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Study and Simulation of Droplet Formation and Nozzle Design for Graphene Inkjet Printing

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***Abstract*—Conductive inks made of graphene are widely used in printed electronics. Considerable research interest is now focused on graphene. Nevertheless, graphene faces disadvantages such as stability, dispersion, and the tradeoff between conductivity and transparency. This paper deals with graphene ink characteristics with simulation using a software that mimics inkjet printing technology. This printing inkjet facilitates the realization of several film characteristics, including pattern shape, pattern placement, film thickness and conductivity. Graphene is primarily prepared from oxides of graphene/graphite and reduction methods using thermal, microwave, chemical and laser reduction are explored. Recent studies have revealed that the energy storage devices made using graphene hybrids and nanocomposites have improved efficiency. In this article, the application of graphene and its derivatives are speculated and helps in creating visions for added advance of graphene-based materials.**

***Index Terms*—graphene, graphene ink, inkjet-printing.**

I. INTRODUCTION

A. Inkjet printing and graphene

Inkjet printing is a non-contact approach of producing droplets from an ink reservoir with minimal human participation and directly deposit the amount of materials required from a computer-designed picture on a specified substrate [1]. As far as the type of substrate is concerned, there are no limitations. The support might be flexible or stiff. It is often seen as a cost-effective and versatile approach of micro and nano-manufacturing utilizing functional materials. Inkjet printing has been so powerful that the process is used to print various functional materials such as conductive tracks, Light Emitting Diodes and even tridimensional structures [2]. Large number of scientific publications and conferences are taking place every year in this active field. Recently, scientific papers focusing on the inkjet printing of functional materials and properties of the printed patterns has amplified [3].

The long-term use of fossil fuels has accounted for the increase in greenhouse effect, environmental pollution and global warming. Focus recently has enlarged on power conversion and storage systems including fuel cells, solar cells, supercapacitors and batteries, to offset significant carbon emissions.Batteries are subjected to increased research since their use in portable devices. Lithium- ion batteries have been extensively used nowadays because of its high energy density and high storage volume [4]. Carbon-based materials play critical roles in energy storage in lithium-ion batteries [5]. Graphite, duetoits electrochemical and physical properties is extensively usedastheanodematerialinthelithium-ionbatteries[6]. But, traditional graphite materials show low storage capacity due to partial Li-ion storage sites in the carbon structure. Considerable effort has been made to explore new anodematerials to improve the energy density, storage capacity and new anode materials. Graphene which exhibits novel properties was discovered in 2004 [7]. Graphene, with its large surface-to-volume ratio and highly conductive nature, may potentially provide high Li storage capacity to Li-ion batteries. The capacity is expected to increase up to 500–1100 mAhg−1 if graphene is used instead of graphite [7].

B. Synthesis of graphene

Two types of approaches used in graphene synthesis are:

1. Top-downapproach
2. Bottom-upapproach

Top-down approach comprise the disintegration of stacked graphite layers into single graphene sheets. This approach helps to overcome the Vander Waals forces holdingthegraphite layers.Effective separation of the layers without damage and preventing re-agglomeration are the main challenges in using the top-down approach [8]. The separation of graphene from alternative carbon sources is the bottom-up approach. The approach also supports the formation of large-area graphene films. Graphene synthesized from the exfoliation of graphite oxide is referred as functional graphene as complete reduction is not achieved yet[8].

Feixiang Liu et al (2019) have portrayed the synthesis of high- quality graphene based conductive inks on various substrates using various methods [9]. RajeshKumaretal(2019)discussed the synthesis of graphene using reduction approach involving thermal, chemical and laser methods [10]. Rodriguez-Perez Laura et al (2013) clearly reviewed the advantages of graphene, difficulties in the synthesis of graphene and the chemistry behind its wonderful electrical and thermal properties [11]. AndriiTencha et al (2017) have explained the synthesis of two-dimensional graphene oxide using Hummers method for the application of printed electronics [12].

C. Inkjet Printing of Graphene

Yanfei Xu et al (2013) and Naik et al (2018) have done investigation on the preparation of thin-film electrodes using graphene/polyaniline inks for inkjet printing. Two-Electrode supercapacitors are printed and its electrochemical measurements are taken and tested for over 1000 cycles [13,14]. Marcello Romagnoli et al (2016) have described the challenges of printing of graphite ink using a 20μm nozzle in a thermal DOD inkjet printer. The authors also explainedthe influence of factors such as density,viscosity and surface tension on thejettability and droplet stability [15]. Guohua Hu et al (2018) have reviewedtheimportanceofgrapheneandotherrelated2D materials in the field of Charlie toaccelerateadditive patterning on stiff and conformable substratessupporting the design of flexibledevices in large scale [16]. Stephen Lawes et al (2015) have clearly depicted the printing techniques to print carbon nano-materials. Thechallengesandfuturescopeof these printed carbon materials were also discussed [17].

D. Rheological Characteristics of Carbon-Based Materials

Charlie O' Mahony et al (2019) and Ozkan et al (2016) have reviewed the market movement towards eliminating waste and optimization of additive manufacturing. They have also explored the relationship between the rheology of inks and the steps to maintain the properties [18,19]. Francesco Bonaccorso et al. (2016) examined the liquid phase production of graphene, 2D crystals and heterostructures, as well as the capacity to formulate functional inks with rheological for printing processes [20].

E.Applications of Carbon-Based Materials in Printed Electronics

Urooj Kamran et al (2019), Capasso et al (2015) and Paul et al (2012) expressed the applications of graphene and carbon nanotubes in future electronic devices and also proposed a profound study on the applications of carbon materials in flexible electronics, energy devices, etc. [21-23]. RebeccaS.Edwardsetal(2013)intheir researchhave clearly portrayed the importance and amalgamation of graphene and its applications, including composites, batteries, etc. [24]. The current research survey indicated the methods used in the synthesis of graphene, their mechanical, chemical, thermal properties and the application of graphene. The parameters for the simulation and the properties of graphene were extracted from the studies discussed in Section I.

II. NUMERICAL SIMUALTION AND ROCESS PARAMETERS

The process parameters are taken on the basis of studies discussed previously and these values are employed in the FLOW 3D software to get the simulation result of the ink drop let from the nozzle tip diameters of 20 and 40 micrometers as shown in Table 1 and Table 2.

Table I. Parameters of InkjetPrinter Nozzle (20 μm)

Name	Parameters	Value (m)
Subcomponent 1: Cylinder	Radius	1.05e-04
Subcomponent 2: Torus	Minor radius	9.5e-05
	Major radius	1.05e-04
Subcomponent 3: Substrate	Radius	1.05e-04
Subcomponent: Piezo	Radius	9.5e-05

Table II. Parameters of InkjetPrinter Nozzle (40 μm)

Name	Parameters	Value (m)
Subcomponent 1: Cylinder	Radius	1.05e-04
Subcomponent 2: Torus	Minor radius	9.5e-05
	Major radius	1.05e-04
Subcomponent 3: Substrate	Radius	1.05e-04
Subcomponent: Piezo	Radius	8.5e-05

III. MODELLING AND ANALYSIS

The scientific methods which are equipped in the analytical software and reason behind the choosing of cylindrical mesh for this analysis are gave in detail manner for the better understanding.

A.Design module

In this design module, a nozzle for inkjet printing is designed with a nozzle diameter of 20 and 40 micrometer are shown in Fig. 1 and Fig. 2. This design model solves in three- dimensional for a better understanding of the flow of graphene from the nozzle tip to the substrate. For different nozzle tip diameters, the flow of graphene from the nozzle tip is also different because of graphene's physical properties viz., viscosity, surface tension and other parameters.

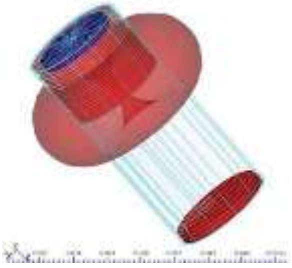

Fig1. Inkjet Nozzle Diameter 20μm

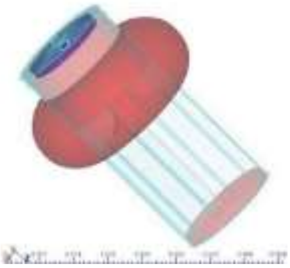

Fig. 2 Inkjet Nozzle Diameter 40µm

The above three-dimensional models consist of a torus, cylinder, and substrate which can be designed into the nozzle by the Boolean operation. The centre of the torus is where the graphene ink is given through fluid initialization in Flow 3D software.

B.Fractional Area Volume Obstacle Representation (FAVOR)

Fractional Area Volume Obstacle Representation (FAVOR) method uses the Volume of Fluid (VOF) method to create rectangular grids whose elements are assigned with fractional areas and volumes. The VOF method works on three rules: a structure to locate the surface, a process to track the surface and a means of applying surface boundary conditions.

The cylindrical mesh is used because of its uniform circular section as shown in Fig. 3 and Fig. 4. The free gridding method is used and it consists of a simple rectangular mesh with variable spacing controlled by a few parameters. In FAVOR technique, which is unique to FLOW-3D, the portions of element surfaces and volumes blocked by obstacles are computed and stored. The computations are done entirely by the pre-processor and require no user interaction. The main advantage of this independent gridding technique is to modify the geometry without changing the grid. The grid can be refined and other changes can be made without affecting the geometric model.

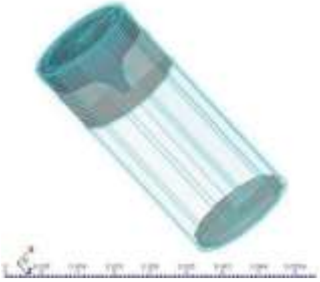

Fig 3. Inkjet Nozzle Diameter (Mesh) 20µm

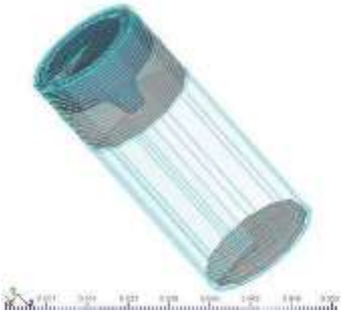

Fig 4. Inkjet Nozzle Diameter (Mesh) 40µm

IV. SIMULATION AND RESULTS

A.Properties of Fluid

The properties of graphene, which are very important for this simulation and these properties are taken from [25].

Table III. Graphene ink properties

Material name	Graphene
Density	0.94 g/cm^3
Viscosity	9.4 mPa.s
Surface tension	33 mN/m
Contact angle	0 degrees

B.Velocity of piezo material

The velocity of piezo material is given to the piston by giving prescribed motion while designing. The prescribed motion is given to the piezo material by selecting the simple movingobjectfromthecomponentproperties,sub-menuand then given the co-ordinates of time vs Z-velocity graph as shown in Fig.5.

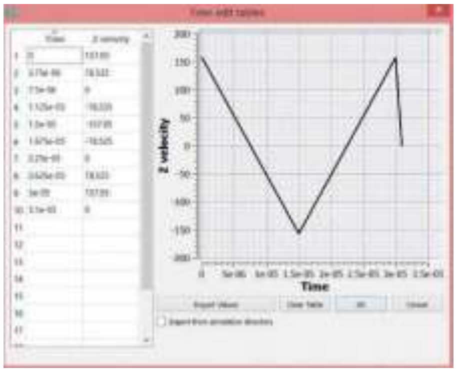

Fig 5. Velocity profile of piezo material

C. Boundary conditions

The boundary conditions for the inkjet printer to print the graphene ink. The boundary conditions on the volume fraction are set as a constant contact angle, here which is zero.

- Inlet – Velocity inlet (Piston driven)
- Outlet – Pressure outlet

D. *Simulated Fluid Flow from Nozzle Tip Diameter of 20 µm*

The simulation shows the ejection and spread of the graphene ink from the nozzle tip to the substrate. The following figures are shown that the ejection of graphene ink from the nozzle with the velocity profile as shown in Fig. 5. From the Fig. 6a, 6b and 6c, we can infer that the pressure values are very high and the spread of graphene ink onto the substrate is not uniform.

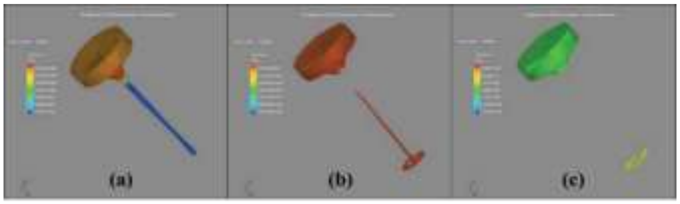

Fig 6. Fluid flow from the nozzle (20 µm)

E. *Simulated Fluid Flow from Nozzle Tip Diameter of 40 µm*

The flow of graphene from the 40 µm nozzle tip is uniform and the droplet is spread uniformly on the substrate. The following figures are shown that the ejection of graphene ink from the nozzle with the velocity profile as shown in Fig. 5. From the Fig. 7a, 7b, and 7c, we can infer that the pressure values are nearly uniform throughout the process and the spread of graphene ink onto the substrate is uniform.

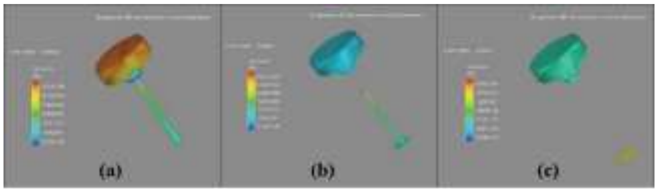

Fig 7. Fluid flow from the nozzle (40 µm)

F. *Pressure Graph for the Nozzle Tip Diameter of 20 µm and 40 µm*

The variation of pressure of graphene ink with respect to distance from the nozzle tip diameter of 20 micrometres and 40 micrometres to the substrate is shown in Fig. 8a and Fig. 8b.

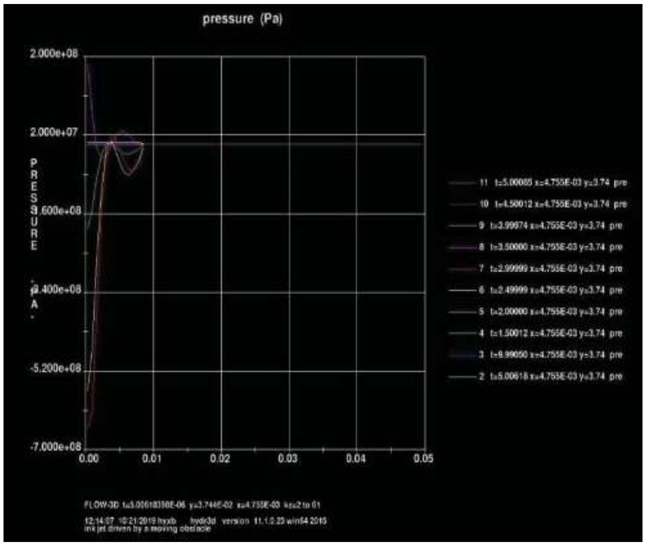

Fig 8a. Pressure of graphene vs Distance from the nozzle (20 µm)

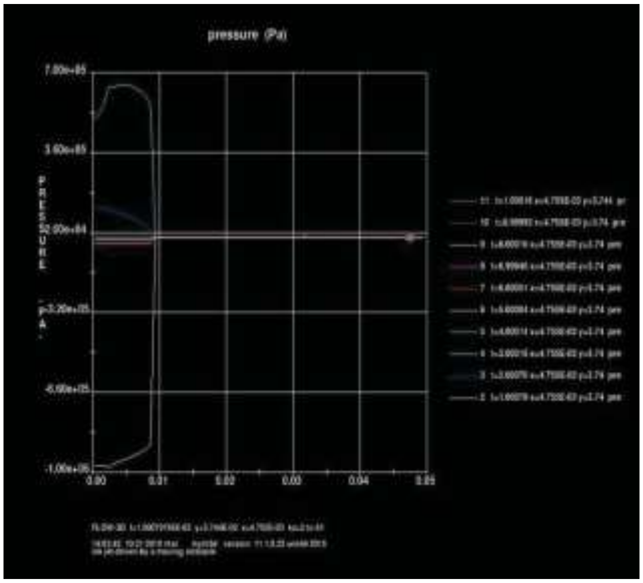

Fig 8b. Pressure of graphene vs Distance from the nozzle (40 µm)

G. *Velocity Graph for the Nozzle Tip Diameter of 20 µm and 40 µm*

The variation of velocity of graphene ink with respect to distance from the nozzle tip diameter of 20 micrometers and 40 micrometers to the substrate is shown in Fig. 9a and Fig. 9b.

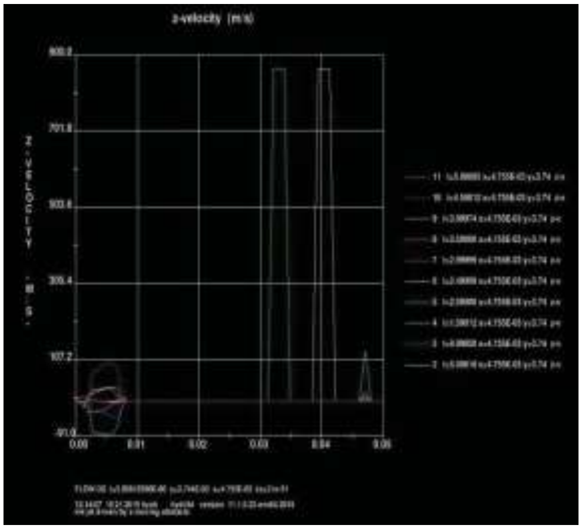

Fig 9a. Velocity of graphene vs Distance from the nozzle (20 µm)

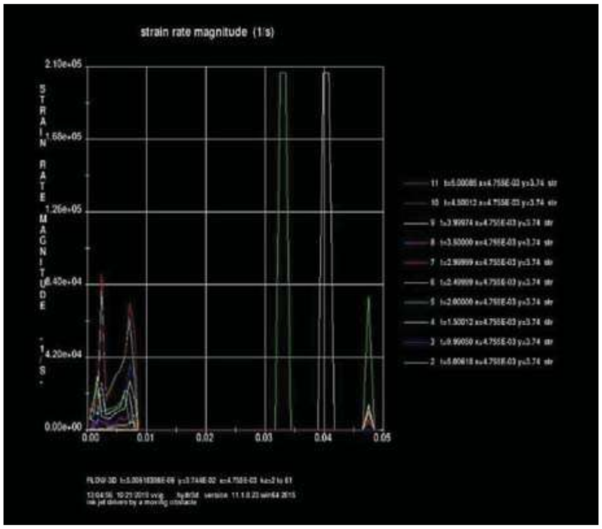

Fig 10a. Strain rate of graphene vs Distance from the nozzle (20 µm)

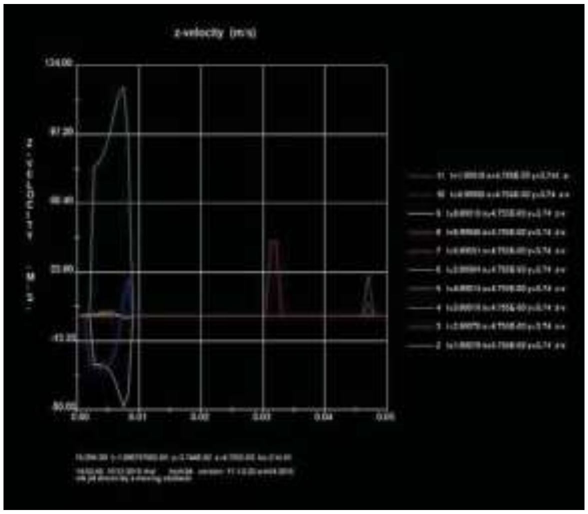

Fig 9b. Velocity of graphene vs Distance from the nozzle (40 µm)

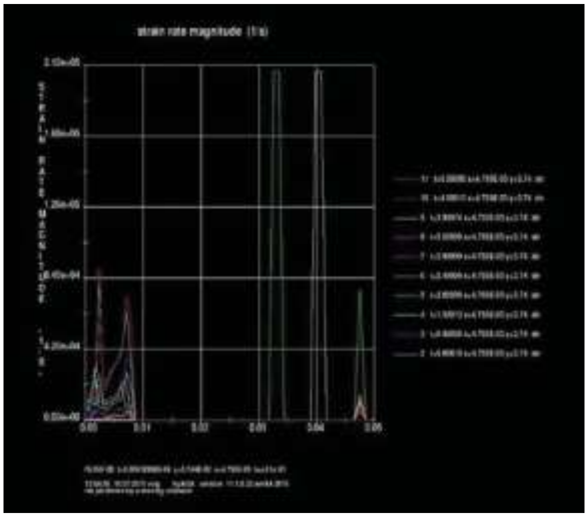

Fig 10b. Strain rate of graphene vs Distance from the nozzle (40 µm)

H. Strain rate magnitude graph for the nozzle tip diameter of 20 µm and 40 µm

The variation of strain rate of graphene ink with respect distance from the nozzle tip diameter of 20 micrometers and 40 micrometers to the substrate is shown in Fig. 10a and Fig. 10b.

V. CONCLUSIONS

In this research work, the inkjet nozzles of 20 and 40 micrometer were designed to print the graphene ink onto to substrates. The simulation results demonstrated that the spread of graphene ink on the substrate was affected on the basis of the nozzle size of the inkjet while the velocity graph of piezo-electric element is constant for the both inkjet nozzles. From the results as we get, the spread of graphene ink onto the substrate is uniform for 40 micrometers inkjet nozzle. Since the drop spread is uniform onto the substrate we might be able to 3D-print the sub-components of the product. Since the lots of research is going on in graphene towards the energy storage and replacing the graphite in Li-Ion battery with this graphene. We might be able to print the graphene-based batteries for the applications in the electric vehicle’s battery.

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