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Influence of filler proportion on mechanical and physical properties of particulate composite

A. Aruniit[1], J. Kers[1] and K. Tall[1]

[1]Department of Materials Engineering, Tallinn University of Technology, Ehitajate tee 5, EE19086 Tallinn, Estonia; e-mail: aare.aruniit@ttu.ee

Abstract. A particle reinforced composite material consisting of unsaturated polyester (UP) resin and fine dispersion of alumina trihydrate (ATH) has good mechanical properties. It performs well as laboratory or culinary bench top, in bathrooms as vanity top or sanitary ware, in maritime and agricultural applications. The goal of this study is to find out how the filler percentage in the composite influences the mechanical properties of the material and if it is possible to increase the concentration to lower the price. Test slabs with different proportion of filler were fabricated with vacuum assisted extruder. Test specimens were cut from post cured sheets and hardness, flexural strength, flexural modulus, deflection, and density were measured. It is shown that with higher percentage of ATH the flexural strength decreases and hardness and flexural modulus increases together with density.

Key words: Particulate composites, mechanical properties of particle reinforced material, filler proportion.

INTRODUCTION

There is a variety of particle filled composites consisting of thermosetting polymer matrix and particulate reinforcement. Resin is usually unsaturated polyester, acrylic, or polyester modified with acrylic type of resin. Most common fillers are sand, calcium carbonate, alumina trihydrate, and quartz (ICPA, 2003).

An important way to classify these composites is to make a difference between products having surface coat (gel coat) and others having consistent composition throughout the material. Products having surface coat are commonly referred to as artificial marble or artificial onyx. Products that are consistent throughout the material are known as solid surface or engineered stone (ICPA, 2003).

In addition, composite often comprises colour pigment, resin chips for imitating natural stone, accelerator or inhibitor to control the polymerisation process, additives to influence flow characteristics, internal release agents, etc. Nevertheless, the matrix and the reinforcement pose the main influence on all properties.

The choice of materials is based on the requirements. Unsaturated polyester resin modified with acrylic is the best compromise between the cost and mechanical properties. Alumina trihydrate (Fig. 1) is a non-toxic, non-corrosive, non-cancerogenic, odourless, flame retardant filler that provides good whiteness if white products are desirable. It is a mineral derived from bauxite. ATH has specific gravity of 2.42g cm^{-3} and Mohs' hardness index of 2.5–3.5 (Ash, 2007).

The composition of the composite depends on the product. Filler content influences the morphological properties of the casting dispersion. If the product is a simple slab the filler content can be as much as 66–92 wt% (DuPont, 2008; 2009). When casting products like washbasins or bathtubs the filler content is usually around 55–62% (Cook Composites & Polymers, 2009). The filler content is lower because the casting dispersion has to have better flow characteristics to overcome narrow openings. The filler content influences the shrinkage of the product as well. More filler means less shrinkage and residual stress (Katz & Milewski, 1987).

Figure 1. Alumina trihydrate.

Besides the physical and mechanical properties the filler amount influences the cost of the product. By lowering the resin content in the composition of the particle filled composite one can lower the costs (Zurale & Bhide, 1998). Resin is relatively expensive compared to all kinds of fillers. This is the main motivation that drives the manufacturers to search for possibilities to lower the filler content.

Flexural strength and surface hardness are most important mechanical parameters for brittle materials like particle reinforced thermosets (Preis, 2004). Flexural strength describes the ability of materials to withstand deformation under load. Material hardness gives info about material resistance to plastic deformation by scratching, abrasion, or indentation. These are the most sought properties for products, such as bench tops, washbasins, shower trays, wall and floor claddings.

An important technological step in the fabrication of thermosetting composites is post cure of the material. Post cure is a process to increase the amount of cross linkage in the composite by raising the composite's temperature. The maximum cross-link density is when every carbon to carbon unsaturated group has been reacted and each end group of each chain is connected to another polyester chain. Short post cure times are made possible with ovens that elevate temperatures to the resin’s Glass Transition Temperature (T_g) and higher (Lipovsky, 2006).

This study investigates how the amount of particulate reinforcement influences the mechanical and physical properties and cost of the particulate composite.

MATERIALS AND METHODS

For determining of mechanical and physical properties 3 material specimens were cast (Table 1). The filler percentage range was chosen based on the recommended filler concentration for casting of shapes (55–62% (Cook Composites & Polymers, 2009)).

For the fabrication of specimens an unsaturated polyester casting resin based on isophthalic acid and neopentyl glycol was used. The resin is developed to produce non-gel coated products and contains methyl methacrylate. It is a pre-accelerated, medium reactive, low viscosity resin. For curing a methyl ethyl ketone peroxide with resin to hardener ratio of 1/100 wt% was added. As filler, ATH with medium particle size was used. Preliminary cure was done at room temperature (23 ± 2°C). That was followed by post cure at 40°C for 12 h. 500 x 1,000 x 10mm slabs were cast. The slabs were cast with a closed mould with a special vacuum assisted casting machine ADM-KSA 10/15 VAC F. 0.85 bar vacuum was applied. A closed mould guarantees equal thickness and flatness of the slab. That is necessary to get the test specimens as specified in the testing standards. Vacuum chamber of the machine removes air from the casting dispersion and helps to achieve non-porous material. The proportion of filler and other components is controlled by the machine. From all materials 5 specimens were cut for all tests. The test specimens were cut from the slabs with water jet.

Table 1. Test specimens.

#	Proportion of filler wt.%	Post cure temperature °C	Post cure time h
1	55	40	12
2	60	40	12
3	65	40	12

The flexural properties of the material were determined by 3 point bending test (Fig. 2) as specified in ISO 178 Plastics – Determination of flexural properties. The test specimens were with dimensions 50 x 300 x 10mm. A test speed of 2mm min^{-1} was used.

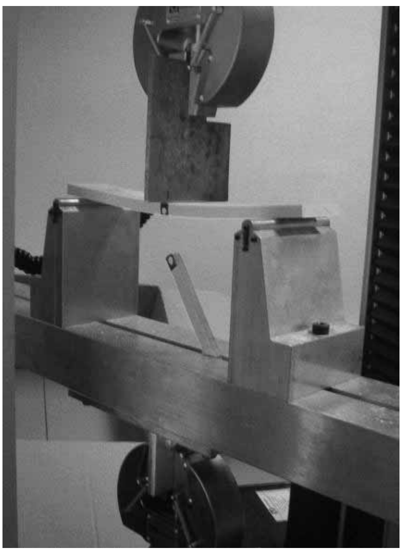

Figure 2. 3 point bending test.

The indentation hardness of the material was measured by Zwick-Indentec 8150 LK according to ISO 2039 Plastics – Determination of hardness. Rockwell M hardness scale was used (ball diameter 6.35mm and major load 980.7 N). Comparative hardness measurements were taken with GYZJ 934-1 Barcol impressor according to ASTM D 2583-99.

The density of the material was determined by weighting method. A kit of analytical scale and a weighting jig was used for weighting a specimen of the material in air and fluid. The density is determined by equation:

$$\rho = \frac{m_{S\tilde{o}} \cdot \rho_v}{m_{S\tilde{o}} - m_{Sv}} \tag{1}$$

where $m_{S\tilde{o}}$ is specimen weight in air, m_{Sv} is specimen weight in fluid, and ρ_v is density of the fluid.

RESULTS AND DISCUSSION

The test results are presented in Table 2. Besides the properties of the tested materials, the table contains also properties of similar material Corian® produced by DuPont™ that is fabricated from acrylic resin and ATH with 33:67 ratio (DuPont, 2009). The table contains properties of a 12mm thick Corian® sheet that is the closest thickness to the test specimens. Additionally, properties of pure resin are presented for comparison.

Table 2. Mechanical and physical properties of particulate composites and resin.

#	Proportion of filler wt.%	Flexural strength MPa	Flexural modulus MPa	Deflection mm	Barcol hardness	Density g cm^{-3}
1	55	61.4±3.21	10,960±1,168.10	8.85±0.45	50	1.64
2	60	60.2±6.51	13,094±2,047.09	7.23±0.40	51	1.69
3	65	57.5±7.32	16,955±2,147.60	5.10±0.50	53	1.77
Corian®	67	57.1–74.0	8,040–9,220	–	56	1.68–1.75
Resin	0	140	3640	–	40–45	1.10

The flexural strength is considered to be a demonstrative parameter for brittle materials, such as polymers and ceramics (Guhanathan, 2002; Preis, 2004). The stress-strain curves showed that the mechanism of fracture was brittle. Only slight plastic deformation occurred after elastic deformation before breakage. Due to unfavourable geometrical features particulate fillers could only moderately increase the modulus, while flexural strength remains the same or decreases (Julson et al., 2004; Xanthos, 2005). The same can be observed from the test results in Table 2. On increasing the weight percentage of the ATH from 55 to 60, the flexural strength decreases 1.2MPa.

On further increase of filler, the flexural strength decreases 2.7MPa. The decrease can be considered small or even nonexistent when considering the standard deviation of the flexural strength values. The material under observation shows similar values of flexural strength as in the commercially available Corian®. The flexural strength of neat resin is 2.3 times higher then that of the composite. This confirms that the elastic properties of the composite decline by lowering the wt% of matrix.

The increase of flexural modulus shows that the stiffness of the composite increases by adding more filler. That trend is confirmed by the deflection values that decrease as the filler content increases. As it was with flexural strength, the change in flexural modulus is small. But when comparing the composite with 65 wt% of filler to the Corian®, the flexural modulus is 7,735MPa higher. This is a great difference. Stiffness is a good property generally when considering the application of the material. On the other hand, the material gets more brittle as the flexural modulus increases. Brittleness is not the material property a manufacturer of bench tops or washbasins has sought for because it makes the products prone to cracks and breakage by falling objects. The flexural modulus of composites containing almost equal amount of matrix and resin is influenced by the ratio of the moduli of the 2 phases and the percentage of the filler (Xanthos, 2005). What is more, stiffness and impact strength are dependent on particle size (Hayes & Seferis, 2001). Smaller particle size provides higher stiffness. Impact strength can be increased by smaller particle size or by adding impact modifier. In this case it might be necessary to increase particle size to decrease stiffness, because the reduction of filler wt% is not desirable. At the same time the effect of bigger particles on other properties must be observed.

Hardness test is a simple one and gives good info on the microstructure relationships of polymer composites (d’Almeida, 2001). The composite was manufactured with a vacuum assisted casting machine that produces air void free casting dispersion. The filler is a medium fraction ATH and the filler wt% is more than half in the mixture. All this should assure homogeneous material. When the hardness test was conducted this assumption was verified by measuring the hardness on both sides of the test specimen. No discrepancy was found. The hardness values show that an increase in filler increases the hardness. Material with 65 wt% of filler has Barcol hardness of 53 compared to the 50 that was measured in material with 55 wt% of filler and 20% higher hardness than with neat resin. The comparison of hardness of plain filler and matrix is complicated because the hardness value of ATH is given in Mohs’ index and there is no direct conversion to other hardness scales. An approximate conversion was done. The conversion of ATH’s Mohs’ index of 2.5 (2.5–3.5) to Brinell scale (10mm ball 500kg load) is 90. The hardness of resin on Barcol scale is 40 (40–45) and its conversion to Brinell scale gives a value of 25 (Jones et al., 1996). When comparing these values it seems understandable that the hardness of the composite increases by the increase in wt% of ATH. Nevertheless, the concentration of particles in the composite must be greater that 30% to obtain a useful increase in hardness. Besides the wt% of filler, hardness is also influenced by the fraction and size of the filler. The filler should contain both small and large particles to fill all the gaps (d’Almeida, 2001). Corian® has in datasheet besides the hardness in Barcol scale also Mohs’ index of 2–3 that is lower than that of ATH (2.5–3.5) (DuPont, 2009). Table 2 shows that the hardness of the tested material is 5% lower than that of Corian.

A downside to the increase of the filler is increase in density that influences the weight of the products. Heavier products are more difficult to handle in production and transport. Heaviness is an undesirable property in marine and other applications. The increase in density is 7% when increasing the filler 10 wt%.

As stated in the objective of the study, one of the goals is to evaluate the effect of filler wt% increase in cost. Fig. 3 presents the price trend when filler concentration rises. By adding 5% of filler the price decreases 3.5% and by adding 10% it decreases 7.5%. Cost reduction is the trend a manufacturer is looking for.

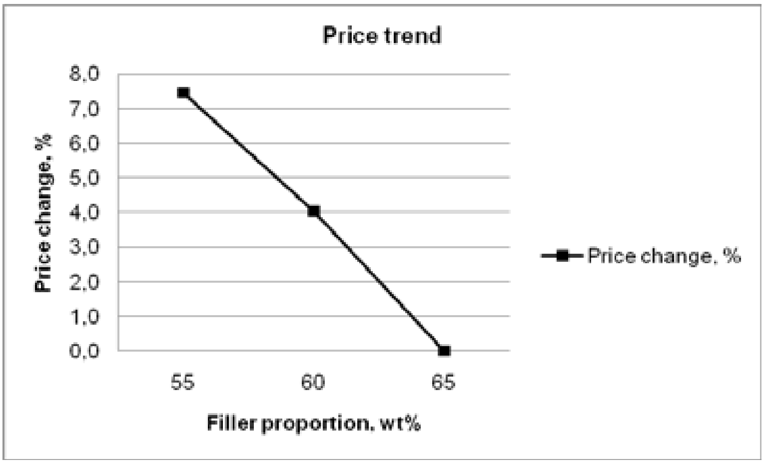

Figure 3. Price change compared to filler wt%.

CONCLUSIONS

This research was carried out to study the effect of filler wt% on the mechanical and physical properties and on the cost of particle reinforced composite. The experimental part included making material specimens and testing their flexural strength, flexural modulus, deflection, hardness, and density.

Flexural strength decreased 3.9MPa when 10% of filler was added. Contrary to that flexural modulus increased 35%. The same is confirmed by the deflection values. The hardness on Barcol scale increased 5.6% by the 10% increase of filler, but compared to neat resin the composite had a 20% higher hardness value. As more filler was added the density also increased 7% in all. Finally the cost reduction of the composite was calculated and that showed a 7.5% decrease in net price.

From the experimental data one can conclude that the manipulation of filler wt% influences many variables and there are aspects like particle size and fraction to be considered. When compared to the materials commercially available on the market today, the tested material showed comparable results with only flexural modulus being remarkably higher. Further study on the influence of particle physical properties on the mechanical properties of the composite should be carried out.

The test results obtained betoken a prospect for the tested material to be used commercially as a material for laboratory, culinary, marine or agricultural products.

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