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Development of new formulation for soft material in paste extrusion-based 3D Printer

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Abstract. Fused Deposition Modelling is a form of additive manufacturing where solid filament is heated into molten state and deposited onto a heating platform to create three-dimensional objects layer-by-layer. Since heating and cooling processes are involved in Fused Deposition Modelling (FDM), this restricts the use of thermoplastic polymers such Room Temperature Vulcanizing (RTV) silicones and gels. Others concern are with the right rheological properties for extrusion and the ability to provide desired mechanical qualities upon quick solidification. To develop a suitable silicone printing technology, it is crucial to understand the silicone polymerization mechanism in terms of its rheological and mechanical characteristics. Due to the numerous factors that can influence silicone paste mixtures, this study utilized the Taguchi method to design experiments, optimize factors, and predict properties, thereby avoiding extensive and resource-intensive experimental work. The study specifically considered the factors of curing method, mass ratio of silicone thinner, and fumed-silica in the silicone paste formulation. Among the 9 samples generated through the Taguchi method, only one sample demonstrated favourable results in terms of mechanical properties and the curing process, with a mixture ratio of base silicone, silicone thinner, and fumed-silica at 15g, <1g, 0.1g; respectively. However, further investigation into the fixed amount of silicone and fumed silica in the selected mixture ratio indicate that the amount of silicone thinner must be less than 1g or can be omitted as the silicone paste can be used as printing ink for the extruder. The shore hardness testing for the silicone samples revealed with zero percent silicone thinner exhibited a hardness value of 21.5 HA (Shore A) while the sample obtained less than 1g displayed a significantly lower hardness value of 10.5 HA. These findings indicate that the addition of silicone thinner in the silicone paste formulation, as optimized through the Taguchi method, contributed to a reduction in the hardness of the material for shore A scale. This suggests that the presence of silicone thinner affects the elasticity and flexibility of the silicone paste, resulting in a lower Shore hardness value. The discrepancy in hardness values between the samples further highlights the significance of formulation optimization to achieve desired material properties for silicone paste-based applications in 3D printing.

1. Introduction

Three-dimensional object produced via additive manufacturing using CAD model or digital 3D model is often known as 3D printing. 3D printing technologies are gaining momentum in a variety of industries,



including aerospace and machine manufacturing. Numerous facets of society have benefited from the use of additive manufacturing, including manufacturing, aerospace, automotive, electronics, robotics, healthcare, prosthetics, fashion, general commerce, food processing and education [1-3]. Different medium or printer inks are used by different types of additive manufacturing techniques. Additive manufacturing is classified into seven types by ASTM International: binder jetting, directed energy deposition, material extrusion, material jetting, powder bed fusion, sheet lamination, and vat photopolymerization. Extrusion-based AM is simpler to set up than liquid- and powder-based AM techniques, especially in domestic conditions, and requires less equipment and energy. CAD model can be created from one of three techniques which are modelling software, reverse engineering laser scanning techniques like photogrammetry or computed tomography, or light-based 3D laser scanning and reverse engineering. These CAD models are then converted into the standard triangle language (STL) file format using slicing software. After that, the STL files are sent to the 3D printer where printing parameters such as layer thickness and printing temperature are configured. Finally, the printed 3D objects are removed from platform adhesion and any supporting structures that may have been there. Post-processing may be used as the final stage to solve issues like anisotropy and unacceptable surface quality.

This has led to the adoption of this technology by a wide range of users, from amateurs to significant manufacturers in several industrial areas. The desired product is created by fusing layers upon layers of plastic or metal with an additive manufacturing technique. Comparing 3D printing to normal manufacturing methods, the key benefits are its speed, design flexibility, testability, and cost savings. The usage of 3D printing is superior to conventional industrial processes for small-scale production, prototyping, small enterprises, and educational purposes. Despite how far additive manufacturing has come, there are still many ways to expand the technology.

Silicone is a polymer that is extensively utilised in the healthcare, aerospace, and textile industries. Silicone is a particularly desirable material for manufacturing due to its unique properties of high flexibility and resistance to heat and stress. Silicones are frequently utilised in many technical fields because of their distinctive qualities, such as chemical stability, high thermal consistency, optical clarity, and high oxygen absorption. They are primarily formed from $(\text{Me}_2\text{SiO})_n$ polymers [4]. Due to their unique qualities, silicone elastomers are of economic importance in a variety of fields. Using lower viscosity inks could help solve some of the problems caused by high viscosity materials, such as the difficulty in combining and switching inks to construct devices made from multiple types of silicone.

One of the limitations of the technique is that RTV silicones and gels are not suitable for FDM. Direct Ink Writing (DIW) is a further technique that is widely used for silicone and hydrogel printing. However, it is challenging to produce a customised ink that has the right rheological properties for extrusion and that can provide the required mechanical qualities after quick solidification. In order to develop a suitable silicone printing technology, it is crucial to understand the silicone polymerization mechanism in terms of its rheological and mechanical characteristics [5]. Additionally, the appropriate shore hardness of the silicone must be established in order to modify the shore hardness to meet user requirements. On top of that, the silicone for paste extrusion-based 3D printing is not a single syringe dispenser friendly. In terms of silicone, it is typically non-reactive, stable, and resistant to extreme environments and temperatures. However, during printing, the silicone paste may eventually solidify in the syringe [1].

The driving wheels used in filament-based material extrusion are substituted in this type of extrusion-based AM with a motor-driven plunger or syringe to help in flow of material. This approach is utilised with feedstocks suitable for printing, such as pastes and AM substances with low melting point and fluid properties. The syringe or plunger, respectively, forces the fluid-like substance and feedstock material out of the nozzle and onto the building platform. The two plunger-based extrusion AM techniques most frequently used are robocasting and DIW [6]. A screw extruder has multiple zones. By friction and heat, materials are transported from the delivery zone to the melting zone and melted. The soft material is then forced out and placed in the metering zone using a rotating screw and high pressure. Compared to plunger and filament-based processes, controlling the flow of the material can be more challenging with

screw-based AM extrusion. In order to guarantee that the material is of a fixed size and to maintain a consistent material flow, design modifications may thus be required [6, 7].

2. Materials and Methods

Figure 1 shows the process for flow chart of the overall from experimental. This flow chart elaborates the experimental work to be done. This experimental consists of 3 stages which are material selection for stage 1, sample preparation for stage 2 and sample testing for stage 3. The most important stages are the material selection because this stage need to do more research to develop a soft material with right combination of material that suitable for paste extrusion-based 3D. When satisfied with the material selection of soft material, the sample preparation can be set up by creating the G code and identify the suitable printing parameters. After that the 3D sample will be test using Elastomer Tensile Strength Testing to analyse the mechanical properties.

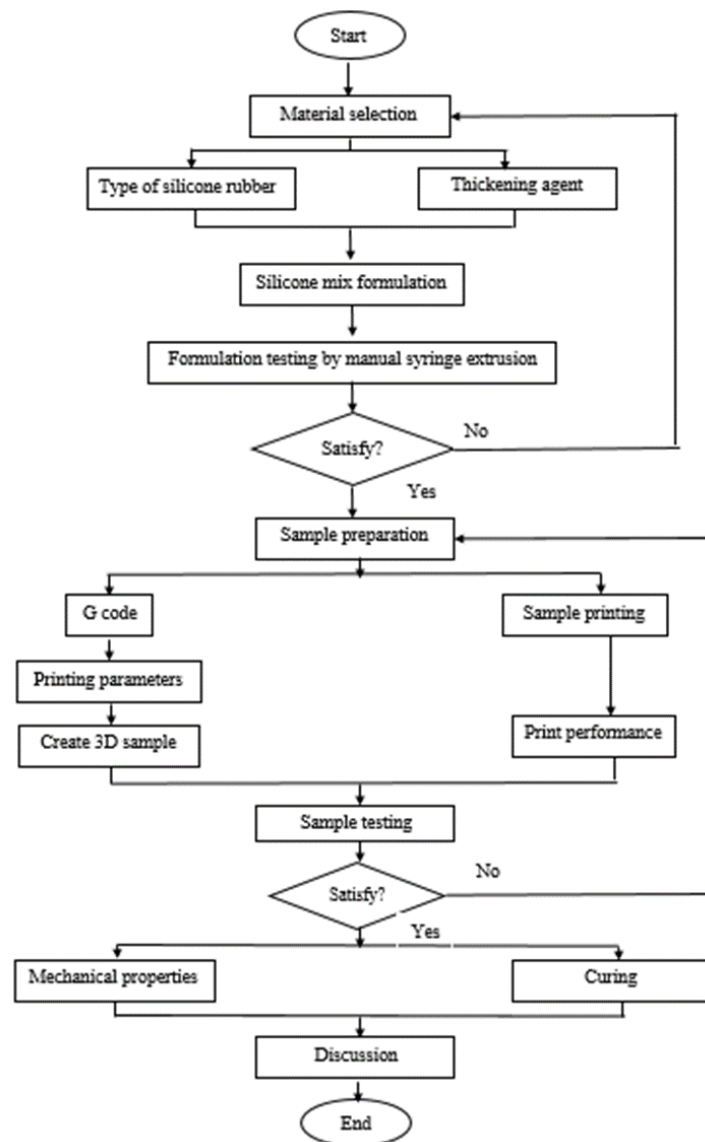


Figure 1. The flow chart for overall experiment.

The key component of the experiment is silicone elastomer or liquid silicone rubber. Long polysiloxane chains are strengthened with chemically treated silica in the two-component method known as liquid silicone rubber. The two components, known as Parts A and B, include a platinum catalyst in Part A and methylhydrogensiloxane in Part B, which also serves as a cross-linker and an alcohol inhibitor. The benefits of utilising liquid silicone rubber are that it has highly compatible qualities that allow it to be used for components that come into contact with humans and for safe physical interaction with humans. In addition, it can tolerate of temperatures ranging from -60°C to $+250^{\circ}\text{C}$ while its excellent mechanical capabilities are consistently maintained. This ensures long-term stability and chemical resistance.

The Taguchi method can be used to optimise the 3D printing process's utilisation of silicone and Thi-vex. To get the desired printing outcomes, it is possible to carefully analyse the impacts of many elements and their interactions by using this robust experimental design approach. The Taguchi approach as shown in Table 1 involves carrying out a series of carefully monitored tests in which the concentrations of silicones, Thi-vex and fumed silica, are changed in each trial. These experiments are made to efficiently collect data and identify the most suitable combination of variables that produce the intended result.

Table 1: L9 orthogonal array Taguchi experimental design matrix.

Samples	Silicone (A & B) (g)	Thinner (g)	Fumed Silica (g)
1	15	3	0.5
2	15	1	0.3
3	15	<1	0.1
4	10	3	0.3
5	10	1	0.1
6	10	<1	0.5
7	8	3	0.1
8	8	1	0.5
9	8	<1	0.3









3. Results and Discussion

Taguchi method results showed that Sample 3 had the best results for all response variables. This specific sample showed great printability, producing accurate and precise prints. It was found that the smooth surface quality improved the overall appearance of the printed goods. Sample 3 displayed outstanding elasticity, demonstrating that the silicone, silicone thinner, and fumed-silica combination ratio utilised in this sample produced a silicone paste with desired elastic qualities. In a study conducted by S. Walker *et al.*, silicone mixture ratio of (Part A) 44.5 g of base silicone, 0.5 g of silicone thinner, and 1.0 g of Urefil-9, and (Part B) 44.5 g of base silicone, 0.5 g of silicone thinner, and 1.0 g of Urefil-9 was combined for the purpose of studying the curing effect and conducting peel testing [6]. This article verifies that the silicone preparation, achieved through the correct combination of silicone thinner and fumed-silica, exhibits favourable mechanical and rheological properties, making it suitable for sample testing and be used in 3D printing.

The purpose of adding various silicone thinner concentrations into silicone paste for 3D printing is to examine the impact of thinning agent concentration on the printing process and the final qualities of the printed objects. Surprisingly, from the experiment of effects of thinner on silicone results proved that the silicone paste formulation with 0% silicone thinner still generated 3D printed objects of equal quality to Sample 3. The mechanical strength, surface quality, or printability of the printed products

were unaffected by the lack of silicone thinner. Without silicone thinner, the silicone paste's printability was still outstanding, enabling accurate and precise printing. The material's flow characteristics were unaffected by the absence of silicone thinner, and it retained enough viscosity to ensure adequate deposition during printing. Additionally, the prints' surface quality remained excellent and free of flaws, showing that the absence of silicone thinner had no negative effects on the material's capacity to adapt to intricate geometries or preserve fine details.





Table 2: Results and observations of 9 sample of silicone paste using Taguchi method.

No of exp.	Mixture	Printed	Observation
1			<ul style="list-style-type: none"> • Good 3D shape during printing • Easy to extrude • The wall sample collapsed • Moist texture • Fail to cure
2			<ul style="list-style-type: none"> • Good 3D shape during printing • Easy to extrude • The wall sample collapsed • Moist texture • Fail to cure
3			<ul style="list-style-type: none"> • Good 3D shape during printing • Need pressure to extrude • The two layer stick properly, and the wall not collapse • Elastic characteristic • Less than 24 hours to cure
4			<ul style="list-style-type: none"> • Good 3D shape during printing • Easy to extrude • The wall sample collapsed • Moist texture • Fail to cure

5		<ul style="list-style-type: none"> • Good 3D shape during printing • Easy to extrude • The wall sample collapsed • Moist texture • Fail to cure
6		<ul style="list-style-type: none"> • Good 3D shape during printing • Need pressure to extrude • The wall sample not collapse • Grease texture and behaviour like rotten rubber band • Fail to cure
7		<ul style="list-style-type: none"> • Cannot form 3D shape during printing • Easy to extrude • The wall sample collapsed • Moist texture • Fail to cure
8		<ul style="list-style-type: none"> • Good 3D shape during printing • Need pressure to extrude • The wall sample collapsed • Grease texture • Fail to cure
9		<ul style="list-style-type: none"> • Good 3D shape during printing • Need pressure to extrude • The wall sample not collapse • Grease texture and behaviour like rotten rubber band • Fail to cure

In order to examine the impact of thinning agent concentration on the printing process and the final qualities of the printed objects, various amount of silicone thinner concentrations was added into silicone paste. Surprisingly, from Table 3 the outcomes proved that the silicone paste formulation with 0% silicone thinner still generate 3D printed objects of equal quality to Sample 3. The surface quality, or printability of the printed products were unaffected by the lack of silicone thinner. However, the results for shore hardness testing revealed that the sample with zero percent silicone thinner exhibited a hardness value of 21.5 HA (Shore A). In contrast, the sample obtained from the Taguchi method, specifically Sample 3, displayed a significantly lower hardness value of 10.5 HA (Shore A). These findings indicate that the addition of silicone thinner in the silicone paste formulation, as optimized through the Taguchi method, contributed to a reduction in the hardness of the material.

Table 3: Sample with varying thinner to fixed silicone and fumed silica.

Amount of thinner (droplets)	Sample with fixed amount of silicone and fumed silica	Observation
0		<ul style="list-style-type: none"> • Able to cure • Double line layer can stick properly • Elastic • Wall did not collapse
1		<ul style="list-style-type: none"> • Fail to cure • Less viscous • Moist texture
3		<ul style="list-style-type: none"> • Fail to cure • Behaviour like plasticine • Good 3D shape
5		<ul style="list-style-type: none"> • Fail to cure • Behaviour like plasticine • Good 3D shape

4. Conclusion and recommendations.

In this study, an inexpensive custom-made 3D printing ink is developed using silicone rubber for paste extrusion-based 3D printer. The elasticity of the silicone paste was improved through addition of silicone thinner and fumed-silica and increasing the concentration of crosslinker to give the silicone printed have similar mechanical properties to other commercially available products. The experiment on the effect of silicone thinner in silicone also revealed that the silicone paste can still generate 3D printed objects with the required mechanical properties; however, this does not mean that the silicone thinner is not important in this formulation of silicone paste extrusion because altering the silicone thinner can produce various types of silicone paste in terms of rheological and mechanical properties. The silicone paste formulation with 0% silicone thinner still generated 3D printed objects of equal quality to Sample 3. The mechanical strength, surface quality, or printability of the printed products were unaffected by the lack of silicone thinner.

Without silicone thinner, the silicone paste's printability was still outstanding, enabling accurate and precise printing. The material's flow characteristics were unaffected by the absence of silicone thinner, and it retained enough viscosity to ensure adequate deposition during printing. Additionally, the prints' surface quality remained excellent and free of flaws, showing that the absence of silicone thinner had no negative effects on the material's capacity to adapt to intricate geometries or preserve fine details. According to these findings, the formulation without silicone thinner can still produce good printing results that are comparable to the results of Sample 3, indicating the possibility of a cost-saving measure in the production of silicone paste. The performance and the mechanical properties of this recently develop silicone paste formulation can be further investigated through the manufacturing and production for silicone paste-based applications in 3D printing.

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