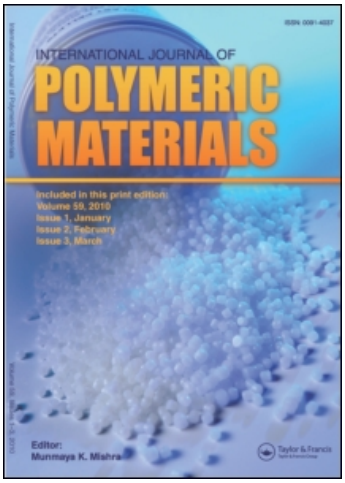

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Utilization of Urea- and Melamine-Formaldehyde Resin Wastes as Reinforcing Materials

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Industrial wastes of urea-formaldehyde (UF) and melamine-formaldehyde (MF) resins have been collected from industrial sources in Jeddah. Such wastes were classified, with the exclusion of unsuitable fractions, according to color or shape. After that, classified wastes were ground into relatively fine powder and the coarse granular parts were excluded with the aid of special sieves. The powdered wastes were mixed in different compositions with unsaturated polyester (UP) in the presence of cobalt octanoate activator and methylethylketone peroxide initiator for crosslinking. Different mixtures were prepared in a cylindrical form and characterized through their ability to absorb water and their compression strength. Results have been discussed according to the variation of the chemical structure of the waste resins used. They reflect the ability of thermosetting polymeric wastes, especially those of UF and MF resins, to be utilized as fillers and reinforcing materials in UP end-products.

Keywords: formaldehyde, melamine, reinforcement, unsaturated polyester, urea

INTRODUCTION

An integrated approach is required in an attempt to manage such large quantities of a diverse, contaminated mixture of plastics in an energy-efficient and environmentally sensitive manner. This would require critically examining various steps in the life of plastics such

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as the raw materials for their manufacture, the manufacturing processes, design and fabrication of the finished products, possible reuse of those items, and the proper disposal of the wastes. Such an integrated waste management concept comprises: (1) source reduction, (2) reuse, (3) recycling, (4) landfill, and (5) waste-to energy conversion [1,2].

The major operations involved in a waste management process include the collection of the plastic waste outside or inside the municipal waste stream, its disposal in landfills, its energy recovery, recycling into useful products, and the establishment of markets for the recycled products [3]. There are different options for the current waste management of thermosetting waste to find out the potential to make new useful construction products through reinforcement. These are increasingly being used due to their light weight, ease of installation, low maintenance, tailor-made properties, and corrosion resistance [4].

Urea-formaldehyde (UF) and melamine-formaldehyde (MF) resins are among the main categories of thermosetting polymers being used in a wide range of household articles. During the manufacturing process some articles have defects and don't fit with the quality requirements. Such articles are excluded and then used in a recycling process. It is worth mentioning that although many works have been published regarding the applications of thermosetting polymeric materials in different fields and their recycling approaches [2,4–17], there is only a small amount of literature concerning the recycling of UF and MF resins. The current work aims at using a facile recycling method for UF and MF resins to be recycled into value-added materials.

EXPERIMENTAL

Materials

Waste of urea-formaldehyde and melamine-formaldehyde end-products was obtained from some factories in Jeddah. Unsaturated polyester resin (UP) based mainly on isophthalic acid diester containing 40% styrene was supplied by Saudi Industrial Resins Limited, Jeddah, KSA. Methylethylketone peroxide and cobalt octanoate were purchased from Aldrich.

Methodology

Grinding

The industrial waste of UF and MF resins was ground repeatedly with the aid of a special mill used for rock grinding. After that, the ground product was sieved and coarse granules were excluded.

The average particle diameter based on the sieving process was about 1.25 μm.

Mixing

Based on the weight of the unsaturated polyester (UP), an amount of 3 wt% of cobalt octanoate was added to a suitable amount of UP. The ground wastes of UF and MF resins were each mixed with UP in different compositions as shown in Table 1. The mixture was stirred carefully for homogeneity. After that, methylethylketone peroxide corresponding to 1 wt% of UP was added. The mixture was then efficiently stirred for a short time and transferred into cylindrical molds of 30 mm inner diameter. The mixtures in the molds were left under vacuum for a while before the gel-time is reached to get rid of the entrapped air bubbles, and then left for curing at 40°C for 6 h followed by annealing of the samples at gradually increased temperature from 40 to 90°C over stages of 10°C interval, while keeping the temperature constant in each stage for 1 h. After reaching 90°C, the samples were kept at this temperature for an additional 3 h and left to cool down to room temperature.

Investigation of the Parameters

The following parameters were followed:

- **Reaction temperature during the curing process:** The reaction temperature was recorded during the course of the reaction, until complete curing of the reaction mixture, by using thermocouple digital thermometer.

TABLE 1 Compression Strength (CS) in kg/cm^2 of UP Samples Reinforced with Different Compositions in Weight% of MF or UF Resin Wastes

	Compression strength (CS), kg/cm^2			
	MF		UF	
Wt.% of waste	No.	CS, kg/cm^2	No.	CS, kg/cm^2
0	1	80.6	1	80.6
17	2	91.9	9	112.4
33	3	101.9	10	161.2
50	4	116.7	11	241.1
55	5	123.8	12	270.1
58	6	125.9	13	281.4
62	7	128.0	14	311.1
64	8	127.3	15	305.5

- **Compression strength:** It is the resistance of samples to an applied external pressure. The samples of different compositions of UP and MF or UF waste material were prepared in a special cylindrical mold of 30 mm inner diameter with varying length. Resistance of samples to the applied external pressure was measured by using a Zwick Tensile instrument and consequently the compression strength has been calculated.
- **Water absorption ability:** Samples of the different compositions were cut into cubes of 1 cm edge and immersed in 25 ml of distilled water at room temperature for 48 h. After that, the samples were wiped with filter tissue and weighed. This experiment was repeated for similar samples after polishing the surface of the samples. The water absorption % was calculated according to Eq. (1):

$$\text{Absorption}\% = \frac{Wi - Wo}{Wo} \times 100$$

where $Wi =$ weight after immersion and $Wo =$ weight before immersion.

RESULTS AND DISCUSSION

A literature survey reflects that recycling the thermosetting polymers MF and UF as reinforcing materials, especially for the unsaturated polyester (UP), is less frequently cited in comparison to other reinforcing materials. This difficulty arises from the inability of such materials to be melted or solubilized in organic solvents as a result of their crosslinking into three-dimensional networks during processing. Therefore, recycled MF and UF have been selected to be investigated as reinforcing materials for UP products. The choice was based on the fact that both materials MF and UF on one side and UP on the other, have functional groups that may be compatible or at least have some sort of compatibility with each other. The result obtained is the mean of three replicates.

Reaction Temperature during the Curing Process

The temperature of curing was noted to rise during the course of the reaction as a result of the exothermic nature of the crosslinking reaction. The curing temperature reaches a maximum and after that it tends to step down until it reaches room temperature after a certain period of time, depending on the mixture composition and the total mass of the sample.

Compression Strength of the Reinforced Material

Table 1 showed the compression strength (CS) in kg/cm^2 of the UP samples reinforced with MF and UF resin wastes in different compositions. In both cases, the compression strength increases with the amount of the reinforcing material. This dependence has been represented in Figure 1 for MF-reinforced UP and in Figure 2 for UF-reinforced UP. It is found from Figure 1 that samples of 17 wt% of MF showed compression strength of ~92 kg/cm^2 that increased to 128 kg/cm^2 for samples of 62 wt% of MF resin waste. Beyond this composition, samples' compression strength tends to decrease. Such dependence has almost linear behavior, leading to the ability to predict the compression strength of samples as a function of MF content. In addition, the linear relationship indicates that MF particles are homogeneously distributed within the UP matrix with good mutual attraction and adhesion. The dependence tends to form a plateau and starts to step down at higher contents of MF. This behavior can be attributed to the poor wetting of MF particles with the UP liquid. Therefore, there were still some MF domains un-wetted with UP which represents points of weakness in the end product after curing.

Figure 2 shows the compression strength (CS) of UF-reinforced UP samples. From the results, we may remark that the UF-reinforced UP samples showed compression strength higher than that obtained with

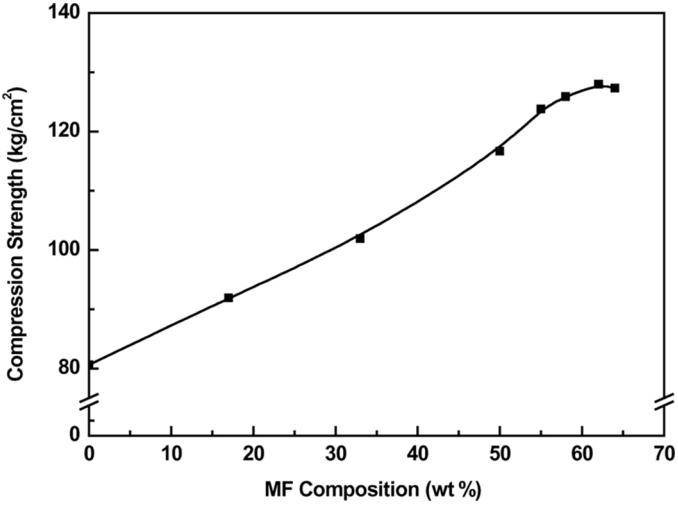

FIGURE 1 Dependence of compression strength (CS) in kg/cm^2 on the MF composition in wt% in the MF-reinforced UP samples.

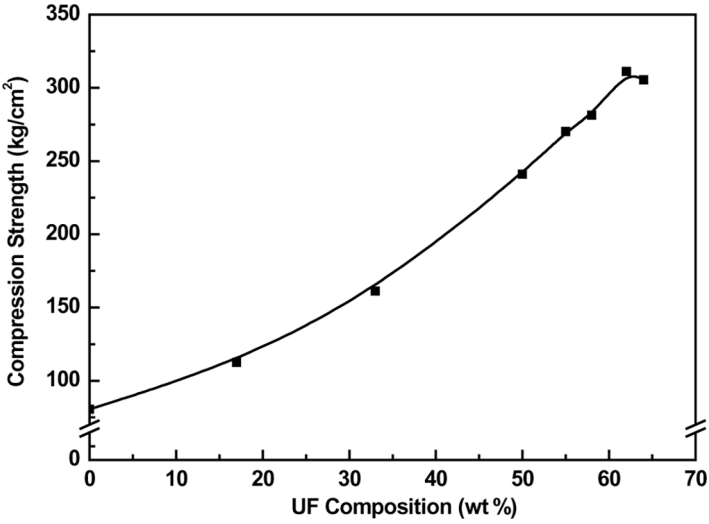

FIGURE 2 Dependence of compression strength (CS) in kg/cm^2 on the UF composition in wt% in the UF-reinforced UP samples.

MF-reinforced ones. For example, samples 17 wt% of UF showed compression strength up to 112.5 kg/cm^2 while samples of similar composition of MF showed compression strength of 92 kg/cm^2 only. Also, samples of 62 wt% of MF showed compression strength of 128 kg/cm^2 while it reaches to 311 kg/cm^2 for UF-reinforced UP samples of the same contents. This may be attributed to the higher activity of the amine groups in UF than that of MF, in addition to lesser steric hindrance of UF residues than in the MF residues. After that, the dependence tends to form plateaus followed by step-down behavior. This can be attributed to the wetting problem explained above for the MF-reinforced UP samples.

The obtained results are encouraging and promising in the light of the possibility to exclude the addition of the cobalt activator and use only the peroxide initiator. The effect of the activator can be compensated by applying high temperature and pressure during the processing. This may increase the interaction between UP and UF and/or MF as reinforcing materials in addition to facilitating the production of various articles of complicated structures.

Water Absorption Ability of the Reinforced UP

From the preliminary study one can expect good characteristics for the obtained samples. The ability of samples to absorb water was proved

to be negligible. Hence, the increase of the weight of samples as a function of water absorption was found to be in the range of 0.4 to 1.03 wt% after immersion for 48 h in distilled water at room temperature (~25°C). It was also noticed that UF-reinforced samples were able to absorb more water than the MF-reinforced ones. Also, the polished samples showed lower ability to absorb water than the unpolished ones, due to the surface roughness of the unpolished samples. This can be attributed to the ease of water absorption by the unpolished surface as a result of higher surface area, which is composed mainly of UF waste particles of a hydrophilic nature.

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