VOL. 11, NO. 8, APRIL 2016 ISSN 1819-6608
ARPN Journal of Engineering and Applied Sciences

www.arpnjournals.com

# FDM PREPARATION OF BIO-COMPATIBLE UHMWPE POLYMER FOR ARTIFICIAL IMPLANT

M. S. Ramli, M. S. Wahab, M. Ahmad and A. S. Bala
Advanced Manufacturing and Material Center, Faculty of Mechanical and Manufacturing Engineering, Universiti Tun Hussein Onn Malaysia, Parit Raja, Johor, Malaysia
E-Mail: saifulramly@gmail.com

**ABSTRACT**

Due to its properties of high wear, creep resistance, high stiffness and strength, Ultra-High Molecular Weight Polyethylene (UHMWPE) was developed to eliminate most metallic wear in artificial implant, which conventionally found in stainless steel, Cobalt Chromium (Co-Cr) and Titanium (Ti) alloys. UHMWPE has an ultra-high viscosity that renders continuous melt-state processes including one of the additive manufacturing processes, Fused Deposition Modeling (FDM) ineffective for making UHMWPE implant. Attempt to overcome this problem and adapting this material to FDM is by blending UHMWPE with other polyethylene including High Density Polyethylene (HDPE) and Polyethylene-Glycol (PEG) which provide adequate mechanical properties for biomedical application along with the improvement in extrudability. It was demonstrated that the inclusion of 60% HDPE fraction has improved the flowability of UHMWPE in MFI test and showing adequate thermal stability in TGA.

**Keywords:** additive manufacturing, fused deposition modelling, ultra-high molecular weight polyethylene high-density polyethylene, artificial implant.

## INTRODUCTION

Additive manufacturing is another alternative manufacturing process which also widely adopted in most manufacturing field alongside with the conventional and advanced machining. This is due to the obvious advantage of this technology which is relatively fast process by creating a three-dimensional (3D) object directly from CAD data (Chua *et al*, 2014). Although this technologies can be considered as newly emerged, which is just been introduced in 1980s (Chua *et al.*, 2014), there are a lot of patents in wide range of size and fabrication materials up to date on which proved to highly acceptance and interests in markets either for industrial application or even in research and development area.

The development however keeps growing in order to pursue many demands and needs from the various field especially for medical purposes in fabricating surgical model, customized prostheses and artificial implants (Chua *et al*., 2014). The existing of biomaterial in each type of material classification, including metal, polymer, ceramic and composite also acts as driving factor to grab the opportunity to explore the capabilities of this interesting technology. However, the compatibility of additive manufacturing technology towards fabricating biomaterial still been a challenge for certain type of rapid prototyping process due to some specific processing parameters required for specific process and application.

Although the basic process of additive manufacturing is initiated with CAD data and followed up by slicing process, the fabricating process plays one of the important roles to ensure a good output. This is the challenging stage where all processing parameters should match with the properties of desired fabricated material. For example in one of type additive manufacturing technology, Fused Deposition Modelling (FDM), the fundamental of extrusion applied in this fabrication method required correct amount of heat and pressure to cope with material properties such as flexibility, stiffness and viscosity (Novakova and Kuric, 2012) which formally in the form of filament.

### Introduction to FDM

FDM is one of the systems lies under solid based AM. Apart from liquid and powder based AM, this classification technique means that the material are in the form of wire, pellets or laminates. The prominent systems are including laminate object manufacturing (LOM) and FDM. These techniques are employing joining or binding method either with the aid of glue or binder such in LOM or chemically and physically bond with the aid of temperature such in FDM which focused in this study.

In FDM process, the material used is in spools of filaments which generally made of plastic material. Currently, the system has up to two extrusion head, which heats up the filament into a semi-molten or semi-liquid state. The semi-molten filament are then deposited line by line to finish up the corresponding layer and due to the difference in temperature of the surrounding air, which is lower than the melting point of the filament material, the semi-molten will progressively solidify after the deposition. For FDM systems which have a dual extrusion head as shown in Figure-1, the purpose of having two extrusion head instead of one normally is either one for fabricating model material and the other for processing support material or both are fabricating the model material but having a different color.

For the biomedical application, it is found that possibilities in controlling the fabrication process by commanding the selection of "sparse low" and "sparse

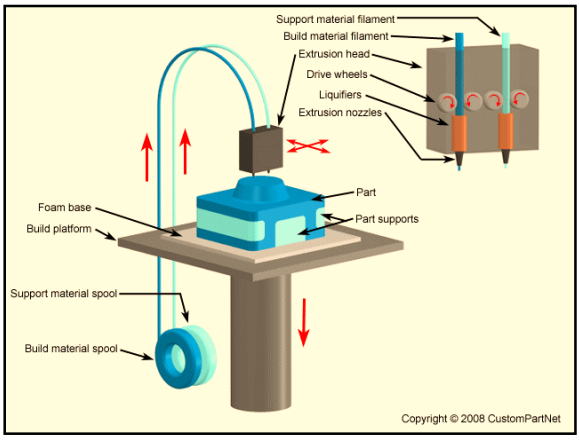


**Figure-1.** Fused deposition modeling (FDM) illustration (Custompart.net, 2009).

high" density of which previously intent to save the material, providing the grid of cavities inside the volume on which can mimicking the structure of human bone (Novakova and Kuric, 2012). Although the common end use of Acrylonitrile Butadiene Styrene (ABS) plastic in FDM process, there are several selection of biopolymer of which can be processed apart from commonly used ABS plastic such as ABS-M30i and PC-ISO (Novakova & Kuric, 2012). These selection of biomaterial gives a room for other potential candidate especially which conventionally used in extrusion based process including high density polyethylene (HDPE) and ultra-high molecular weight polyethylene (UHMWPE) to be adept into FDM process on which not yet been found on most of previous research regarding the development of that material towards additive manufacturing process ability.

**FDM of biomaterial**

There is a little change in the basic process of rapid prototyping when involving biomaterials. The specific application either scaffold tissue engineering or vascular segment need to have a sliceable virtual model before the fabrication takes place due to the existing of customization among living organism including human. For the example, in order to get a model of tissue engineering scaffold, a scaffold library have to be developed due to the customized structures (Chua *et al*., 2003). It is same goes to in order to build a prototype of certain organ, what is so called "blueprint" representing the organ have to be acquired first. From various of previous study, there are some existing method that can be employed to obtained the data including the 3D surface scan (Leong *et al*., 2003), CT scan or magnetic resonance imaging (MRI) scan (Ciocca *et al*., 2009).

However, up to today, most of present rapid prototyping systems can only process the material which supplied and recommended by the vendors. In fact, most of them are not biocompatible. This material constraint gives some room for improvement and opportunity to introduce numerous of potential biomaterials into the appropriate rapid prototyping system either direct method or indirectly.

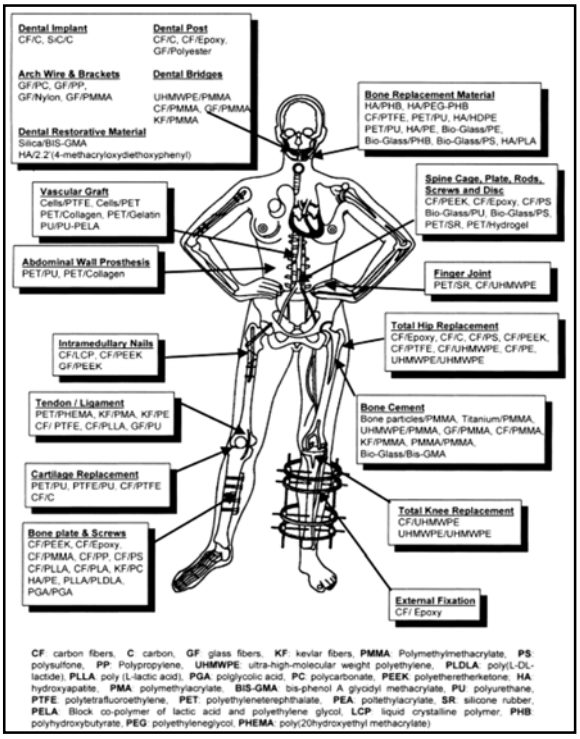


**Figure-2.** Various application for corresponding potential biopolymers (Ramakrishna *et al*., 2001).

Some progress and modifications have been done in some of commercial rapid prototyping systems including FDM (Zein *et al*., 2002). As shown in Figure-2, despite the widely employment of titanium in most clinical application, other potential biomaterials which are mostly from polymer also being employed such as polycaprolactone (PCL), polyether ether ketone (PEEK), ultrahigh molecular weight polyethylene (UHMWPE), polypropylene (PP), poly(methyl methacrylate) (PMMA) and some other biopolymer depends on their corresponding potential application.

Although the findings of numerous biomaterials especially biopolymers seem promising, the challenge to meet various parameters and characteristic including the material, processing and applications are still there. Those elements which are in concern should be treated and altered thoroughly to meet the corresponding requirements without compromising the biocompatibility and mechanical characteristics.

**The bio-compatible UHMWPE**

UHMWPE is one of unique polymer which having exceptional mechanical and physical properties. It becomes prominent due to its high toughness, chemical inertness, abrasion and impact resistance, lubricity and biocompatibility. Although with the potential characteristics which can be exploit into clinical application, but since 1950s, more than 90% of the

UHMWPE produced was used in industries such as gears, liners, unlubricated bearings, seals, pickers for machinery textile, runners for bottling production lines and also bumpers and siding for ships and harbors (Steven, 2004).

But in 1962, these properties have made UHMWPE progressively employed as a load bearing material especially for use in many implant application such as total joint replacement including knee, hip and shoulder of which have received significant attention in both academics and industries (Ahmad *et al*., 2013). Since that year, more than 1.4 million artificial joint replacement procedures have been performed around the world. But despite having those successful times, UHMWPE only have a finite lifetime due to wear and damage on its components. This reason is due to the performance of UHMWPE which directly affected by intrinsic weaknesses such as such as high creep when compared to metal and bone. As a result, the concern in improving the UHMWPE mechanical properties has encouraged many researchers to developed new UHMWPE blend or composite for the application.

Presently, UHMWPE based material are usually produced either by direct compression moulding process or by ram extrusion process which is under the definition of formative fabrication type and then these conventional process are followed by machining including turning or milling of which lies under the subtractive type of fabrication. By using the ram extrusion technique, the common grade of UHMWPE can extrude up to 350 mm diameter and 150 mm diameter for a medical grade of UHMWPE respectively (Allen, 2003). The extremely (ultra) high molecular weight have led directly to high viscosity of UHMWPE makes the processing a challenging task for present fabrication technique and even for additive manufacturing technique if comparing to the wire fabrication for FDM filament material of which about 1.75 mm diameter. Therefore, most of the works are concentrating on the refining the processability of UHMWPE on the extrusion based process and introducing the material towards FDM processing.

Several method have been identified based on previous researchers including the incorporation of processing aid such as with polyolefin or other polyethylene either high-density polyethylene (HDPE) and polyethylene glycol (PEG). The purpose of this method are in order to improve creep resistance, reducing extrusion pressure and melt viscosity (Ahmad *et al*., 2013). The addition of PEG is a unique processing additive due to its biocompatibility of which not compromising the chemical inertness of UHMWPE. It was found effective in reducing the surface roughness of polyethylene extrudate as well as the extrusion pressure. In addition, it is found that along with the reduction of die pressure, the melt viscosity during the extrusion also significantly reduce (Xie and Li, 2007).

## EXPERIMENTAL

### Materials

The UHMWPE used was GUR 1020 (Ticona, Telford, UK) and provided in powder form with a weighted average molecular weight of $3.5x10^6$ g/mol and density of 0.93 g/cm$^3$. HDPE (Etilinas HD5403AA) comes with a density of 0.954 g/m$^3$, weight average molecular weight of $1.2x10^5$ g/mol, and melt flow rate of 0.25 g/10 min at 190 $^oC$. PEG with an average molecular weight of 4000 comes with a density of 1.027 kg/m$^3$.

### Blending and sample preparation

A preliminary test was done as a pilot on mixing the UHMWPE with a small amount of PEG. Another test was also done in conjunction by mixing the UHMWPE with HDPE. The uniformly mixed polyethylene batches were blended in Brabender Plastograph with the speed of 20 rpm, with the addition of 2%, 5% and 10% of PEG respectively. For the UHMWPE/HDPE formulation, the addition of HDPE by 50%, 40% and 30% was assigned. The blending temperature was set as 190 $^oC$ for all formulations. The resin of HDPE was prior grinded into powder before mixing with HDPE.

**Table-1.** The composition of the UHMWPE/PEG blends.

| Sample Formulation | UHMWPE (wt %) | PEG (wt %) |
|---|---|---|
| U98PEG2 | 98 | 2 |
| U95PEG5 | 95 | 5 |
| U90PEG10 | 90 | 10 |

**Table-2.** The composition of the UHMWPE/HDPE blends.

| Sample Formulation | UHMWPE (wt %) | HDPE (wt %) |
|---|---|---|
| U60H40 | 60 | 40 |
| U50H50 | 50 | 50 |
| U40H60 | 40 | 60 |

### Thermogravimetric analysis

Thermogravimetric analysis (TGA) was performed from the ambient temperature of 30 $^oC$ – 600 $^oC$ under a controlled environment with a heating rate of 10 $^oC$/min by using Linseis PT1000 to determine the thermal degradation behavior of the composites.

### MFI analysis

The melt flow index (MFI) analysis was carried out using CEAST MF10 Melt Flow according to ASTM D1238 (Procedure A) standard. The method is described on which a hand loaded mass of the polymer (in grams), presented through a capillary of 24.50 mm length and 9.51 mm diameter, and leave it to flow through 2.095 mm diameter die (orifice) by applying standard load and temperature (190 $^oC$, 5kg) in ten minutes under a pressure

applied. The outcomes indicate the flowability for each sample formulation and reflect the inversely proportional to the viscosity under the same controlled parameter.

**Pre-processing on FDM**

By having the sample in the form of filament with average diameter of 2.03±0.04 mm, a preliminary test was carried out using Wanhao Duplicator 4 machine to evaluate the extrudability through the print head nozzle of 0.4 mm diameter. The print temperature was set as 230ºC for all formulation.

## RESULTS AND DISCUSSIONS

**Thermo gravimetric analysis**

TGA of pure UHMWPE and UHMWPE/PEG blends was carried out to assess the effects of blend composition on the thermal stability of the polymers. The TG curves are shown in Figure-3 whereas the degradation temperature of UHMWPE/PEG blends obtained from TG and DTG curves are presented in Table-3.

The initial thermal stability is categorized by temperatures at 10% weight loss, which referred as $T_{10\%}$. The TG curves showed only one step of degradation for both pure UHMWPE and UHMWPE/PEG blends. In general, the $T_{10\%}$ decreased with the increasing PEG fraction up to 5% ratio and it is shown that thermal degradation of blend with 10% ratio of PEG took place most rapidly compared to pure UHMWPE and the blends. This results was also consistent with a previous study describing the reason for adding only low content of PEG not more than 10% ratio into the blends because of the reduction in thermal stability on UHMWPE/PEG blends. This is due to the presence of PEG particles hinder the heat transfer and reduces the surface area needed for the kinetic effects, which in turn reduces the degradation temperature of the UHMWPE/PEG blends (Ahmad *et al*., 2013). It is also observed that the sample weight percentage decreased continuously as the temperature increase with some of the blends had char residue even at temperature reach 500ºC with no significant changes for the peak of decomposition temperature ($T_p$).

**Table-3.** Degradation temperature of components and UHMWPE/PEG blends obtained from TG and DTG curves.

| Formulation | Percentage composition (wt %) | | Initial decomposition temperature, $T_{10\%}$ (ºC) | Peak of decomposition temperature, $T_p$ (ºC) |
|---|---|---|---|---|
| | UHMWPE | PEG | | |
| U100 | 100 | 0 | 398 | 477 |
| U98PEG2 | 98 | 2 | 394 | 476 |
| U95PEG5 | 95 | 5 | 381 | 475 |
| U90PEG10 | 90 | 10 | 359 | 475 |

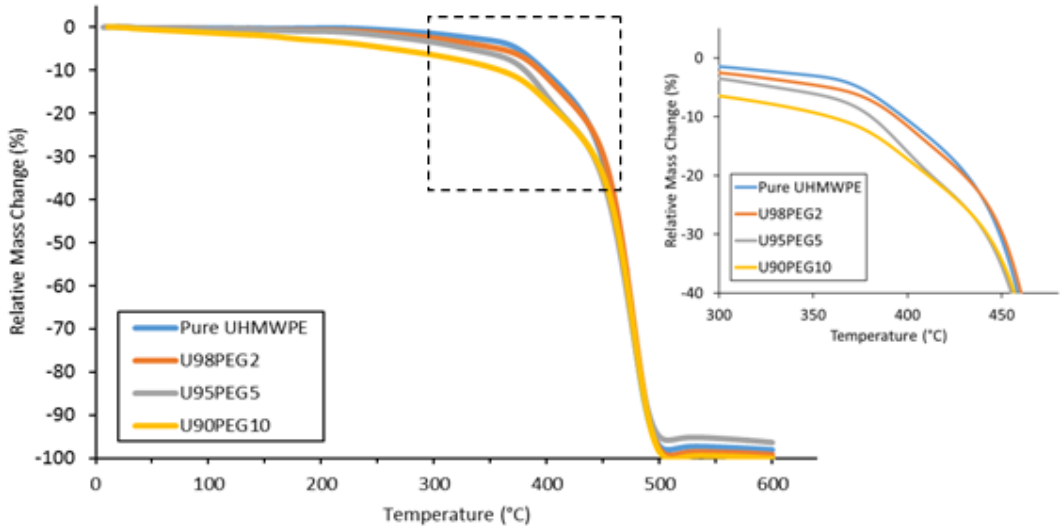


**Figure-3.** Thermograms of UHMWPE and UHMWPE/PEG blends.

The TG curves for the UHMWPE/HDPE blends are illustrated in Figure-4 and the summary of the degradation temperature is shown in Table-4. The result shows that degradation is a one step process and occurs in broad temperature region between 300ºC and 490ºC. Thermal degradation of the blend containing 60% ratio of

HDPE took place earliest compared to other blends. However, it is clearly observed that there are ambiguities upon the trends on which the blend containing 70% of HDPE fraction lies beyond the neat UHMWPE trend. This behavior also reported and illustrated that the UHMWPE/HDPE blend lies between those of the individual polymers and higher than the UHMWPE within the range of 437 ºC – 481 ºC due to the random scissions and branching occurs simultaneously (Ahmad *et al*., 2013). In general, the $T_{10\%}$ decrease with increasing HDPE fraction. The sample weight percentage decreased continuously with some of the blends not degrade completely above 500 ºC.

**Table-4.** Degradation temperature of components and UHMWPE/HDPE blends obtained from TG and DTG curves.

| Formulation | Percentage composition (wt %) | | Initial decomposition temperature, $T_{10\%}$ (ºC) | Peak of decomposition temperature, $T_p$ (ºC) |
|---|---|---|---|---|
| | UHMWPE | HDPE | | |
| U100 | 100 | 0 | 398 | 477 |
| U50H50 | 50 | 50 | 389 | 473 |
| U40H60 | 40 | 60 | 371 | 468 |
| U30H70 | 30 | 70 | 410 | 476 |

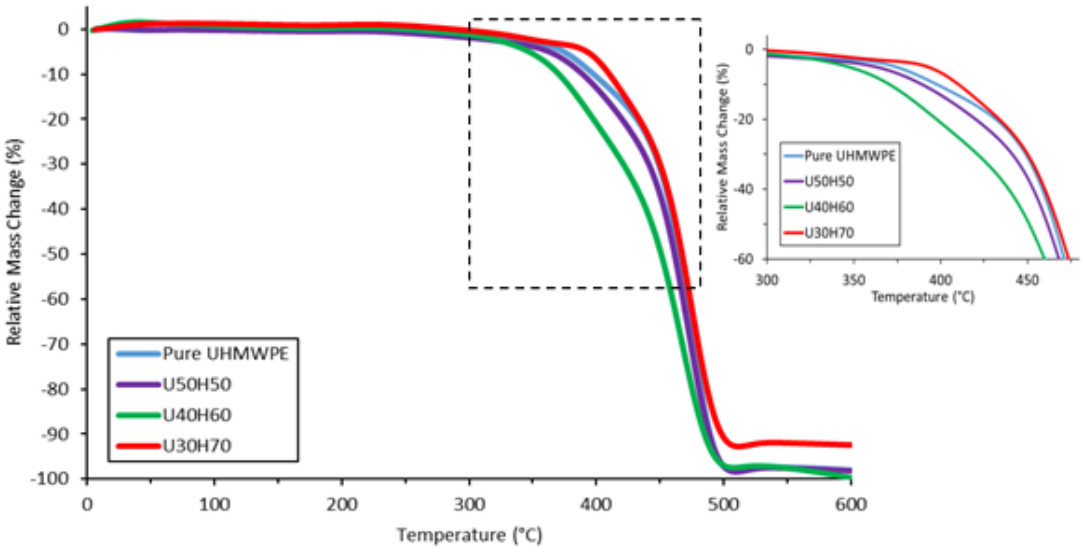


**Figure-4.** Thermograms of UHMWPE and UHMWPE/PEG blends.

**MFI analysis**

The melt flow index (MFI) analysis of UHMWPE/HDPE blends was carried out to measure the ease of flow of the melt blends as the results also can reflect to the inverse effect of viscosity of the blends. Figure-5 illustrate the results of MFI obtained for each of the blends along with the benchmarking of neat UHMWPE and HDPE whereas also been presented in Table-5.

The intention of blending UHMWPE with HDPE was to improve the processability of UHMWPE by implementing the processing ability of HDPE to the material properties superiority of UHMWPE. In general, the MFI increase with the increasing HDPE fraction up to 60%. This results was also reliable with a study which explained that most of the mechanical properties possessed by UHMWPE was found maintained up to 60 wt% HDPE before a sudden drop was observed with increasing HDPE content (Ahmad *et al.*, 2013). This is possible due to the weak interface combination between UHMWPE particles and HDPE matrix, thus resulting the uneven shear distribution throughout the blends.

**Table-5.** Melt flow index of UHMWPE/HDPE blends.

| Formulation | Trials (g/10 min) | | | Average (g/10 min) |
|---|---|---|---|---|
| | 1 | 2 | 3 | |
| U50H50 | 0.285 | 0.453 | 0.288 | 0.342 |
| U40H60 | 0.915 | 1.072 | 0.969 | 0.969 |
| U30H70 | 0.044 | 0.049 | 0.057 | 0.050 |

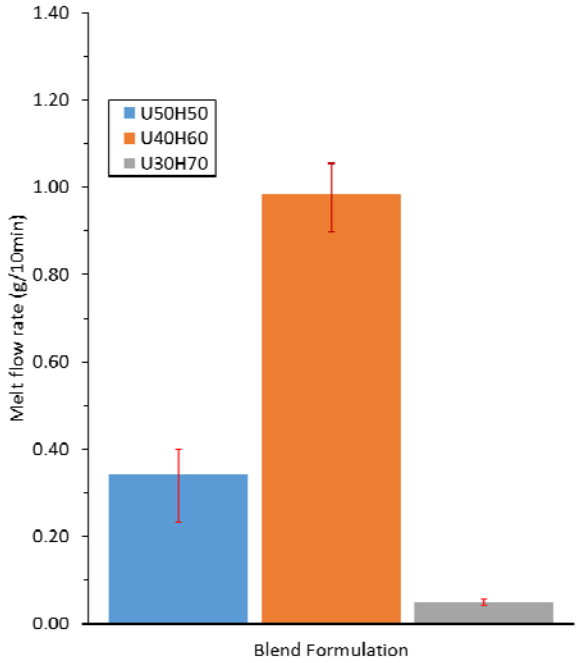


**Figure-5.** Melt flow index of UHMWPE, HDPE and UHMWPE/HDPE blends.

## CONCLUSIONS

From this study, it was discovered that the PEG is not suitable to blends directly with UHWMPE even though it is found successful as internal lubrication. This is support by numbers of the previous study used HDPE, hydroxyapatite (HA) and polypropylene (PP) as a compatibilizer in conjunctions for blending. The results also proved the reasons why blending UHMWPE with only small amount of PEG often used. In the other hand, the melt flow index result indicates that HDPE was found successful in improving the processability of UHMWPE with the optimum blend formulation not more than 60 wt% of HDPE. Whereas on the pre-processing on FDM, although by using the MFI samples filament, it is showing promising results where the sample filament of U50H50 formulation able to be extruded with a constant diameter as illustrated in Figure 6 and ready for further filament fabrication.

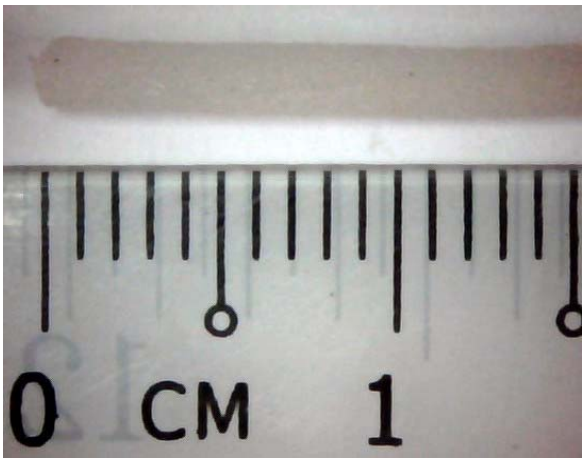


**Figure-6.** The UHMWPE/HDPE filament wire from MFI analysis (ruler for scale).

## ACKNOWLEDGEMENT

This research was supported under grant MDR 1312 by Office for Research, Innovation, Commercialization and Consultancy Management (ORICC), Universiti Tun Hussein Onn Malaysia (UTHM).

## REFERENCES

Ahmad, M., Wahit, M. U., Kadir, M. R. A., Dahlan, K. Z. M., Jawaid, M., Rafiq, M., Mohd, K. Z. (2013). Thermal and mechanical properties of ultra-high molecular weight polyethylene / high-density polyethylene / polyethylene glycol blends. Journal of Polymer Engineering, 33(7), 599-614.

Allen, M. 2003. UHMWPE processing : techniques and problems.

Chua, C. K., Leong, K. F. and An, J. 2014. Rapid Prototyping of Biomaterials, 1-15.

Chua, C. K., Leong, K. F., Cheah, C. M. and Chua, S. W. 2003. Development of a Tissue Engineering Scaffold Structure Library for Rapid Prototyping. Part 1 : Investigation and Classification, 291–301.

Ciocca, L., De Crescenzio, F., Fantini, M. and Scotti, R. 2009. CAD/CAM and rapid prototyped scaffold construction for bone regenerative medicine and surgical transfer of virtual planning: a pilot study. Computerized

Medical Imaging and Graphics: The Official Journal of the Computerized Medical Imaging Society, 33(1), 58-62.

Custompart.net. 2009. Fused Deposition Modeling (FDM). Retrieved from http://www.custompartnet.com

Leong, K. F., Cheah, C. M., and Chua, C. K. 2003. Solid freeform fabrication of three-dimensional scaffolds for engineering replacement tissues and organs. Biomaterials, 24(13), 2363-2378.

Novakova-marcincinova, L. and Kuric, I. 2012. Basic and Advanced Materials for Fused Deposition Modeling Rapid Prototyping Technology, 11(1), 24-27.

Ramakrishna, S., Mayer, J., Wintermantel, E. and Leong, K. W. 2001. Biomedical applications of polymer-composite materials : a review, 61.

Steven M. Kurtz. 2004. The UHMWPE Handbook : Ultra-High Molecular Weight Polyethylene in Total Joint Replacement (1st Editio). Elsevier.

Xie, M. and Li, H. 2007. Viscosity reduction and disentanglement in ultrahigh molecular weight polyethylene melt: Effect of blending with polypropylene and poly (ethylene glycol). European Polymer Journal, 43(8), 3480-3487.

Zein, I., Hutmacher, D. W., Tan, K. C. and Teoh, S. H. 2002. Fused deposition modeling of novel scaffold architectures for tissue engineering applications. Biomaterials, 23(4), 1169-1185.