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Dynamic Mechanical Properties of Copper-ABS Composites for FDM Feedstock

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ABSTRACT

This paper presents an investigation of dynamic mechanical properties of a copper-ABS composite material for possible fused deposition modeling (FDM) feedstock. The material consists of copper powder filled in an acrylonitrile butadiene styrene (ABS). The detailed formulations of compounding ratio by volume percentage (vol. %) with various combinations of the new polymer matrix composite (PMC) are investigated experimentally. Based on the result obtained, it was found that, increment by vol. % of copper filler ABS affected the storage modules (E') and tan δ results. The storage modulus and tangent delta increased proportionally with increment of copper filled ABS. It can be observed that, the storage modulus and tangent delta are dependent on the copper filled ABS in the PMC material.

Keywords - *Polymer Matrix Composites, Dynamic Mechanical Properties, Storage Modulus, Loss Modulus, Tan δ, Fused Deposition Modelling.*

I. INTRODUCTION

Rapid prototyping (RP) technology is a new technique for part fabrication in layers by a layer process. Normally, previous RP applications focused on build a final product for fitting and testing. Customer needs for the end used product and continued demand for low-cost and time saving have generated a renewed interest in RP. A shift from prototyping to manufacturing of the final product will give an alternative selection with different material choice, low cost part fabrication and achieving the necessary mechanical properties. One of the RP technologies available today is Fused Deposition Modelling (FDM), which involves extrusion of plastic filament wire as feedstock material. Existing FDM machine have been able to deposit only thermoplastic filament with limited mechanical properties through the liquefied nozzle. Fig. 1 shows the schematic diagram of the FDM filament through heated liquefied nozzle [10]. The FDM machine can process acrylonitrile butadiene styrene (ABS) plastic material [4,9,10,11,19,21,22,23,24] and Nylon [16,17] material. Other material and polymer matrix composites (PMC) used in FDM are ABS-Iron [9,10,16,17,26], Aluminum epoxy [15] and stainless steel-ECG2[25]. Meanwhile, some researchers focused on the optimal process parameters by Taguchi method [4,13,21,22,24], improved FDM product [22,23,25], mixing torque [9], thermal conductivity [11, 16], deposition rate [19,23] geometrical modeling and mathematical model [20], [23,24] and mechanical properties [16,21,22,23,26]. Another manufacturing process for PMC is injection moulding. In various research work, the composites material used in injection moulding are basalt-LDPE [1], iron-HDPE [2,14], iron, nickel-HDPE [3], zirconia-PE [5], copper-PE [6], stainless steel-PE [7,14], stainless steel-PEG, PMMA [8,2], iron-PP, EVA [14] and stainless steel-EVA [18]. Wu et al. [25] have investigated the development of stainless steel-ECG2 ceramic composite for time and cost saving using Fused Deposition Ceramic (FDC). It was mentioned that, the compounding is a very critical process to provide homogenous powder polymer mixture of stainless steel filament. The fabrication of ABS-Iron filament wire for FDM machine has been done by Masood et al. [16,17] and Nikzad et al. [9,10] with proper formulation and mixing processes. They mentioned that the critical properties requirements for high-quality composite are depending on the desired viscosity, strength and modulus. High iron powder loading in the Nylon will lead to dispersion issues, and the viscosity will increase with increment of metal powder in PMC material. Nikzad et al. [10,27] have investigated the thermo mechanical properties of new composite for rapid tooling in injection moulding application. It was mentioned that, the addition of 10% iron into ABS matrix increase the storage modulus up to 40% approximately. The tensile strength result drops proportionally with increment of 10% by volume of iron powder. From the DMA results obtained, it was found that with high concentration of copper (30% by vol. %) above glass transition temperature of ABS, the thermal conductivity increased proportionally. The conductively begins to form at only in 20 vol. % concentration of copper particles[10]. Nowadays, application of FDM material requires an enhanced and higher mechanical property toward rapid manufacturing. Pure metal is unsuitable for deposit through the heated liquefied head by FDM machine because of higher melting temperature and viscosity. Nevertheless, there are limited data/research available particularly dealing with the deposition of PMC through the heated liquefied nozzle. Layered of rapid deposition polymer composites (RDPC) with highly filled metal powder in the polymer matrix may offer the possibility of introducing new composite material in FDM.

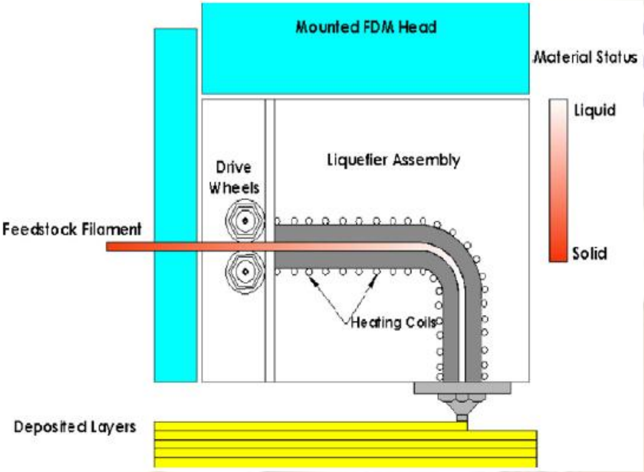

Figure 1 Schematic diagram of the FDM Filament through heated liquefied nozzle [10]

The aim of this study is to develop composite feedstock with copper, ABS and surfactant powder for FDM process. The research focuses on developing a proper formulation and mixture procedure of constituent materials for obtaining the homogeneous condition to produce the feedstock form. The main outcome from this study is to produce a strong, flexible, smooth and conductive feed stock to be produced by extrusion machine. The optimum composition of mixing ratio of constituent material was made by an injection moulding process for dynamic mechanical analysis. In this study, two main types of constituent materials were used to develop the new composite material. The first material, used for the matrix, is the ABS thermoplastic powder with lower melting temperature approximately 266°C, flexible and stiffness and suitable material for extrusion process. The copper particle was mixed with the ABS material as binder content to form the composite in Brabender mixer. The additive material is needed in order to control the melt flow behavior during an extrusion process. Results will be presented for dynamic mechanical properties, including storage modulus (E'), loss modulus (E") and Tan δ.

II. EXPERIMENTAL

2.1 Fabrication of FDM Feedstock

The fabrication of a new composite of FDM feedstock, required three types of the constituent material, which is a copper powder, ABS plastic and surfactant material. The ABS material is supplied by Dutatek Sdn. Bhd. (Selangor, Malaysia). The density of ABS was 1.03 g/cm^3 and melting temperature 266 ºC. ABS materials are an environmental friendly material because they are completely recyclable. Fig. 2 shows the ABS material for injection process. The copper powder is supplied by Saintifik Bersatu Sdn. Bhd. (Johor, Malaysia). The chemical composition is 99.9 % copper powder, and the particles size distribution is 17µm ~ 130 µm respectively with melting temperature 1080 °C and specific gravity 9.84 g/cm^3. Fig. 3 shows the copper material used in compounding process and Fig. 4 shows the SEM images of pure copper powder (300x) with different grain size distribution in a millimetre.

Figure 2 ABS Material

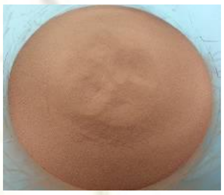

Figure 3 Copper Powder

The amount of each compounding composites is depending upon the volume percentage weight. In this experiment, the actual amount of compounding was determined and calculate by equation (1) following relationship[10]:

$$W_c = \frac{W_{cop} + W_{ABS}}{(1 - W_s\%)} \qquad (1)$$

Where;

Wc	Weight of composite material in grams
Wcop	Weight of copper powder in grams
WABS	Weight of ABS powder in grams
Ws%	Weight percentage (wt. %) of surfactant material

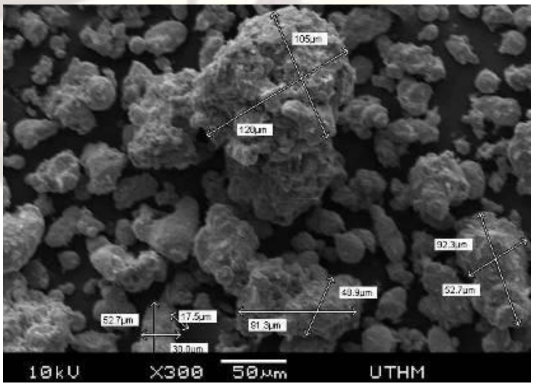

Figure 4 SEM Image of pure copper powder (300x)

The distributions of the copper powder, ABS and surfactant composition are 57% to 63% ABS, 22%

to 24 % copper powder and 15 % to 19% surfactant by volume percentage. A binder material based ABS plastic and surfactant (S) powder was added as the surfactant agent for smoothest flow of mixture of materials.

Firstly, ABS material was cut into the smallest size and approximately 2 mm - 5 mm in length. After that the ABS materials were then loaded in Brabender mixer, type W50 at 180 °C for compounding process in constant speed. In order to achieve a homogeneous mixture, the compounding procedure was followed. The maximum volume of the mixer is 55 cm^3 per mix approximately and one to three hour mixing time. Fig. 5 shows the (a) Brabender mixer, (b) Compounding process of ABS and copper materials. The PMC compounding were analyzed by scanning electron microscope(SEM) images to ensure a homogeneous compounding is acceptable for feedstock preparation. At the end, the surfactant was added into compounding to reduce the inter particle forces and to lubricate the powder.

Figure 5 (a) Brabender Mixer (b) Compounding process of Copper, ABS and surfactant Material

The compound was crushed by machine to form the pellets with 2 mm – 6 mm in size approximately. Fig. 6 shows the (a) PMC material after the Brabender mixer and (b) PMC material after crushing process. The feedstock pellet was injection moulded on a horizontal NP7-1F moulding machine for dynamic mechanical analysis (DMA) properties test specimens. Table I shows the composition of constituents of copper filled ABS composite material for five samples of PMC compounded with the different density value. The volume percentage ratio of copper, ABS and surfactant material are shown in Fig. 7

(a) (b)

Figure 6 (a) The PMC material after the brabender mixer (b) The PMC material after crushing process.

Due to the copper powder loading in the ABS matrix, the density of the composite increases proportionally with an increment of copper powder. An important issue in the extrusion process is the viscosity of the composite material to flow through the injection mould. In this study, two methods were used to determine the value of composite density. Firstly, the density of composite was measured by experiment and secondly by equation. In the experiment, the measurement of density was carried out by the Mettler Toledo apparatus. The theoretical density of the composite was calculated by equation (2) by Kate, 1987 [29].

$$dc = Vfdf + Vpdp \qquad (2)$$

where,

dc	Theoretical density of the composite
Vf	Volume percentage of filler copper
Vp	Volume percentage of ABS polymer
df	Density of filler copper
dp	Density of ABS polymer

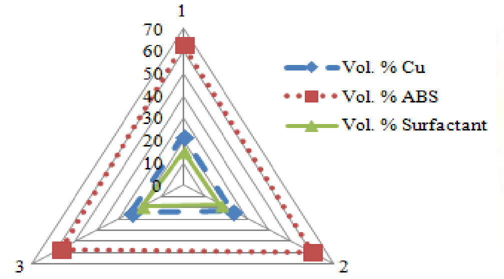

Figure 7 Distribution of volume percentage of copper, abs and surfactant materials

Table I. Constituents of copper filled abs composites material

Material	By Volume Percentage (vol. %)		Density by equation (g/cm^3)	Density by experimental (g/cm^3)
	ABS	Copper		
A	100	0	-	1.03
B	78	22	2.77	2.23
C	77	23	2.82	2.34
D	76	24	2.87	2.40
E	75	25	2.93	2.43
F	74	26	2.98	2.52

Figure 8 Injection molding machine [26]

2.2 The Specimen Fabrication by Injection Moulding

The injection moulding process was used to fabricate the specimens. The machine specifications are the screw diameter 19 mm, injection capacity 14 cm^3, injection rate 50 cm^3/second and injection pressure 161 MPa. Standard test specimens of DMA were prepared based on ISO 178 or ASTM D790. Due to the numerous type of dynamic mechanical instrument, specimen size is not fixed by this practice. The specimen size used was 80 mm x 10 mm x 3.2 mm in the analysis. Zone temperatures consist of five areas, where the nozzle temperature was 170 ^{o}C, front and middle were set to 175 ^{o}C and 165 ^{o}C, while for rear 2 and rear 1 was 130 ^{o}C and 160 ^{o}C. The feeding temperatures were set to 70 ^{o}C with the cooling time is10 second. In order to prevent the melt from sticking at the screw, the barrel temperature was used from low to high temperature. Fig. 8 and Fig. 9 shows, the injection moulding machine and the zone temperature in injection moulding machine.

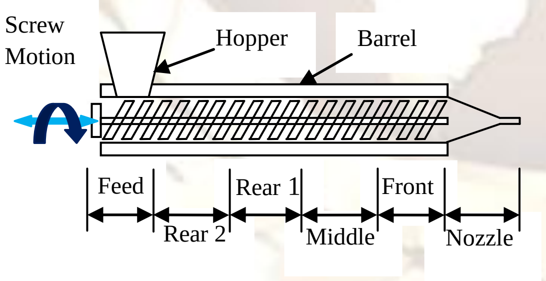

Figure 9 Zone temperature in injection molding machine[26].

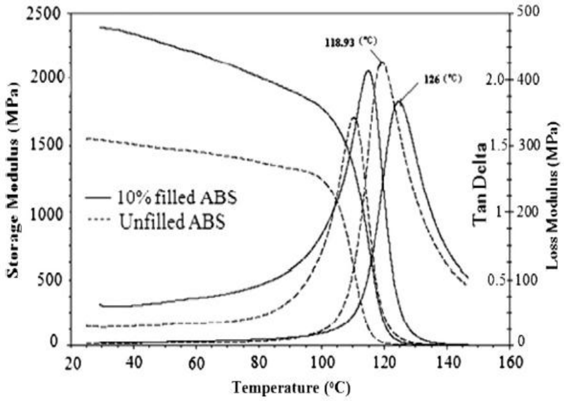

Figure 10 Comparison of dynamic mechanical properties of virgin ABS and 10% iron-powder filled ABS [10]

2.3 Dynamic Mechanical Analysis (DMA)

DMA is a technique used to measure the mechanical properties of an elastic, inelastic and viscous material. Normally, DMA works in the linear viscoelastic range, and it is more sensitive to structure. In dynamic mechanical test, the material stiffness and the loss modulus were measured. The complex modulus(E^*) will be calculated based on a modulus equals stress/strain. From the result obtained on the graph for E^* and measurement of Tan Delta (tan δ), the storage modulus (E') and loss modulus (E'') will be calculated[28]. The storage modulus (E') was used as the elastic component, and it is related in the stiffness sample of polymer composite material. Furthermore, the loss modulus (E'') was used as viscous component and is related in the ability of the sample to dissipate mechanical energy through molecular motion. Finally, tangent of phase difference (tan δ) will provide the information about the relationship between the elastic and inelastic for polymer matrix composite material. According to Nikzad et al. [10], it was found that the glass transition temperature increased proportionally with an increment of 10% iron in ABS. The peak glass transition temperature(Tg) of unfilled ABS is 126 ^{o}C and glass transition temperature(Tg) of 10% filled ABS is 118.9 ^{o}C. Furthermore, the Tan δ will be calculated based on the loss modules divide by storage modulus value. Fig. 10 shows the comparison of dynamic mechanical properties of virgin ABS and 10% iron-powder filled ABS[10].

III. RESULTS AND DISCUSSION

In this study, the experiment was done on DMA 2980, manufactured by TA instrument, which is a common equipment used for analysis. In this study, two main dynamic mechanical properties observed are the storage modulus (E') and the loss factor (tanδ) for the copper-ABS feedstock. Storage modulus value is a measure of the maximum energy stored in the composites material during one cycle of oscillation [30]. Three (3) sample with different compounding of PMC has been done by extrusion process for sample preparation used in DMA analysis. Each PMC compounding consists of five pieces for each sample for analysis. The DMA setting was dual cantilever with amplitude 15 µm and temperature ramp of 10 ^{o}C/min up to 160 ^{o}C. The specimen size from injection moulding were cut into 57 mm x 10 mm x 3.2 mm and clamped in the dual cantilever mode in DMA machine.

Table II Storage modulus, loss modulus and tan delta of composites with 22 % copper filled in ABS polymer.

Temperature (^{o}C)	Max Storage Modulus (MPa)	Loss Modulus (MPa)	Tan δ
40	52731	10018	0.190
50	35176	9118	0.259
60	16941	4341	0.256
70	11827	2780	0.235
80	9787	2404	0.246
90	7333	2022	0.276
100	2360	809	0.343
110	**551**	**459**	**0.833**
120	145	88	0.608
130	90	27	0.306
140	68	16	0.235

Fig. 11 shows the results of temperature scan for the three dynamic properties for the 22% copper filled in ABS material. The testing was done for a temperature range of 30 ^{o}C to 160 ^{o}C at fixed frequency of 1 Hz. In Fig. 11, the complex viscosity is in Y-1 axis, the storage modulus in Y-2, the loss modulus in Y-3 and tan delta in Y-4 axis with temperature variation in X axis. The results shows that for the 22 % copper filled in ABS polymer, the complex viscosity, the storage and loss modulus decrease with increase in temperature. The maximum glass transition temperature of 22 % copper filled in ABS was 113 ^{o}C and the tan delta is 0.8813 approximately. Increment of 22 % copper filled in ABS increased the storage modulus up to 551MPa and the loss modulus up to 459 MPa. Table II shows the detailed values for the storage modulus, loss modulus and tan delta of composites with 22 % copper filled in ABS polymer.

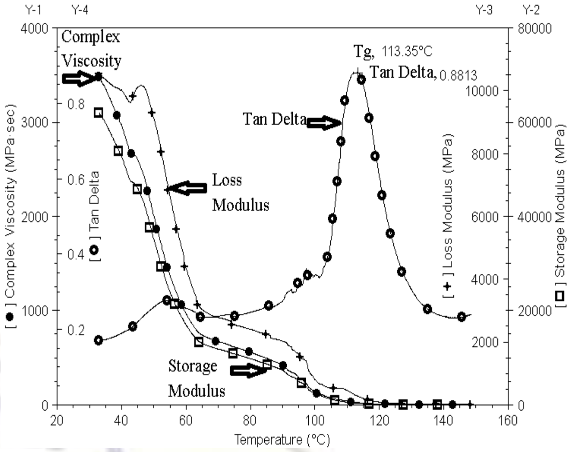

Figure 11 Dynamic Mechanical Properties of 22% copper powder filled ABS.

Table III Effect of copper filled in ABS on the Tg

Material	Heating rate (°C/min)	Heat flow Onset (W/g)	Tg Onset (°C)	Tg Midpoint (°C)
Abs Pure	10	-0.1235	104.5	106.2
Co71%,29%Abs	10	-0.0661	98.9	102.0
Co72%,28%Abs	10	-0.0881	100.5	102.7
Co73%,27%Abs	10	-0.0881	100.8	102.7
Co74%,26%Abs	10	-0.0523	101.7	103.6
Co75%,25%Abs	10	-0.0688	100.5	102.7

3.2 Differential Scanning Calorimetry(DSC)

The measurements of glass transition temperature (Tg) and heat flow were carried out by TA Instrument Q50 for differential scanning calorimeter (DSC). In this analysis, the samples were encapsulated on aluminum pans with 10.5 mg sample weight, prepared by extrusion method. The selected heating rate was 10 °C/min for each ABS and compounding material to achieve good resolution. The DSC test was carried out mainly to investigate the effect of copper filled in ABS material from injection molding with different compositions on the Tg temperature value. The results for ABS pure and injected compounding sample with different compositions are shown in Table III. Fig. 12 shows the DSC results for pure ABS material, where the glass transition temperature is found to be 104.5 ^{o}C on onset. From the results obtained, the increment of copper filled in ABS matrix does not much affect on the glass transition temperature(Tg). The maximum Tg onset of compounding 74% copper filled in 26 % ABS is 101.7 ^{o}C approximately. The minimum value of Tg onset is 98.9 °C with heat flow -0.0661 W/g for 71% copper filled in 29 % ABS. It is observed that, the Tg value decreases in the copper filled in ABS matrix. The detail of the heat flow of pure ABS and 74% copper powder filled in 26% ABS for FDM feedstock are shown in Fig. 13 and Fig. 14.

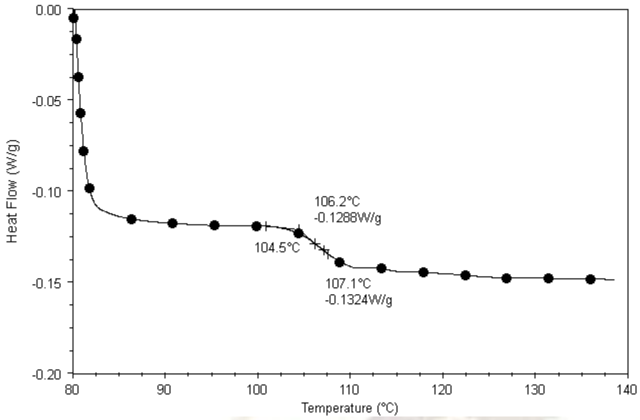

Figure 12 Glass transition temperature(Tg) of pure ABS

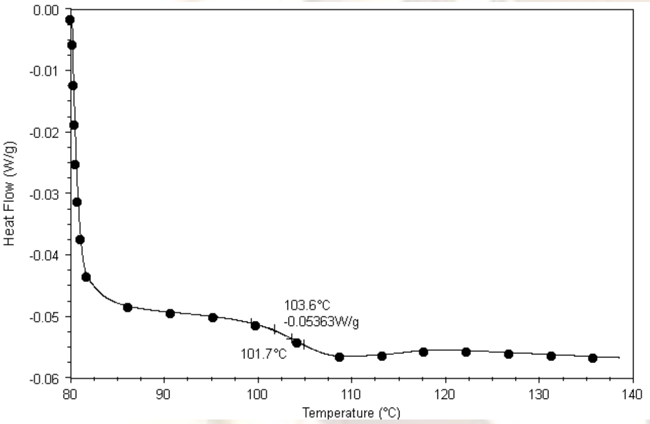

Figure 13 Glass transition temperature(Tg) of 74% copper filled in 26% ABS

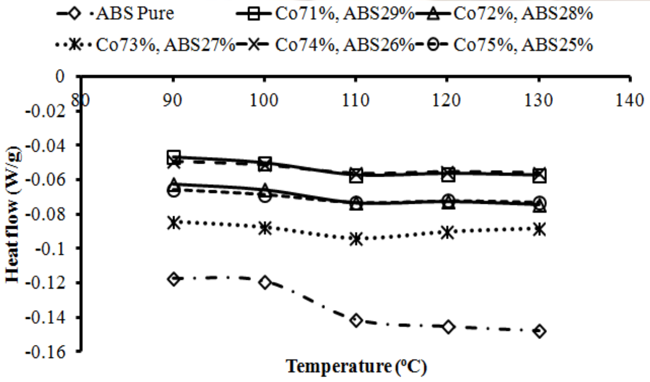

Figure 14 Heat Flow of copper powder filled in ABS with different composition.

IV. CONCLUSIONS

A new PMC feedstock with copper powder filled in ABS was successfully produced for dynamic mechanical analysis. The DMA testing has been successfully used to produce results for the storage modulus (E'), loss modulus (E") and Tan Delta (δ). The results show that, the dynamic mechanical properties of copper filled in ABS are greatly affected by the volume percentage of the copper content. The suitable material and binder selection, mixing method and parameter setting on melting temperature, pressure and cooling time may offer a great potential area for the metal feedstock fabrication in the wire filament extrusion through the extruder machine.

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