Influence of silane coupling agents on the mechanical properties of recycled glass fiber-reinforced composites

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Abstract. In this paper a feasible process to reuse glass fibers separated from a wasted glass mat as reinforcement in high density polyethylene (HDPE) was described. Glass fibers were recovered by pyrolysis, chopped and surface was treated by silane as adhesion promoters for increased composite interfacial strength before compounding. FTIR spectroscopy was used to observe the silane coupling agents adsorbed onto glass fiber surface after hydrolysis. Composite was produced by hot compression. Macroindentation and compressive strength tests were performed to characterize interface strength of composite material.

Introduction

In fabrication of glass fiber mat a felt made of randomly oriented short fibers cuts to shape called preforms, which are impregnate with urea-formaldechide. In-plant scarp or end-of-life products made of cross-linked fiber-reinforced thermosetting polymers cannot be re-melted and shaped into new products, like in themoplastic composites technology. So, there is a need to developp a process for recovery and reuse of glass fibers from wasted mat as reinforcement in another polymer composite materials. One of the techniques to re-use thermosetting composites is pyrolysis- the reinforced composites recover by heat- annealing of material.

In this study in-plant scarp of glass fiber mat has been undertaken of pyrolysis, and recovered glass fibers have been chopped and compounded by compression molding.

Since the innovation of the manufacturing techniques for polymer composites, the performance of composites has been improved, and the study of the interfacial adhesion between the fiber and the resin is also becoming important. Organo-silicon compounds are an obvious choice as potential coupling agents [1] for glass fiber-reinforced polymers since the silicon ends of the molecules are similar to glass, and the organic groups on silicon could be synthesized for compatibility with organic polymers [2]. When silane coupling agents are introduced onto the glass fiber surface in the composites, two interfaces exist between the glass fiber and the polymer matrix: the interface between the glass fiber and the silane coupling agents, and the interface between the silane coupling agents and the polymer matrix. There is a need to characterize the interfacial properties between the fibers and the matrix of the composites, as the degree of adhesion at the interfaces between two dissimilar solids is largely dominated by weakly intermolecular or physical forces [3–5]. **A** number of experimental techniques have been devised to measure the mechanical properties of the fiber-matrix interfaces in composites. The single fiber compression test and microindentation test were the earliest test methods developed based on microcomposites to measure the bond strength of glass fibers [6]. In regarding of these methods, in this study the compressive strength and macrointendation were performed as measure of the adhesion at interface fiber matrix.

The silane coupling agents used in this study, 3 -methacryloxypropyltrimethoxysilane (MEMO), 3 -aminopropyltriethoxysilane (AMEO), and 3-glycidoxypropyltrimethoxysilane (GLYMO), were supplied by Dynasilan, Germany. The chemical structures of the various silane coupling agents are given in Fig. 1.

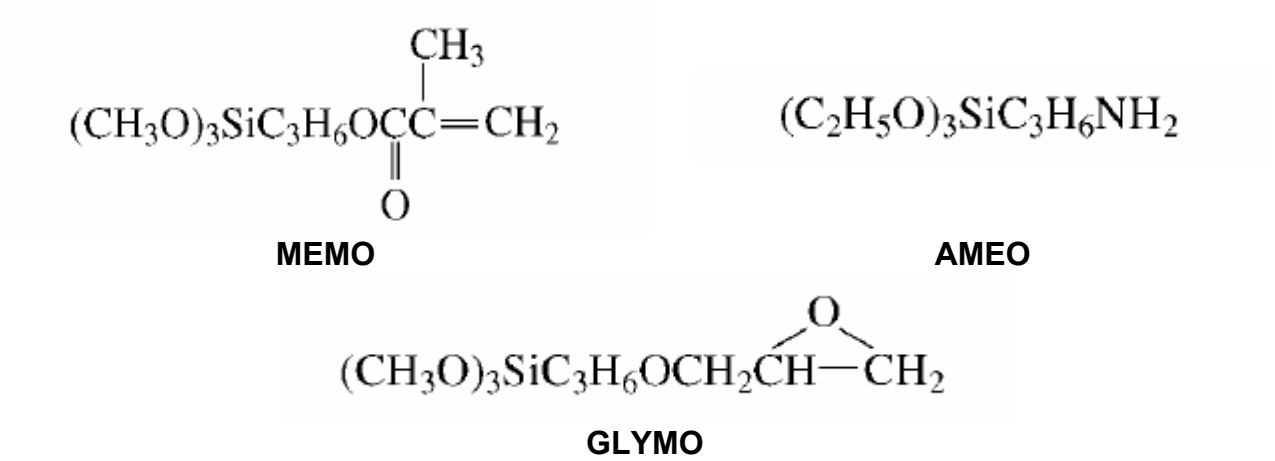


Fig. 1. Chemical structures of the silane coupling agents used

Experimental

The started material was a wasted glass mat and in-plant scarp impregnated by urea-formaldechide. It was cured by annealing at 550°C in a conventional oven. After that the glass fibers was chopped, separated by set of sieves in respect of fibers dimensions. The aspect ratio L/D for the separated mass fraction of started material was obtained by image analyzing of glass fibers microphotographs. The mass fraction of with the largest amount of glass fibers (aspect ratio L/D = 16.71) was used for the composite processing. The surface modification was achieved by treatment of glass fibers with AMEO, GLYMO or MEMO silane. The granulated HDPE TR-130 (density 938 kg/m^3, Vicat temp. 120 ºC) supplied by Petrohemija Pancevo, Serbia was used as thermoplastic matrix of composite. Four series of samples were processed by compression molding, (pressure 41.38 MPa, temperature 160-170ºC, time 26 min):

I- HDPE-as received after heat treatment glass fibers (10; 20; 30 and 40 vol. %;)
II- HDPE-glass fibers treated by MEMO silane (10; 20; 30 and 40 vol. %)
III- HDPE-glass fibers treated by AMEO silane (10; 20; 30 and 40 vol. %)
IV- HDPE-glass fibers treated by GLYMO silane (10; 20; 30 and 40 vol. %)

Surface treatment of Glass Fibers. A solution of 0.5% silane in de-ionised water was prepared by slowly dripping silane into water during continuous stirring. The solution was stirred for another 20 minutes, allowing hydrolysis and silanol formation, before glass fibers into the solution. After 15 min in the solution, with continuous stirring, the glass fibers were rinsed in de-ionised water and dried at 125º C for 45 min to cure silane layer.

Fourier Transform Infrared Spectroscopy (FTIR). FTIR spectroscopy was used to observe the silane coupling agents adsorbed onto the glass fiber surface after hydrolysis. The instrument scanned in the range 4000-400 cm^{-1}using a BOMEM spectrophotometer (Hartman & Braun MB-series, Baptiste, Canada) by 4 cm^{-1} resolution..

Mechanical testing .Compressive properties of composite were measured on universal testing machine R-10 Mashriborintorg Moscow. The compression tests of samples were performed in direction normal to processing compression direction. Compressive strength was calculated according:

$$\sigma = \frac{2F}{\pi d^2 h^2} \tag{1}$$

where F is the maximum load, d is sample diameter and h is sample thickness.

Macroindentation tests were performed on Briviskop WPM VEB Werkstoffprüfmasinen-Leipzeg. The modified Brinell hardness test with 5 mm diameter still ball indenter and max. 1840 N load was performed. The hardness was calculated as:

$$HB = \frac{2 \cdot F}{\pi \cdot D \cdot \left(D - \sqrt{D^2 - d^2} \right)} \tag{2}$$

where F is the load, D is the ball diameter, and d is diameter. The d was obtained by image analyzing of indentation microphotograph (Fig. 2).

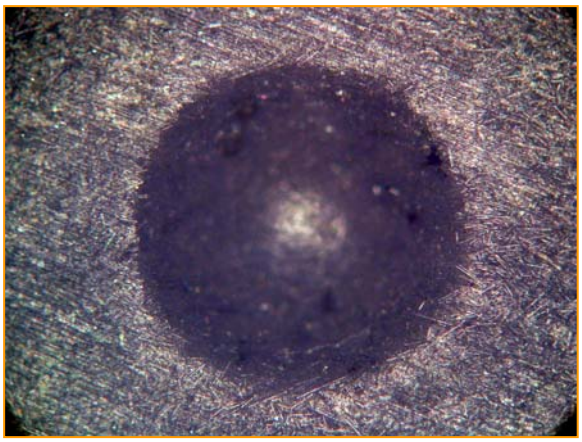
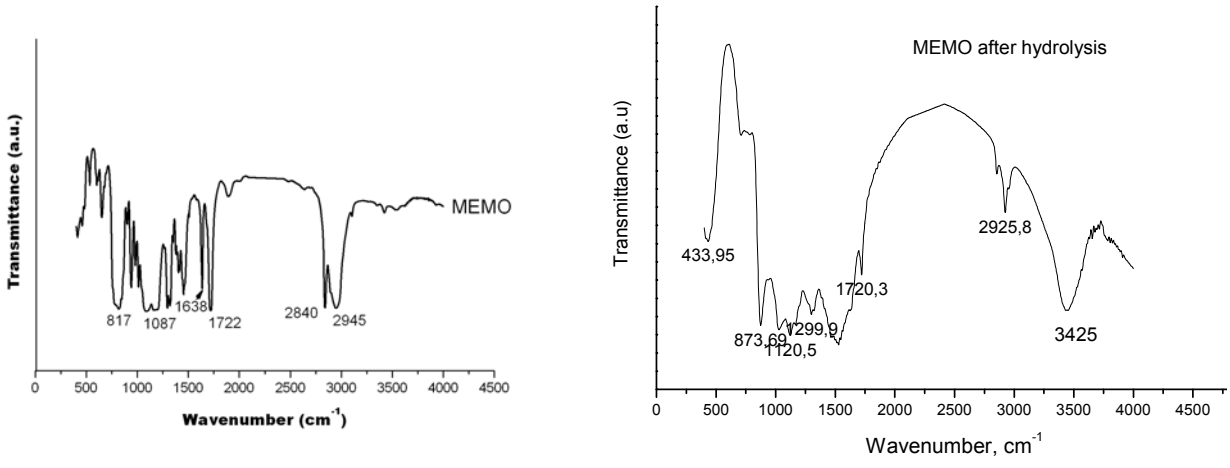


Fig. 2. Indentation microphotograph

Results and discussion

FT-IR spectroscopy was used to observe the silane coupling agents adsorbed onto the glass fiber surface after hydrolysis. The FT-IR transmittance spectra of MEMO-silane before and after hydrolysis are shown Fig. 3. The spectrum of Fig. 3a, before hydrolysis, shows two strong peaks at 2947 and 819 cm^{-1} due to the Si- OCH_3 group of MEMO-silane. Two peaks at 3425 and 1022 cm^{-1} due to Si-OH group are observed in all spectrums due to hydrolysis (Fig. 3b). The peaks corresponding to carbonyl group (1720 cm^{-1}) of MEMO-silane appeared on the spectra of the fiber glass surface.



a) b)

Fig. 3. Transmission spectra of MEMO-silane (a) before hydrolysis and (b) after hydrolysis.

The results of compressive strength and macroindentation tests are presented on the figures 4. and 5. respectively. The influence of fiber content is presented too.

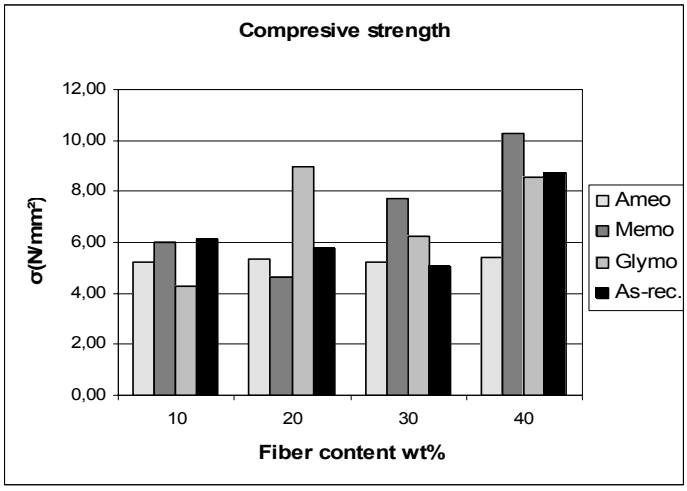


Fig. 4. The dependence of compressive strength on fiber content and surface treatment

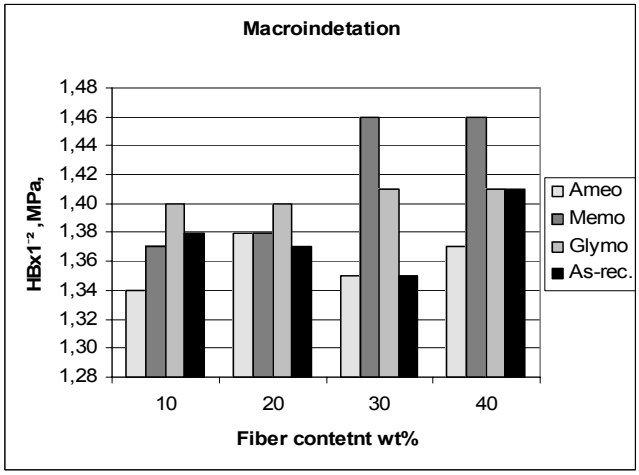


Fig. 5. The dependence of hardness on fiber content and surface treatment

The reinforcement effect of is obvious only with higher content of glass fibers (30% and 40%). The best improvement of interfacial strength with MEMO silane was achieved in samples with higher content of glass fibers (30% and 40%). From the Fig. 4. and 5. it is apparent that the compression strength and hardness shows the similar behavior and tendency with fiber content and the class of silane. So, the hardness could be introducing as mechanical property for characterization the interfacial properties between the fibers and the matrix of the composites.

Summary

The aim of this paper was to investigate the influence of the silane as adhesion promoters for increased composite interfacial strength before compounding, and detect the appropriate mechanical testing for characterization composite interfacial strength. Additionally, the process of recovering and reuse of wasted glass fibers was investigated and developed too.

The samples of composite short glass fiber-HDPE were processed with different content of glass fibers and with surface modification by different class of silane. The compressive strength and macrointendation were performed as measure of the adhesion at interface fiber matrix. The best improvement of interfacial strength with MEMO silane was achieved in samples with higher content of glass fibers (30% and 40%). From this preliminary investigation the hardness could be introduce as mechanical property for characterization the interfacial properties between the fibers and the matrix of the composites.

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