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Research Article

Effect of Polypropylene Fiber Type, Content and Length on Mechanical Performance of Rigid Polyurethane Composites

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ABSTRACT

The purpose of this work is to improve mechanical properties of rigid polyurethane (RPU) composites used in traditional ceramic casting industry. Therefore, monofilament (mono) and fibermesh (fibril) polypropylene (PP) fibers with various lengths (3, 6, 12 and 18 mm) were incorporated to polymer matrix at different rates (0.5, 1.0, 1.5 and 2% by weight). Effects of fiber type and content on flexural strength, bending strength and compressive strength of composites were investigated. Surface morphology and thermal characteristics of composites were evaluated by SEM and TGA analysis, respectively. Bulk densities of specimens with and without PP fibers vary between 72,15-146 kg/m^3. Compared to pure rigid polyurethane foam, bulk density of monofilament PP reinforced composites significantly increased and the highest density value (146,86 kg/m^3) was reached in M6/2.0 sample. On the other hand, incorporation of fibrilmesh caused a decrease in bulk density. While the increase in percentage of mono PP increased flexural strength, the presence of fibril PP had a negative effect on strength. Compressive strength of all mono PP reinforced composites is higher than that of pure RPU, except for M6/0.5 sample. Besides, SEM analysis revealed that the presence of PP fibers generally reduced number of closed cells in composite structure. Experimental findings indicate that fiber type, content and length affect mechanical performance of RPU composites. In addition, it is possible to use mono PP fiber reinforced RPU composites as support apparatus in ceramic casting industry.

Keywords: *Rigid polyurethane,* Polypropylene *fiber, Composite, Mechanical performance*

Polipropilen Lif Türü, İçeriği ve Uzunluğunun Sert Poliüretan Kompozitlerin Mekanik Performansına Etkisi

ÖZ

Bu çalışmanın amacı, geleneksel seramik döküm endüstrisinde kullanılan sert poliüretan kompozitlerin mekanik özelliklerini iyileştirmektir. Bu nedenle, çeşitli uzunluklarda (3, 6, 12 ve 18 mm) monofilament (mono) ve fiberağ (fibril) polipropilen (PP) elyaflar polimer matrisine farklı oranlarda (ağırlıkça %0.5, 1.0, 1.5 ve %2) dahil edildi. Elyaf tipi ve içeriğinin kompozitlerin eğilme mukavemeti ve basınç mukavemeti üzerindeki etkileri araştırılmıştır. Kompozitlerin yüzey morfolojisi ve termal özellikleri sırasıyla SEM ve TGA analizi ile değerlendirildi. Saf

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poliüretan ve PP lif içeren numunelerin kütle yoğunlukları 72,15-146 kg/m^3 arasında değişmektedir. Saf poliüretan kompozite kıyasla monofilament PP takviyeli kompozitlerin kütle yoğunluğu önemli ölçüde artmış ve M6/2.0 numunesinde en yüksek yoğunluk değerine (146,86 kg/m^3) ulaşılmıştır. Öte yandan, fiberağ (fibril) ilavesi, yığın yoğunluğunda bir azalmaya neden oldu. Mono PP yüzdesindeki artış eğilme mukavemetini arttırırken, fibril PP'nin varlığı mukavemeti olumsuz etkilemiştir. Mono PP takviyeli kompozitlerin basınç dayanımı (M6/0.5 numunesi hariç) saf polüretandan daha yüksektir. Ayrıca SEM analizi, PP liflerinin varlığının genellikle kompozit yapısındaki kapalı hücre sayısını azalttığını ortaya koymuştur. Deneysel bulgular, elyaf tipi, içeriği ve uzunluğunun sert poliüretan kompozitlerin mekanik performansını etkilediğini göstermektedir. Ayrıca, mono PP elyaf takviyeli rijit poliüretan kompozitlerin seramik döküm endüstrisinde destek aparatı olarak kullanılması mümkündür.

Anahtar kelimeler: *Sert poliüretan, Polipropilen lif, Kompozit, Mekanik performans*

I. INTRODUCTION

Polyurethane polymers have high strength and wear resistance, superior durability against oxygen, ozone, gas, oil and aromatic hydrocarbons [1]. Due to their production methods and ease of use, they are used in many sectors such as automotive, furniture, clothing, construction, medical, and ceramic industry [2,3]. Of these, polyurethane foams cover the largest segment in thermoset foam industry. They have many physical properties such as different density varieties, flexibility, viscoelasticity, semi-bendability, hardness, heat and flame resistance. Polyurethane foams are prepared by reacting polyols and polyisocyanates with the help of blowing agents. Molecular weight and functionality of polyols affect the properties of resulting foams [4–6]. Polyisocyanates act as binders for polyols, hence they are considered to be building block polymers of urethane and related foams [5,7,8].

Rigid polyurethane (RPU) composites are preferred in construction and industrial insulation and sealing applications [9–11] due to their superior properties such as low thermal conductivity, low density, shape stability, low moisture permeability and low water absorption. Besides, they are also used in the wings and fuselage of airplanes [12] and in production of sandwich panels. Moreover, since this type of cellular materials can absorb impact energy, it has also found use for packaging and cushioning [13]. Main characteristic of RPU composites is that cells of the polymer matrix in form of a network are closed [4]. On the other hand, studies in which various additives such as nanofibers [14], organoclay [15], multi-walled carbon nanotubes [16] and various inorganic nanofillers [17,18] are used to further reduce thermal conductivity of RPU composites and improve their mechanical properties are found in literature. In addition to additive-polyurethane matrix compatibility, particle size is also important in selection of additives [19]. The presence of large particles and use of excess additives affect nucleation and growth mechanisms of gas bubbles during polymerization reaction and it may cause destruction of cell walls. In such a case, strength and thermal insulation performance of RPU composite decreases. In this sense, it is known that strength of fiber-containing polymer composites is affected by a number of parameters, most of all, the concentration and length of fibers, and degree of interfacial adhesion between fiber and matrix [20–23]. Wilberforce and Hashemi [24] reported that tensile strength of polyurethane composites raised with increase of short-glass fiber concentration from 10% to 30% wt. In another study, in which rosella fiber was added to polyurethane matrix at different rates (10, 20, 30, 40 and 50% by weight), it was stated that fiber content played a serious role on tensile and flexural and impact strength. Also, the best mechanical and thermal properties were achieved in polyurethane composites containing 40% rosella fiber content [25].

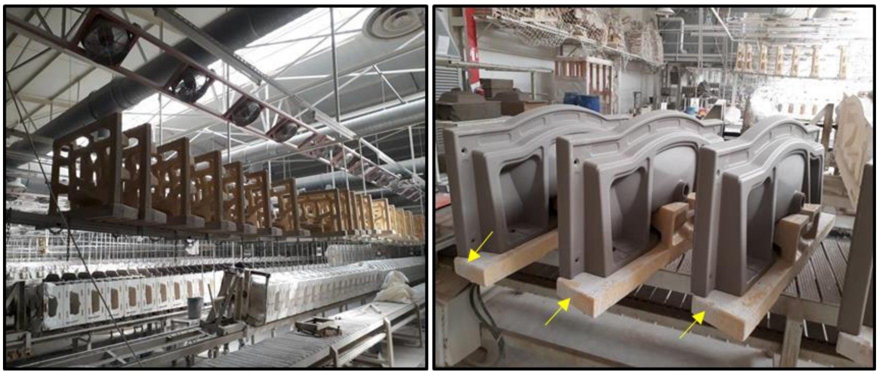

Figure 1. *Examples of RPU materials used as a modeling support apparatus in production of ceramic sanitary ware*

Use of RPU composites in traditional ceramic molding industry (Figure 1) further highlights mechanical strength performance of these materials. With the approach that the presence of fiber can improve mechanical performance of RPU, monofilament (mono) and fiber mesh (fibril) PP fibers in different ratios (0.5, 1.0, 1.5 and 2% by weight), various lengths (3, 6, 12 and 18 mm) were incorporated in polymer matrix. And, investigations were done into how fiber type and content affected compressive, bending, and flexural strengths of composites. SEM and TGA analyses, respectively, were used to assess surface morphology and thermal properties of composites.

II. MATERIALS AND METHODS

A. MATERIALS

Isocyanate MDI (Tekpol) and polyol 1232-140 mixture (Tekpol) were obtained from Turkuaz Seramik in Kayseri. Isocyanate MDI is dark in color. Polyol mixture composes of high functionality polyols, catalysts, stabilizers and new generation blowing agents which don't harm the ozone layer. Some physical and chemical properties of isocyanate and polyol are presented in Table 1. Two different PP fibers (monofilament 3, 6, 12, 18 mm, fibermesh 6, 12, 18 mm) supplied from Polyfibers company. The types of PP fiber are presented in Figure 2.

Table 1. *Some physical and chemical properties of polyol and isocyanate*

Property	**Polyol**	**Isocyanate**	**Method**
	Value	**Value**	
Density (g/cm^3)	1.10-1.12	1.23	DIN 51757-2011
Viscosity (25˚C, mPa.s)	550±50	230±30	DIN 53018
Free isocynate content (%)	-	31.5±1	ASTM D5155

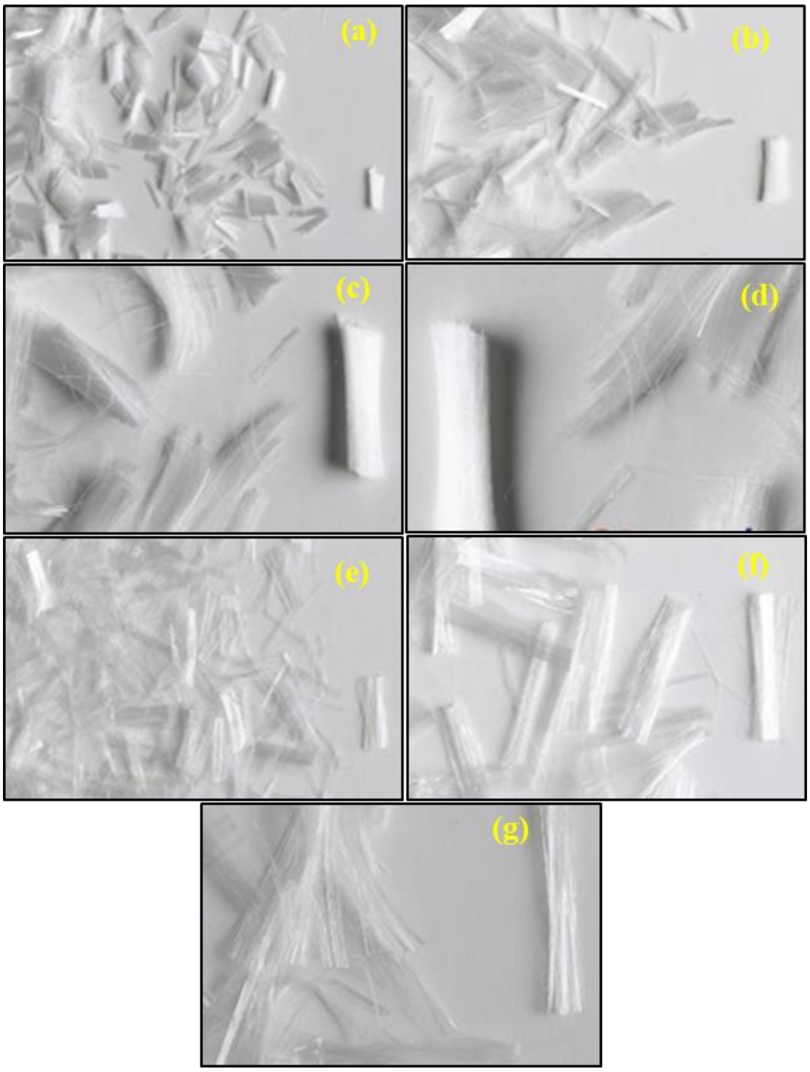

Figure 2. *Types of PP fibers: monofilaments (mono) of 3 mm* ***(a)****, 6 mm* ***(b)****, 12 mm* ***(c)*** *and 18 mm* ***(d)****; fibermesh (fibril) of 6 mm* ***(e)****, 12 mm* ***(f)*** *and 18 mm* ***(g)*** *[9]*

B. METHODS

B.1. Preparation of RPU Composites Mixture

Polyol/isocyanate ratio for RPU composite production was determined as 1.0/1.1 by weight. Isocyanate and polyol were placed in separate containers. Polyol was slowly added to isocyanate and mixed with a mechanical mixer at 3000 min^{-1} for 15 sec. Prepared mixture was poured into mold and cured at 40˚C for 30 min. Cured RPU removed from mold were left at room temperature for 24 h to complete polymerization reaction. Production steps of RPU composites are given in Figure 3. Composition of RPU with/without PP fiber are given in Table 2. A total of 20 samples were produced.

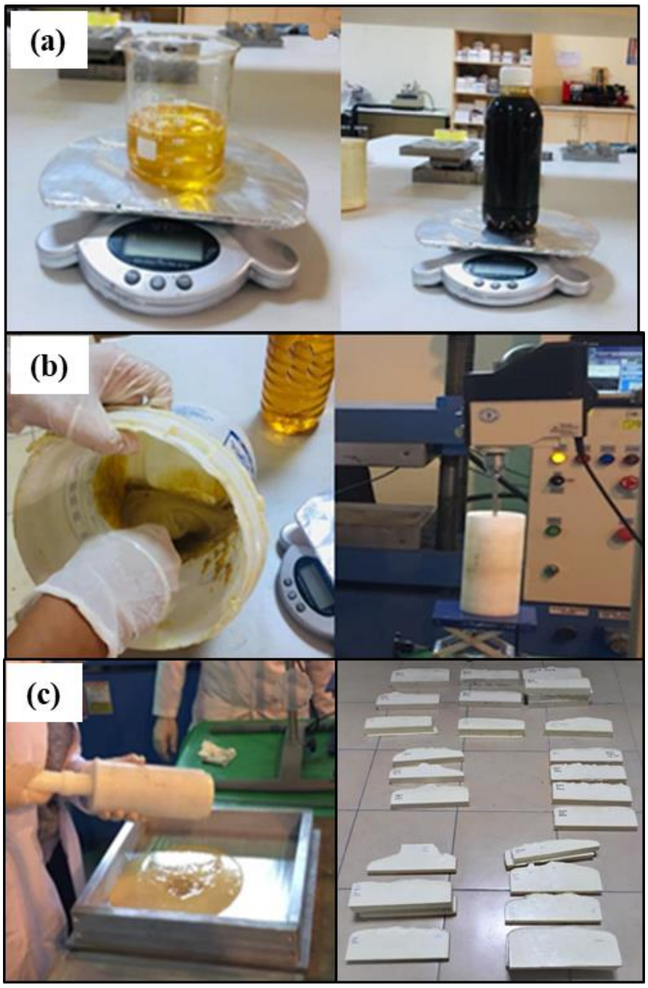

Figure 3. Processing stages of RPU ***(a)*** *weighing,* ***(b)*** *preparation of mixture,* ***(c)*** *molding and product*

B.2. Instrumental Testing And Analysis

Density measurements of RPU composites were calculated according to ASTM D 1622-08 standard [26]. Flexural strength properties of RPU composites were determined using three-point bending method (Shimadzu AG-XD 50) in accordance with ASTM D 790-03 standard [20]. Samples with dimensions of 25 x 25 x 100 mm were prepared and test speed was set at 2.5 mm per minute. Compressive strengths of RPU composites were determined according to ASTM D 1621-10 [21]. For this test, samples with dimensions of 50 x 50 x 40 mm were prepared. Tests were carried out at a speed of 2.5 mm per minute (Shimadzu AG-XD 50). Thermal decomposition behavior of RPU composites with/without PP fiber were analyzed by TGA (Perkin Elmer Diamond) at a heating rate of 10˚C/min in oxygen medium. Surface morphologies of foams were characterized by scanning electron microscopy (SEM, ZEISS LS-10). All mechanical tests were performed in at least three replicates.

Table 2. *Mixing ratios for* RPU composites *production*

Fiber type	**Sample**	**Fiber length** *(mm)*	**Polyol** *(% wt.)*	**Isocyanate** *(% wt.)*	**Fiber** *(% wt.)*
	Pure	0	50	50	0
Monofilament (mono) PP fibre	M3/0.5	3	49,75	49,75	0,5
	M3/1.0	3	49,5	49,5	1
	M3/1.5	3	49,25	49,25	1,5
	M3/2.0	3	49	49	2
	M6/0.5	6	49,75	49,75	0,5
	M6/1.0	6	49,5	49,5	1
	M6/1.5	6	49,25	49,25	1,5
	M6/2.0	6	49	49	2
	M12/0.5	12	49,75	49,75	0,5
	M12/1.0	12	49,5	49,5	1
	M12/1.5	12	49,25	49,25	1,5
	M12/2.0	12	49	49	2
	M18/0.5	18	49,75	49,75	0,5
	M18/1.0	18	49,5	49,5	1
Fibermesh (fibril) PP fibre	F6/0.5	6	49,75	49,75	0,5
	F6/1.0	6	49,5	49,5	1
	F6/1.5	6	49,25	49,25	1,5
	F12/0.5	12	49,75	49,75	0,5
	F18/0.5	18	49,75	49,75	0,5

III. RESULTS AND DISCUSSION

Density of PP fiber is 0.91 g/cm^3, and average density of polyurethane matrix is 0.11041 g/cm^3. Density values of RPU composites vary between 72.16-146.86 kg/m^3 (Table 3). Reason for this may be that densities of monofilament PP fibers are higher than that of pure polyurethane matrix. While a significant increase was observed in density of monofilament PP fiber reinforced composites, density of fibermesh PP fiber reinforced composites decreased compared to pure RPU. Compared to fiber-free foam samples, density of M6/2.0 sample is the highest. Monofilament PP fibers are harder and can be dispersed more homogeneously in mixture. Fibermesh PP fibers are clustered in groups and can be stretched in accordion form. Therefore, because they are thin and short and do not disperse homogeneously in mixture, it adversely affected density of RPU composites.

Table 3. *Density results of RPU composites*

Sample	**Fiber length** *(mm)*	**Fiber amount** *(%)*	**Bulk density** *(kg/m^3)*
Pure	0	0	108,17±2,12
M3/0.5	3	0,5	114,35±1,09
M3/1.0	3	1	114,49±1,25
M3/1.5	3	1,5	123,00±2,01
M3/2.0	3	2	114,90±1,91
M6/0.5	6	0,5	110,34±1,88
M6/1.0	6	1	122,78±2,71
M6/1.5	6	1,5	129,33±2,34
M6/2.0	6	2	146,86±1,85
M12/0.5	12	0,5	112,86±2,74
M12/1.0	12	1	112,16±2,67
M12/1.5	12	1,5	123,37±1,86
M12/2.0	12	2	118,20±2,68
M18/0.5	18	0,5	115,53±1,98
M18/1.0	18	1	140,24±1,69
F6/0.5	6	0,5	104,11±2,19
F6/1.0	6	1	72,16±1,94
F6/1.5	6	1,5	106,49±2,36
F12/0.5	12	0,5	108,37±1,85
F18/0.5	18	0,5	106,11±1,84

Flexural strength and flexural modulus results of unreinforced and PP fiber reinforced RPU composites are given in Figure 4. As seen from Figure 4, flexural strength values of PP fiber reinforced RPU composites of M3/2.0, M6/2.0 and M18/1.0 are higher than non-reinforced polyurethane. Besides, fibermesh (fibril) PP fiber reinforced RPU composites were found to have lower flexural strength values. This may be due to aggregation of fibermesh (fibril) PP fibers. As mono PP fiber percentage is increased, flexural strength of structure also increased.

Flexural modul of mono PP reinforced RPU composites were higher than fibril PP reinforced RPU composites (Figure 4). Reason for this is that when mono PP is reinforced to RPU composites, monofilament fiber structure maintains its straight shape in matrix resulting in high bending strength. On contrary, fibermesh (fibril) of low strength can not maintain their shapes, resulting in low bending strength. Monofilament PP fibers are one-dimensional and can be independently and homogeneously dispersed in a polymer matrix. On the other hand, mesh type PP fibers have a mesh-like structure and performs a multi-dimensional stress distribution under one-dimensional tensile load. Due to their mesh-like structure, it is thought that the mesh type PP fibers are more heterogeneously distributed at same addition ratios as monofilament PP fibers and regional discontinuities occur. Threfore, their bending strengths become weaker.

Flexural elongation values of fiber-reinforced RPU composites were found to be lower than non-reinforced structure. It can be attributed to bond between fiber and matrix preventing deformation of structure [27]. On the other hand, PP fibers disrupt air bubble structure in reinforced RPU composite.

As a result, while reduction in amount of PP fiber decreased degradation, a raise in fiber length increased it.

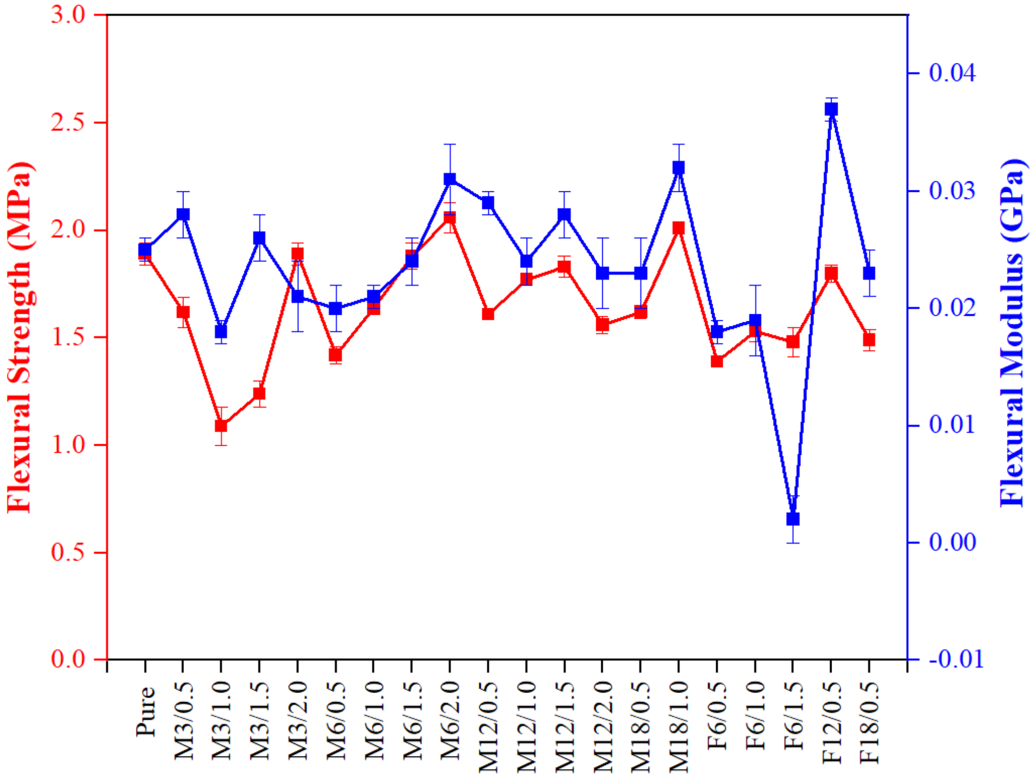

Figure 4. *Comparison of flexural strength and flextural modulus of RPU composites*

According to data in Figure 5a, all compressive strength values of mono PP reinforced RPU composites are higher than unreinforced RPU composites except for M6/0.5 sample. As seen from Figure 5b, compression shortening of half of fiber reinforced RPU composites is higher than compression shortening of unreinforced composites. This shows that when fiber reinforcement is applied to RPU composites compression increases. M3/0.5 PP reinforced specimens were found to have the highest compression shortening. In addition, all monofilament fiber reinforcement of 3 mm fiber length at different amounts (M3/0.5 – M3/1.0- M3/1.5 – M3/2.0) demonstrated very high compression shortening.

Thermal degradation behaviors of unreinforced RPU composite, monofilament PP and M6/2.0 PP fiber reinforced RPU composite specimens were analyzed by TGA (Figure 6). Polyurethanes are unstable and their degradation temperatures vary depending on chemical composition of polyurethane. Decomposition usually begins with degradation of urethane bond (approximately 150-220˚C) and release of CO_2 and isocyanate [28,29]. Maximum weight loss occurs after 400˚C, possibly representing polyol degradation [30].

Thermal decomposition temperatures of M6/2.0 PP fiber and reinforced RPU composites are 500˚C and 270˚C, respectively. Maximum weight losses are between 270˚C and 600˚C and total weight losses at 800˚C is over 80%. The highest weight loss of monofilament PP fiber occurred between 400˚C and 500˚C. At the end of 800˚C, total weight loss is almost 100%.

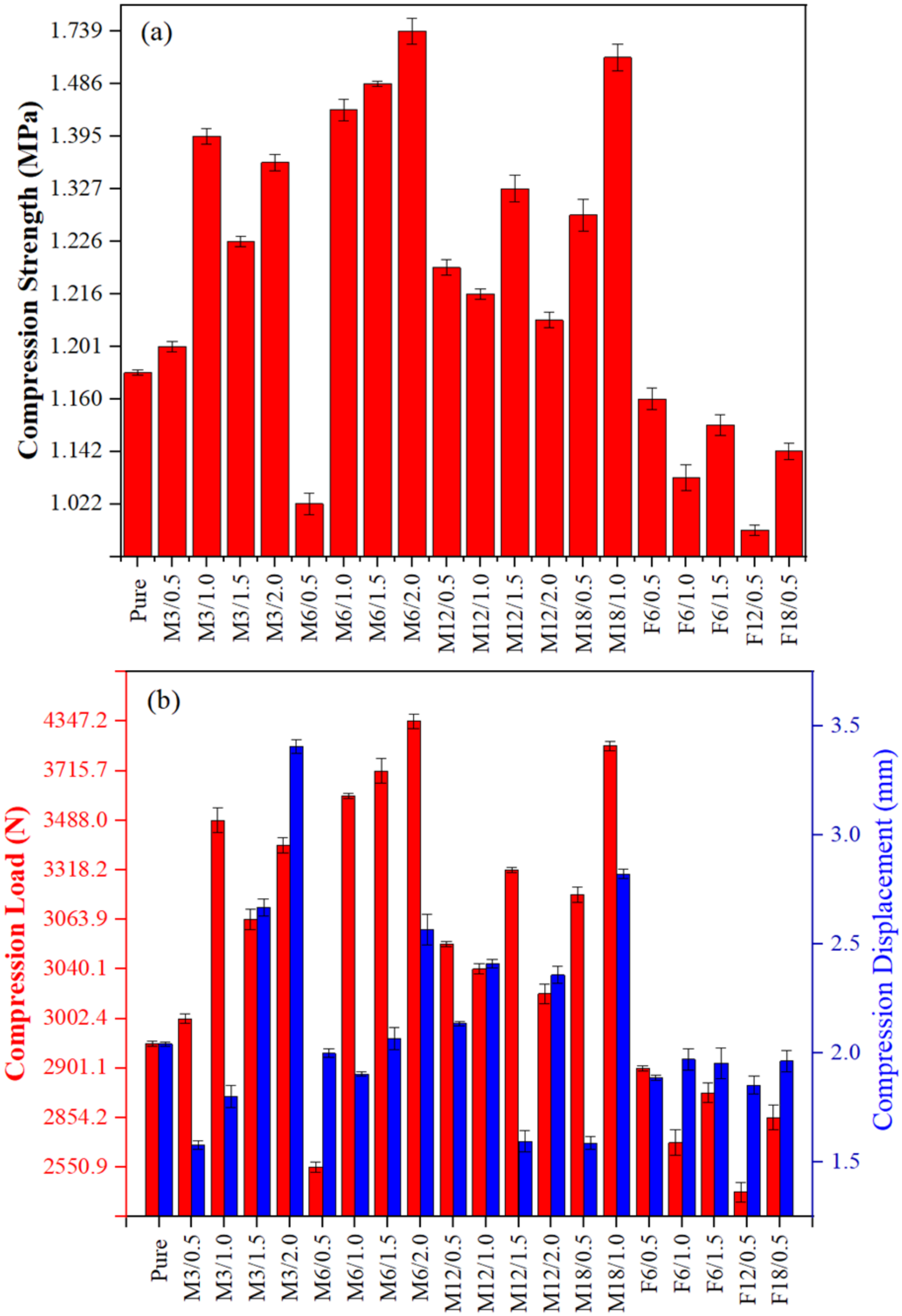

Figure 5. *Compression test results of RPU composites*

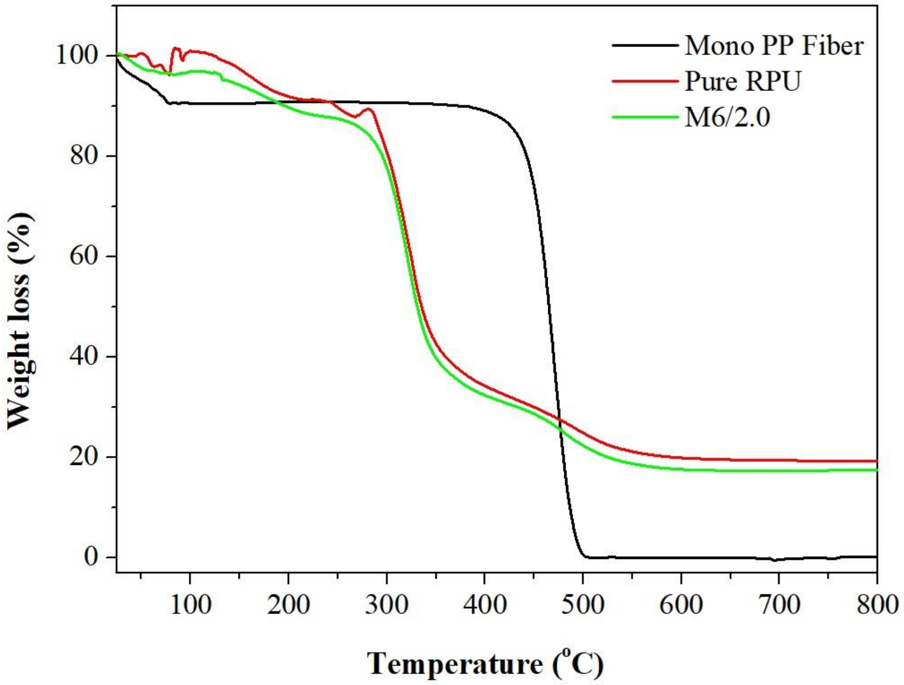

Figure 6. *TGA curves of monofilament PP fiber, unreinforced RPU and RPU composites (M6/2.0)*

In the light of SEM images in Figure 7, it can be stated that composites contain closed cells in spherical and polygonal form. Also, cell sizes generally appear to decrease with increasing PP fiber content. The presence of small cells around PP fiber indicates that cell growth is inhibited by fiber. In other words, PP fibers in RPU composites physically inhibit growth of bubbles [31,32]. Moreover, this phenomenon may result in a uniform infusion of components and formation of homogeneous and high-density composites.

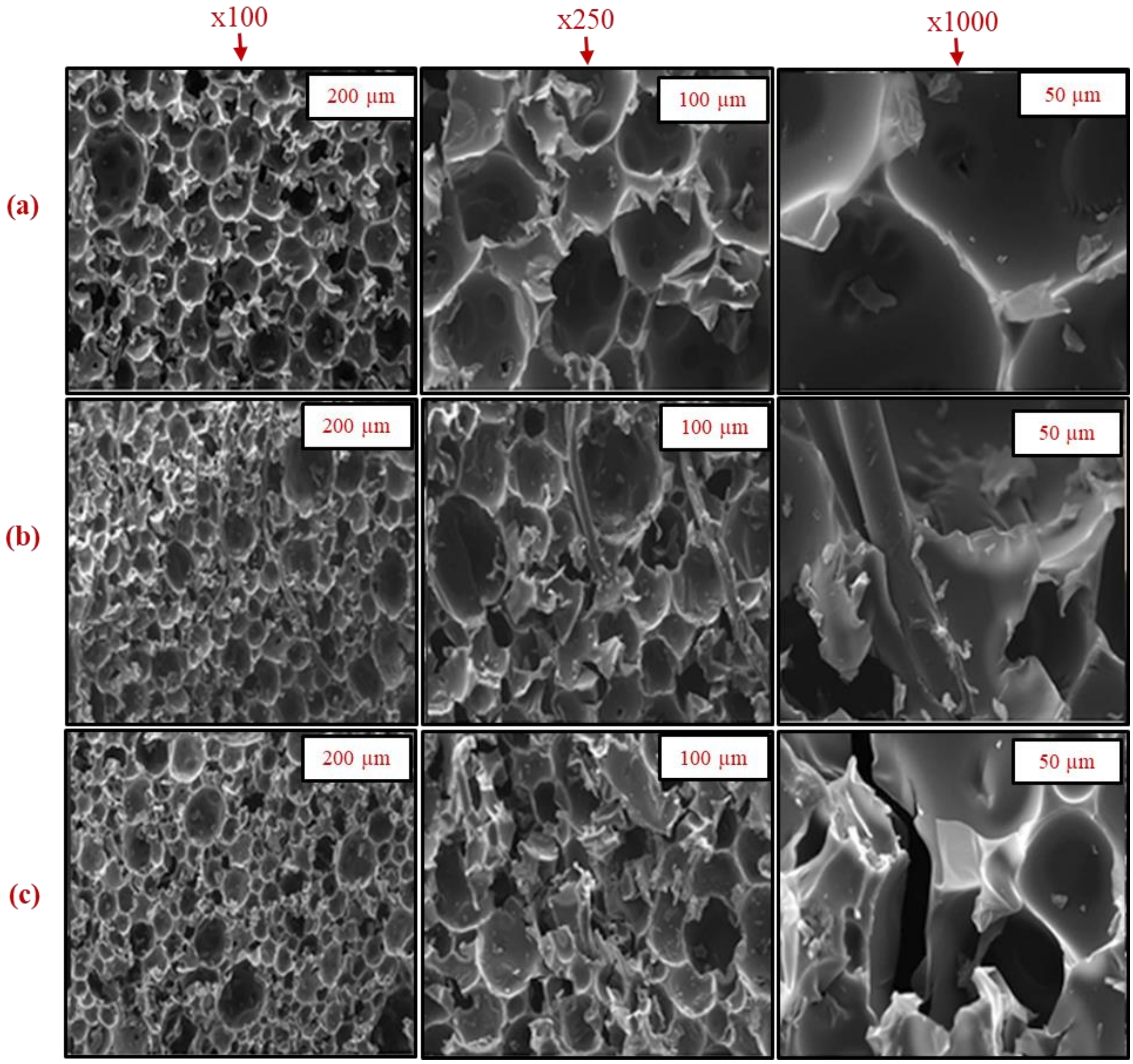

Figure 7. *SEM images at different magnifications of pure RPU* ***(a)****, M6/1.5* ***(b)*** *and M18/1.0* ***(c)***

IV. CONCLUSION

In current paper, PP reinforced RPU composites that were employed as modeling support apparatus in ceramic casting industry were successfully produced to improve mechanical properties of RPU composites. For this, PP fibers in monofilament (mono) and fibermesh (fibril) forms were added to RPU matrix at varied rates (0.5, 1.0, 1.5, and 2% by weight) and in lengths ranging from 3 to 18 mm. Research results are as follows;

• Specimens' bulk densities, with and without PP fibers, range from 72.15 to 146 kg/m^3. Compared to pure RPU, bulk density of monofilament PP reinforced composites significantly increased and the highest density value (146.86 kg/m^3) was reached in M6/2.0 sample. However, addition of fibrilmesh resulted in a reduction in bulk density.

• Flexural strength increased with an increase in mono PP %, but decreased with the presence of fibril PP. All mono PP reinforced composites, with exception of M6/0.5, have compressive strengths greater than pure RPU. M6/2.0 PP reinforced RPU composites showed excellent results in both bending and compression tests, increasing strength by 47%. The presence of fibrilmesh PP fiber had a negative effect on mechanical strength of composites. This can be attributed to fiber not being homogeneously dispersed in matrix.

- According to SEM analysis, incorporation of PP fibers generally decreased quantity of closed cells in composites.

Experimental findings indicate that fiber type, content and length affect the mechanical performance of RPU composites. In light of this, support equipment for ceramic casting sector can be made of RPU composites reinforced with mono PP fiber.

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