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The Influence of Industrial Fiber Waste Amount on Physico-mechanical Properties of Polyester Matrix Composite

Acikbas G[1,2], Gocmez H[2]

[1]Vocational School, Bilecik Şeyh Edebali University, Metallurgy Program, Bilecik, Turkey
[2]Department of Materials Science and Engineering Dumlupınar University, Kutahya, Turkey

***Corresponding author**
Acikbas G

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Abstract: Glass fibers find use in automotive, building, marine, wood and numerous other industries, reaching 5 million tons annual consumption, mainly comprising the reinforcing phase in polymers. The high amount of industrial demand have also caused an increasing research interest for their recovery, recycle and reuse as solid waste treatment and utilization for cost effective and environmentally friendly production processes. In this study, the effects of matrix/fibers ratio on the mechanical and physical properties of glass fiber reinforced composites were investigated. The properties of polyesters were compared with the reinforced polyester. The microstructure of the composites was characterized by SEM-SE technique. It was determined that with the increasing glass fibers content, the flexural and impact strength were reduced, while the flexural modulus and hardness were enhanced and the total porosity was increased.

Keywords: characterization, glass fiber, waste, mechanical properties, polyester, composite

INTRODUCTION

Many of our modern technologies require materials with unusual combinations of properties that cannot be met by the traditional materials. Combination of material properties are extended with the development of composite materials [1]. A composite term is defined the combination of physical bonded two or more constituent materials and developed composite material properties are superior than the constituents. One of the constituents is generally stronger, stiffer and low cost is called filler (active or reactive) and the other phase is called as matrix, its less stiff and weaker [2].

The properties of composites materials are affected by the type of filler in composite mixture. Generally the nature of filler, shape and size and size distribution of the filler, filler to resin ratio, the bonding between the filler and the resin, chemical compositions of filler and additives for production (e.g. hardener, accelerator etc.) affect the properties of polymer matrix composites [3-7]. Mechanical properties of fiber reinforced composites are depending on the properties of the constituent materials (type, quantity, fiber distribution and orientation, void content). On the other hand interfacial bond strength between the matrix and reinforcement and the mechanisms of load transfer at the interphase also play an important role [8, 9].

In recent years, fiber reinforced polymer composites are becoming increasingly widespread in manufacturing industries, including aerospace, marine, automotive, furniture, electrical and sport equipments. Glass fiber is the most common reinforcement due to low cost and good mechanical properties. There are lots of studies about glass fiber reinforced composites [10-12]. Annually, fiberglass industries purchase 3 million tons of recycled glass, which is used in the production of new fiberglass products [13]. There are lots of benefits to use of waste fiber glass in respect of energy saving, ecology, environment and so on. Besides, governmental policies support reuse and recycling of products. The management of fiber waste is an important issue because of the increase in the production of composite materials [14, 15]. There have been quite a number of studies on the use of waste materials as filler in polymer matrix. With regard to the environmental aspects it is very important to reevaluate this kind of waste materials to convert to value added product [14, 16-18]. Therefore this study focused on the evaluation of waste glass fibers in polyester matrix in order to enhance the mechanical properties of polymer and convert glass fiber waste into eco-friendly and value added product. In this scope varying amounts of glass fiber waste was added into polyester matrix and mechanical and physical properties of composite was discussed in accordance with to filler:polymer ratio.

MATERIAL AND METHOD

Materials

The matrix material comprises polyester resin, hardener and accelerator which were supplied from Poliya Composite Resins and Polymers Inc., Turkey. Waste glass fiber was obtained from Sisecam Cam Elyaf Company, Gebze, Turkey. Glass fiber waste material was milled with a Fritsch Pulverisette-9 for one minute at 1000 rpm. The grounded glass fiber waste was dryied and then sieved for five minutes with Vibratory Sieve Machine separated to particle size below 90 µm.

After that some characterization techniques were applied to glass fiber waste material. The theoretical density of glass fiber waste was measured by He-gas pycnometer. Scanning electron microscopy images of waste materials were taken with FEG-SEM(Zeiss) with secondary electron detector. The types of phases were detected by means of X-ray diffraction analyses (Panalytical-Empyrean model with Cu-Kα radiation). Elementel analysis of waste materials was done with WD-XRF (Rigaku Primus). The schematic presentations of preparation of glass fiber waste and characterization steps are given in Figure-1.

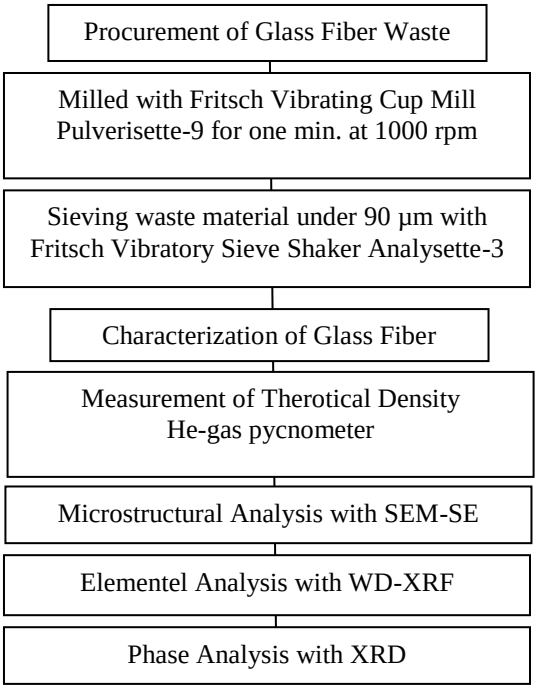

Fig-1: The schematic presentation of preparation of waste and waste characterization steps

Preparation of composites

The objective of the study was to determine the effect of waste glass fiber to polyester resin ratio on the mechanical and physical properties of composites. Also compared properties of neat polyester and reinforced polyester. During the manufacturing of composite material, the amounts of hardener and accelerator was kept constant and waste glass fiber to polyester resin ratio compositions was changed between 0.33-1.00-1.22-1.50-1.86, respectively. Neat polyester is coded as W0. Prepared composite's recipes were shown in Table-1.

Table-1: Prepared composite recipes

Recipe Codes	Filler %wt.	Polyester %wt.	Filler:Polyester
W1	25	75	0,33
W2	50	50	1,00
W3	55	45	1,22
W4	60	40	1,50
W5	65	35	1,86

Waste glass fiber reinforced polyester matrix composites were produced with the casting method. Polyvinyl alcohol (PVA) was used as lubricant to easy remove the composite from mold. Polyester resin, accelerator, waste

particles and hardener were blended in a propeller mixer between 300-1500 rpm for 15 minutes. Initially slow mixing prevents dust from flying. Once the waste filler was incorporated into the liquid polymer the mixing speed was increased stepwise to obtain a homogeneous mixture. Vortex formation was avoided to minimize trapped air bubbles. The blended mixture was poured into the lubricated steel molds. The polyester resin-fillers blend was left for 1-4 hours in the mold at room temperature for curing, and thereafter the polyester resin composites were removed from the metal mold. Finally, in order to complete the curing reaction post cure step was applied at 70^{o}C for 2 hours. The composite preparation process is given in Figure-2, schematically.

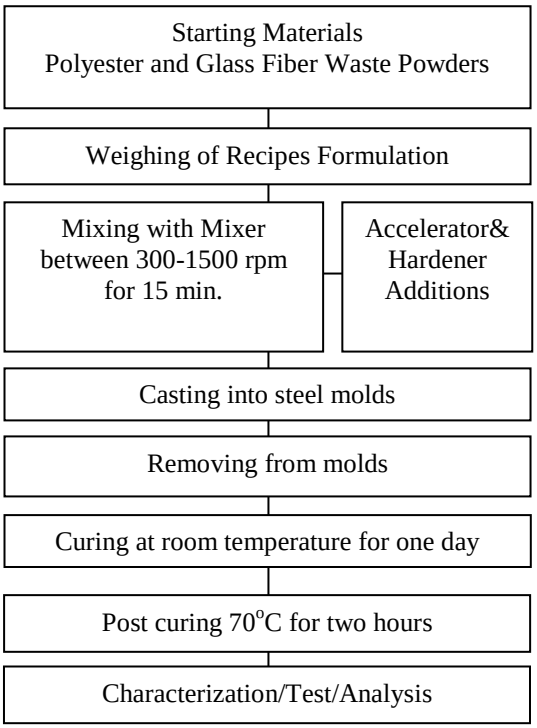

Fig-2: Polymer matrix composite preparation steps

Physical, mechanical and microstructural characterizations

In order to determine the theoretical density value of the composites, the theoretical densities of the glass fiber waste material and the polyester resin were used. Theoretical density of composite samples was calculated from the theoretical densities and the volume fractions of the constituting materials given by Eq. 1 [19]. The known weight fractions were converted to volume fractions.

$$\rho_c = (\rho_m * V_m) + (\rho_g * V_g) \quad \text{(Eq.1)}$$

where, ρc is theoretical density of composite samples, ρm is theoretical density of matrix, ρg is the theoretical density of waste glass fiber filler, respectively, while Vm is the vol. fraction of the matrix, and Vg is the volume fraction of the respective filler.

The Archimedes principle was used to measure the density and porosity of the samples. Bulk density, % theoretical density and % total porosity were calculated by Eqs 2-4.

$$\text{Bulk Density}(\text{B.D.}) = \frac{W_1}{W_3 - W_2} * \rho_{water} \quad \text{(Eq.2)}$$

$$\%\text{Theoretical density} = \frac{\text{B.D.}}{\text{T.D.}} * 100 \quad \text{(Eq.3)}$$

$$\%\ \text{Porosity(Total)} = (100 - \%\ \%\text{Theoretical density}) \quad \text{(Eq.4)}$$

where, W1 is the dry weight, W2 is the wet weight suspended in water, W3 is the wet weight, B.D. is the bulk density, T.D. is the theoretical density of the samples.

The ShoreD hardness was measured with samples in 5cmx5cm dimensions. The average of the five measurements was taken. Three point-flexural strength tests of the samples were done in compliance with ASTM D 790 / ISO 178 standard. At five measurements were used to calculate the flexural strength and flexural modulus (Eqs 5-6).

$$\sigma_{bs} = \frac{3.P.L}{2.W.D^2} \quad \text{(Eq.5)}$$

$$E = \frac{L^3.m}{4.W.D^3} \quad \text{(Eq.6)}$$

where, σ_{bs} is flexural strength (N/mm^2-MPa), P is loading force (N), E is the flexural modulus (GPa), m is slope, L is the distance between the span (mm), W is the width (mm) and D is test sample thickness (mm).

Izod impact resistance of all composite samples was measured with Devotrans impact machine. The tests were carried out in compliance with ISO 180:2000 standard. The samples of sizes 80x10x4 mm were subject to the impact of 6J hammer. Equation 7 was used to calculate the impact energy. In equation 7, the abbreviations mean the following; E_c: Energy value calculated by the device (J), h: sample thickness (mm) b: sample width (mm) a_{iu}: impact strength (kJ/m^2).

$$a_{iu} = \left(\frac{E_c}{h.b}\right) * 10^3 \quad \text{(Eq.7)}$$

The microstructural examinations of the polished composite surfaces were performed by scanning electron microscope imaging. Secondary electron scanning electron microscopy images of composite materials were taken with FEG-SEM(Zeiss Supra 40 VP).

RESULT AND DISCUSSION

Glass fiber waste characterizations

Waste particles were ground and sieved to obtain particle size below 90 µm. Scanning electron microscopy images of waste glass fiber showed that particles had elongated shape with the aspect ratio changing from 1 to 3. On the other hand, the ground waste glass fiber particles were wide size range. The particle shape and distribution are important parameters that affect the process ability (e.g. rheological properties) and a number of mechanical and physical properties of composites. The as received state image of the glass fiber waste that was formed during the production of chopped strand glass fibers is shown in Figure-3a and representative SEM-SE image of glass fiber waste was given in Figure-3b. The mean particle size of glass wastes slightly changed from 5 to 10 µm with broad distributions. Coarse particle's sizes are reach 25 to 30 µm.

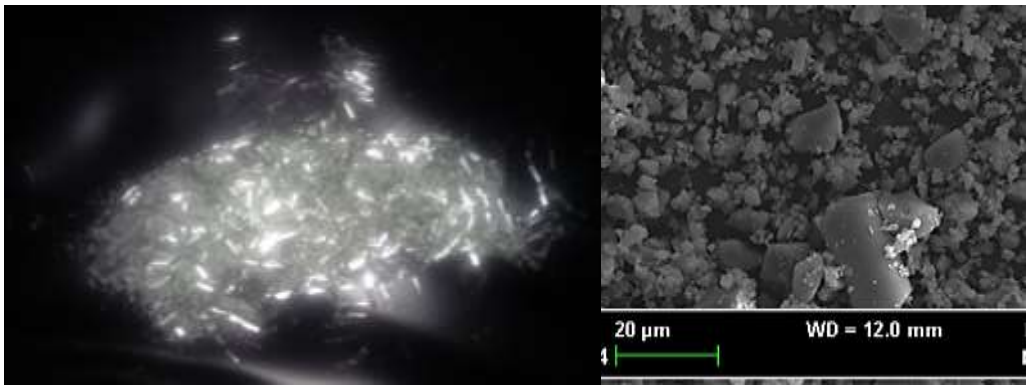

Fig-3: a) As received state of the glass fiber waste image, b)SEM-SE image of waste glass fiber,1000x

He-gas pycnometer results of waste glass filer and polyester matrix were 2,61g/cm^3 and 1,20g/cm^3, respectively.

Elemental analyses of waste glass fiber materials were carried out by WD-XRF technique. Si, O, Ca and Al were the main elements in the waste glass fiber sample with little amounts of Mg and others. The results showed that the

waste material composed of SiO_2, CaO, Al_2O_3 and little amount of MgO. The result of XRF analyses of the waste material is shown in the Table-2.

Table-2: WD-XRF analysis result ofwaste glass fiber

Oxide	wt.%
SiO_2	60,02
CaO	20,87
Al_2O_3	15,23
MgO	1,48
Others	2,40

Phase analysis of waste glass fiber materials were conducted by XRD technique. The very broad peak in waste glass fiber X-ray diffraction pattern indicated that the main phase of waste glass fiber was amorphous. XRD patern of waste glass fiber was given in Figure-4.

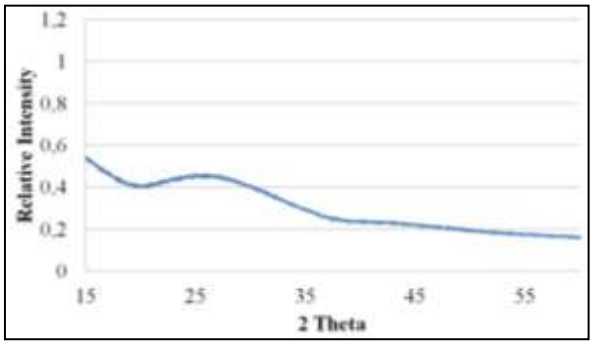

Fig-4: X-ray diffraction pattern of waste glass fiber

Physical properties of composite materials

Bulk density, theoretical density, and %total porosity increased while %theoretical density decreased with increased waste glass content. Increase in bulk and theoretical density with increase in waste amount is resulted from the density of waste glass fiber is higher approximately 117% than polyester matrix. Decrease in theoretical density% and increase in total porosity is arises from porosity evolution during casting due to high viscosity of mixture with increase in waste content. With the decrease in waste glass fiber content, viscosity of mixture decreases and hence wetting of matrix to filler is enhanced. Total porosity content of W5 coded composite is higher about 79% than W4 sample. This can be resulted from worse wetting behavior of W5 composition with increases in filler content.

In our previous study showed that when the coarse waste glass fiber particle size (150 µm) was used better process ability behavior and hence has the lowest porosity evolution and highest bulk density value of composite was achieved [20]. The theoretical density, bulk density, %theoretical density values increased and total porosity level reduced with an increase in glass fiber waste content and particle size of added waste material. Finer particle size lead to higher surface area and processability of composite mixture is deteriorates. Therefore, a composite consisting of fine particle size of waste causes more porosity evolution and less theoretical density. Physical properties of composites were given in Table-3.

Table-3: Theoretical density, bulk density, %total porosity %theoretical density of composite samples

Recipe Codes	Bulk Density	Theoretical Density	%Theoretical Density	%Total Porosity
W5	1,770	1,85	95,49	4,51
W4	1,735	1,78	97,48	2,52
W3	1,670	1,71	97,56	2,44
W2	1,610	1,65	97,65	2,35
W1	1,370	1,39	98,39	1,61
W0	1,185	1,20	98,75	1,25

Mechanical Properties of Composites

The mechanical test results of the composites are given in Table-4. Addition of waste glass fiber particles into poly**es**ter resin caused an increase of the hardness of the monolithic polyester resin. The hardness increased only slightly with increased amount of waste glass fiber particles. However with the addition of the fillers after that 1,5 filler : resin ratio the hardness was not affected appreciably. With increase in filler content, Shore-D Hardness increased with % 4.9, 7.3, 8.5, 11.0 and 11.0, respectively according to polyester matrix.

When the flexural modulus is analyzed, as expected, the rigidity of the glass fiber amount is more and its flexural modulus value is much higher. Flexural modulus of the monolithic polyester is much lower compared to that of the composite which have high glass filler content. This is arises from the high filler amount and high hardness of filler. The porosity level in composites is another influencing factor causing the flexural modulus to be low. The results indicated that flexural modulus increased with increase in filler content.

Flexural strength of composites is lower compared to monolithic polyester. The amount of filler is increased in composite mixture which causes the flexural strength to decrease. Since waste glass fiber filler phase is more rigid compared to the polyester, as the filler phase amount increases, the flexural strength decreases. Due to filler addition and thereby reducing the deformability of matrix, so that the composite tends to form a weak structure also. According to the study conducted by Hanna and co-workers, it is stated that polyester matrix composites contain 15%wt. $CaCO_3$ and 9%wt.$MgCO_3$ have flexural strength of 97.86 MPa, 75,46 MPa, respectively [21]. This is resulted from the high flexural strength of polymer matrix. Koleva *et al*., noted that as the filler phase increases, the flexural strength decreases [22]. The reason for the decrease in flexural strength is because the interaction between matrix and filler phase weakens as the filler amount increases. In order to strengthen the interaction between the phases and to improve the properties of the composite, either the filler phase or the polymer should be modified. Madugu and et al. stated that the homogenous distribution of the filler in the polyester matrix is the most influencing parameter that affects the mechanical properties [23]. Ahmad and et al. state that the agglomeration of the filler phase within the material causes non-homogenous distribution which results with the weakening of filler-matrix interface and strength [24].

The main increase in flexural modulus value is because the ceramic filler phase has a higher flexural modulus value compared to that of polymer matrix. Elongation ability decreases with the addition of the filler phase because the movement of the filler phase or the deformation of the matrix gets harder and as the filler phase increases, deformation decreases. The studies under the literature data reflect similar results [21, 25].The increase in glass fiber content led to weaker interphase bonding, and hence, caused deterioration of the impact resistance of composites.

Table-4: Mechanical properties of composites and monolithic polyester

Recipe Codes	Shore-D Hardness	Flexural Strength (MPa)	Flexural Modulus (GPa)	Impact Resistance (kj/m^2)
W5	91	57,8	9,52	5,8
W4	91	59,0	8,56	6,1
W3	89	62,0	7,62	6,2
W2	88	70,8	6,54	6,8
W1	86	80,8	3,76	8,3
W0	82	105,2	3,20	10,7

Microstructural Characterization of Composites

SEM-SE images of composites were given in Figure-5. The images reflected that increased in filler content lead to weak interface bonding between matrix and reinforcement. Besides grain pull-out and pore evolution were increased with increase in glass fiber content. These kinds of faults caused to deterioration of physical and mechanical properties.

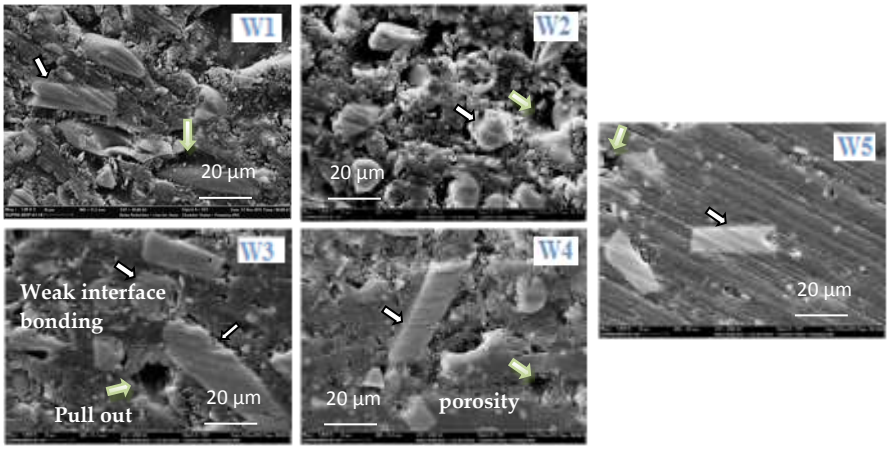

Fig-5: SEM-SE images of composites

CONCLUSIONS

The study was carried out in three main parts. The first one was characterization of glass fiber waste particles, the second was the effect of waste glass fiber filler:polyester matrix ratio on properties of composites and the third was comparison of properties of neat polyester and reinforced composites.

The characterization of waste glass fiber particles was performed with different characterization techniques. The waste glass fiber particules had smooth surfaces which in turn caused a weak polyester matrix-filler interphase bonding with the matrix, affecting the mechanical properties of composites negatively.

Theoretical densities of the glass fiber and polyester matrix were 2.61 gr/cm^3 and 1,20 g/cm^3, respectively. XRF analyses showed that the waste glass fiber was mainly composed of SiO_2, CaO and Al_2O_3. XRD analysis revealed that glass fiber had amorphous phase.

Bulk and theoretical density values of composites were increased and changed between 1.37 g/cm^3 to 1.77 g/cm^3 and 1.39 g/cm^3 to 1.85 g/cm^3, respectively.

Addition of hard particles into polyester matrix caused increase in hardness, flexural modulus and decrease in flexural strength and impact resistance. With increase in waste glass fiber content, viscosity of mixture increased and hence wetting behavior of matrix was getting worse. Therefore bond strength between the matrix and filler was weakened, voids formation was observed and mechanical properties deteriorated.

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