelio kontaktinio ploto sumažėjimą ant neapdorotų paviršių galima numatyti apskaičiuojant paviršiaus energiją, rašalų paviršiaus įtempimą ir sąsajos energiją. Lašelio virpesiai sąveikos metu priklauso nuo rašalo savybių, spausdinimo galvutės purškimo parametrų ir paviršiaus energijų charakteristikų.

Skaitinio modeliavimo tyrimas yra naudingas spausdinimo galvučių struktūros tobulinimui, spausdinimo kokybės gerinimui, spausdinimo greičio didinimui ir naujų rašalų kūrimui. Atliekant papildomus tyrimus, svarbu išnagrinėti lašelių generavimo signalus ir lašelio išstūmimo iš spausdinimo galvutės procesą. Taip pat aktualu nustatyti, kokie rašalinio spausdintuvo galvutės valdymo parametrai turi tiesioginę įtaką formuojant lašelius.

#### 4 straipsnio tyrimo rezultatai

Šiame straipsnyje siekiama nustatyti spausdinimo galvutės lašelio išpurškimo parametrų  $(t; V_{in})$  įtaką lašelių formavimuisi ir judėjimui. Pasitelkus skaitinį modeliavimą nustatomos lašelio formavimosi tendencijos, esant skirtingiems išmetimo parametrams, ir nagrinėjama sužadinimo laiko įtaka lašelių susidarymui. Taip pat nagrinėjama lašelių masės ir judėjimo greičio kitimo priklausomybė bei paviršiaus dengiamas plotas. Visi šie aspektai turi įtakos galutinei spaudos kokybei. Skaitinis modeliavimas trimatėje erdvėje puikiai pritaikomas, siekiant detaliai ištirti, kaip sužadinimo impulso laikas (t) ir dažų padavimo greitis  $(V_{in})$  lemia lašelio formavimąsi. Jei sužadinimo impulsas ir skysčio greitis ties purkštuku bus per mažas, lašelio energijos neužteks atsikabinti nuo spausdinimo purkštuko. Jei lašelio purškimo impulsas bus per stiprus, lašelis suformuos daugybę piktybinių palydovų ir po smūgio į paviršių susidarys purslai, kurie pakenks spausdinimo kokybei dėl nekontroliuojamo rašalais dengiamo ploto.

Lašelių judėjimas modeliuojamas, naudojant *COMSOL Multiphysics* programinę įrangą. Lašelio formavimosi procesą nuspėja rašalo įtekėjimo greitis ir sužadinimo užlaikymo laikas. Tyrimo metu rašalų įtekėjimo greitis buvo fiksuotas 0,5 m/s, esant skirtingoms sužadinimo laiko reikšmėms:  $t_1 = 1 \mu s$ ;  $t_2 = 3 \mu s$ ;  $t_3 = 12 \mu s$ . Lašelių formavimosi ir judėjimo rezultatas atvaizduotas S3.6 pav.



**S3.6 pav.** Lašelių formavimasis ir judėjimas esant skirtingiems sužadinimo laiko parametrams: (a)  $-t_1 = 1 \ \mu$ s; (b)  $-t_2 = 3 \ \mu$ s; (c)  $-t_3 = 12 \ \mu$ s (Tofan, 2022)

Išstūmimo fazėje lašelių masė didėja proporcingai spausdinimo galvutės rašalo įtekėjimo greičiui. Lašelis, kurio sužadinimo laikas 3 µs, pasiekia paviršių tuo pačiu metu, kaip ir lašelis, kurio sužadinimo laikas buvo 12 µs, tačiau pastebimas akivaizdus skirtumas sąveikos su paviršiumi metu. Lašelis, kurio sužadinimo laikas yra 3 µs, beveik nesvyruoja paviršiuje, o lašelis, kurio susidarymo laikas yra 12 µs, sąveikos metu išsitaško purslais ir lieka netolygios formos ant paviršiaus. Šis rezultatas rodo, kad, priklausomai nuo spausdinamo vaizdo skiriamosios gebos, galima naudoti visus 3 skirtingus formatavimo laikus, siekiant padidinti spausdinimo greitį ir kokybę. Jei reikalaujama aukštos raiškos kokybės spaudinių, geriau naudoti nedidelį sužadinimo laiką, kad būtų formuojami maži lašeliai, nesant papildomai susidariusių mažesnio skersmens palydovinių lašelių. Jei nereikia aukštos kokybės ir reikalaujama padengti storą to paties rašalo sluoksnį, galima naudoti ir didelio užlaikymo suformuotus lašelius.

Lašelių masės kitimas pagal skirtingus sužadinimo laiko tarpsnius atvaizduojamas S3.7 pav.



**S3.7 pav.** Lašelių masės kaitos esant skirtingiems išmetimo parametrams vaizdavimas. Masė ir formatavimo laikas (Tofan, 2022)

Skirtingas sužadinimo laikas tiesiogiai turi įtakos lašelių masės formavimuisi. Didžiausias 12  $\mu$ s sužadinimo laikas lemia masės sumažėjimą po 30  $\mu$ s, kai lašelis suskaidomas dalimis, ir padidėja po 35  $\mu$ s, kai lašelį paveja ir prisijungia susiformavę palydoviniai lašeliai. Skirtingų sužadinimo laikotarpių lašelių masė prieš sąveiką užfiksuota: 1  $\mu$ s – 2,5 ng, 3  $\mu$ s – 16 ng, 12  $\mu$ s – 65 ng. Pateiktas skaitinis modelis yra puikus įrankis skirtingiems spausdinimo galvučių nustatymams derinti, siekiant aukštos kokybės spaudos.

## Bendrosios išvados

 Atlikus skaitinį eksperimentą nustatyta, kad rašaliniam spausdinimo procesui turi įtakos šie parametrai: laikas, per kurį išspausdintas lašelis suformuoja sferą žingsnyje C (angl. versijoje Fig. 1.4), ir greitis, kuriuo jis pasiekia paviršių žingsnyje D. Taip pat svarbu numatyti lašelio sąveikos su paviršiumi rezultatus: dengiamas lašelio plotas, vilgymo kampas, laikas, per kurį lašelis pasiekia stabilią fazę (žingsnyje nuo D ir F).

- Ištyrus įvairių tipų rašalų poveikį spausdinamam paviršiui, nustatyta, kad, priklausomai nuo rašalo fizikinių savybių (μ; ρ; σ; *Oh*; *Z*), vyksta skirtingas lašelio judėjimas ir formavimasis, taip pat susidaro skirtingos sąveikos aplinkybės. Taigi šių minėtų parametrų pokytis yra svarbus kontroliuojant spausdinimo procesą ir užtikrinant spausdinimo kokybę.
- 3. Skaitinis eksperimentas leidžia įžvelgti spausdinimo proceso ypatybes. Lašelio formavimosi ir judėjimo pokyčius nusodinimo metu galima kontroliuoti priklausomai nuo spausdintuvo galvutės ir išstūmimo parametrų. Kontroliuojant lašelio trajektoriją ir lašelio masę, galima numatyti galutinę lašelio vietą ir dengiamą plotą paviršiuje.
- 4. Iš grafinių rezultatų matyti, kad lašelio formos pokyčius laiko atžvilgiu galima geriau paaiškinti, padalijus visą procesą į atitinkamus etapus. Lašelio judėjimą siūloma suskirstyti į 6 etapus: lašelių išmetimas, lašelio siūlo atskyrimas nuo purkštuko, lašelio sferos formavimas, lašelis prieš sąveiką su paviršiumi, lašelių sąveika (skleidimas) ant pagrindo paviršiaus ir lašelio stabilumo fazė. Siūlomas šešių etapų tyrimas leidžia greičiau aptikti išstumto lašelio formos pokyčius per atitinkamą laiko intervalą, kai jis laisvai krenta link sąveikaujančio paviršiaus.
- 5. Naudojant COMSOL CFD modeliavimo programinės įrangos modulį, buvo išanalizuotas lašelių formavimasis, judėjimas ir sąveika su paviršiais. Po skaitinių eksperimentų pastebėta, kad pateiktame tyrime svarbu išanalizuoti skirtingų dažų fizinių savybių poveikį spausdinimo procesui, skirtingus lašelio generavimo parametrus ir skirtingus spausdinamų paviršių parametrus.
- 6. Sudarytas skaitmeninis modelis rašalinio lašelio judėjimui ir parametrams tirti. Naudojant COMSOL programinę įrangą, buvo rastas lašelio formos pokytis ir lašelio atsiskyrimo nuo purkštuko laiko momentas, lašelio sferinės formos sukūrimo laiko momentas, lašelio būsena sąveikos metu ir galutinė lašelio būsena ant paviršiaus. Skaitinis modelis suteikia galimybę detaliai išnagrinėti skirtingus purkštukų parametrus, skirtingus lašelio generavimo parametrus ir išbandyti spausdinimą su skirtingų parametrų rašalais.
- Taikant 3D CFD modulį, kai nagrinėjamas skaitmeninio spausdintuvo galvutės veikimas, galima įvertinti tokių komponentų įtaką: lašelio sužadinimo laikas, lašelio masės ir jo parametrų kitimas, lašelio greičio pokytis. Šiame darbe buvo įvertinta skirtingų lašelių generavimo parametrų įtaka lašelių masei: 1 μs 2,5 ng; 3 μs 16 ng; 12 μs 65 ng.
- 8. Skaitinis eksperimentas parodė, kad, siekiant padidinti spausdinimo greitį ir atspaudo kokybę, atsižvelgiant į spausdinamo vaizdo skiriamąją gebą, galima naudoti skirtingus išpurškimo parametrus. Tuo atveju, kai reikalingas aukštos raiškos atspaudas, rekomenduojama nustatyti nedidelį išpurškimo laiką, kad būtų formuojami maži rašalo lašeliai be kenksmingų juos lydinčių palydovų. Jei nereikalaujama aukštos raiškos atspaudo ir reikia padengti storą to paties rašalo sluoksnį, galima nustatyti ilgesnį išpurškimo laiką.

Tim TOFAN

INVESTIGATION OF THE INKJET PRINTING PROCESS, EVALUATING THE DYNAMICS OF THE INK AND THE PROPERTIES OF SPECIFIC PRINTING SURFACES

**Doctoral Dissertation** 

Technological Sciences, Mechanical Engineering (T 009)

RAŠALINIO SPAUSDINIMO PROCESO TYRIMAS, ĮVERTINANT RAŠALO DINAMIKĄ IR SPECIFINIŲ SPAUSDINIMO PAVIRŠIŲ SAVYBES

Daktaro disertacija

Technologijos mokslai, Mechanikos inžinerija (T 009)

Lietuvių kalbos redaktorė Dalia Markevičiūtė Anglų kalbos redaktorė Jūratė Griškėnaitė

2023 11 17. 11,5 sp. l.. Tiražas 20 egz. Leidinio el. versija *https://doi.org/10.20334/2022-047-M* Vilniaus Gedimino technikos universitetas Saulėtekio al. 11, 10223 Vilnius Spausdino UAB "Ciklonas" Žirmūnų g. 68, 09124 Vilnius