INFLUENCE OF SONICATION TIME ON THE PROPERTIES OF EPOXY RESIN

Roberta Motta Neves[1*], Lídia Kunz Lazzari[1], Ademir José Zattera[2], Sandro Campos Amico[1]

1 – Postgraduate Program in Mining, Metallurgical and Materials Engineering (PPGE3M), Universidade Federal do Rio Grande do Sul (UFRGS), Av. Bento Gonçalves, 9500, Porto Alegre/RS
robertamneves@gmail.com

2 – Postgraduate Program in Engineering of Processes and Technologies (PGEPROTEC), Universidade de Caxias do Sul (UCS), R. Francisco Getúlio Vargas, 1130, Caxias do Sul/RS)

Abstract
Epoxy resins are widely reinforced by micro or nanofillers, producing nanocomposites. To obtain desired properties of the final composites, the filler must be well through the matrix, and the sonication technique is applied for this purpose. However, this process has some undesired effects on the polymers such as decreasing properties. Due to it, a study regarding sonication time on the epoxy resin is highly recommended. This study aims to verify the influence of sonication time on neat epoxy resin. The epoxy resin was sonicated at different times: 0, 5, 10, 15, 30, and 60 minutes. The samples were characterized by viscosity, dynamic-mechanical and impact, and tensile strength. The results showed that the influence of sonication time was more pronounced in the viscosity values and in the dynamic properties than in the mechanical properties. Considering the results reported here, lower sonication times seem to be the most adequate conditions for epoxy resin, without fillers.

Keywords: epoxy resin, sonication time, viscosity, mechanical properties.

Introduction

Epoxy resins are widely used as a matrix for composites due to their excellent mechanical properties [1]. Epoxy-composites could be reinforced by several types of micro or nanofillers such as cellulose, clays, graphene, carbon nanotubes, aramid pulp… [2]. The final properties of this kind of composites are related to the dispersion of the filler through the matrix. To achieve improvement on the properties, well-dispersed particles need to be achieved [3]. The sonication method has been widely used to disperse nano or micro fillers on the epoxy resin.

The sonication process is based on acoustic cavitation. During the process occurring formation of bubbles, which growth and collapse in a liquid, such as epoxy resin. Then a rarefied field generate and the bubbles will continuously increase in diameter and initiate another cavitation process, dispersing the filler through the polymeric matrix [3,4]. The process has outstanding power in order to disperse particles, however, this process has some undesired effects on the polymers.

During sonication, the liquid is heated up and, due to it, external cooling such as an ice bath is recommended to keep the epoxy resin below the temperature of degradation. Additionally, the cavitation is able to change the morphology of the polymer, varying in the number of monomers and oligomers, and, sometimes, possible recombination of radicals happens. These effects can change the cure reaction of the epoxy, decrease the viscosity, and consequently, decrease the chain length of epoxy [3,5].

Polymers, such as epoxy resin, have a strong relationship with properties versus chain length. So choosing the process parameters that always operate without break the polymeric chain is highly recommended [6]. As aforementioned, a composite is produced always looking forward enhance properties and, the sonication process may influence the final properties of the composites. Thus, a study regarding sonication time, before the incorporation of fillers in the epoxy resin is necessary.

Apart from it, this study aims to determine the influence of sonication time (0, 5, 10, 15, 30 and, 60 minutes) on the viscosity, dynamic-mechanical and mechanical properties (impact and tensile strength) of epoxy resin.

Experimental

Materials

Bisphenol A diglycidyl ether (DGEBA) epoxy resin and HT1564/E150 hardener were obtained from Advanced Vacuum (São Paulo, Brazil) and acetone was purchased from Dinâmica Química (São Paulo, Brazil). All reactants were used as received.

Methods

Epoxy resin (150 ml) and acetone (150 ml) mixture were sonicated in a SONICS Vibracell VCX-505 equipment, 40% using amplitude and 500 W. A thermometer was coupled to the system to track the temperature during the process. Different times of sonication were applied: 0, 5, 10, 15, 30 and 60 min. Then, the sonicated epoxy/acetone mixture was magnetically stirred at 70°C for 3 h and, later, placed in an oven at 80 °C for 12 h for acetone removal. After cooling to room temperature, the hardener was added to the epoxy (15:100 hardener:resin w/w) and manually mixed. A degassing step was add in order to eliminate bubbles in vacuum oven at −0.8 bar for 3 cycles of 10 min. The specimens were molded by pouring the mixture into silicone rubber molds, cured in situ for 48h and post-cured in the following cycle according to the manufacturer: 2h at 70 °C, 2h at 90 °C, 2h at 120 °C and finally 15h at 150 °C. The final samples were named using the sonication time used.

Characterizations

The samples were characterized by viscosity (prior hardener addition) and by mechanical properties (post-cured resin). Viscosity was measured in a Brookfield DV-IIþPRO equipment at 25°C, 100 r/min, Spindle S64. Tensile testing was performed according to ASTM D638 with Type I test specimens in an EMIC DL-3000, 200 kgf load cell, at a speed of 1 mm min^{-1}. Unnotched IZOD impact testing was done in a CEAST Resil-25 equipment with 4J hammer energy, according to ASTM D256. Five to seven specimens were tested in each case. Dynamic-mechanical analysis (DMA) was performed in a TA instruments Q800 AT equipment, with single cantilever clamp, temperature range 35–200 °C, heating rate of 3°C/min, frequency of 1 Hz and deformation amplitude of 0.1%. Fig. 1 summarized the samples manufacturing flowchart.

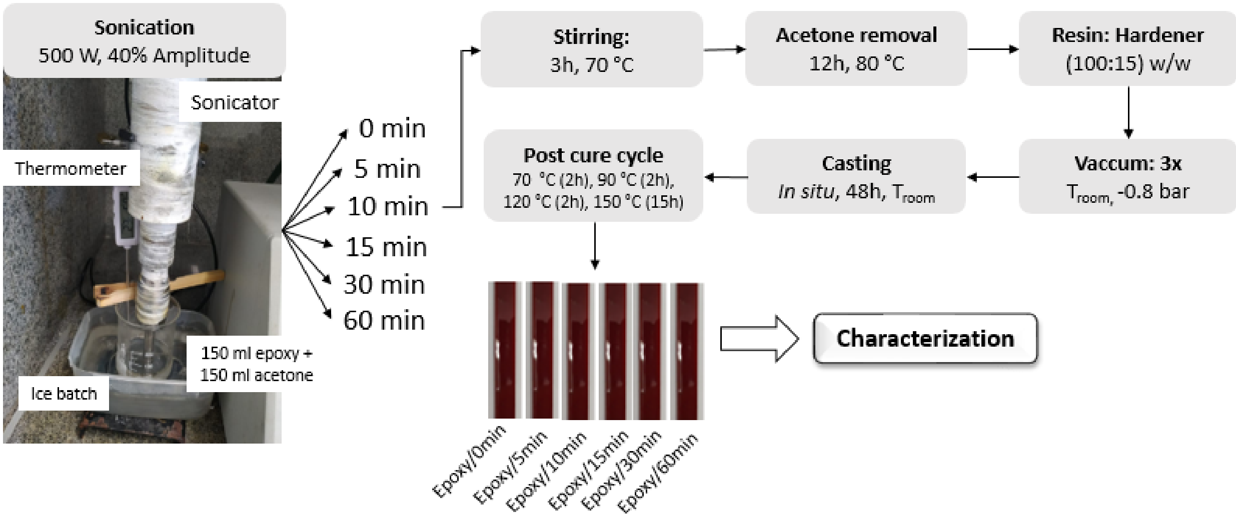

Figure 1– Experimental flowchart

Results and Discussion

Table 1 shows the effect of sonication time on resin viscosity and the final temperature reached during the sonication. The resin without sonication time (0min) and the resin sonicated by 60 minutes presented the highest and lowest viscosity, respectively, as expected. Additionally, higher sonication times reached higher temperatures. The samples sonicated for 5, 10, 15, 30 and 60 minutes showed a viscosity value reduction around 37%, 43%, 50%, 56% and 61% respectively (compared with 0 minutes sample). Suslick [6] attribute this reduction in the resin viscosity to a possible cleavage of the polymeric chain bonds due to cavitation. Additionally, when polymeric chains undergo sonication energy, a thickening of these chains happens, increasing the free volume between them, decreasing the viscosity [3,7].

Table 1 – Viscosity values of the different sonication times applied and the final reached temperature during the sonication

Sample	Viscosity (mPa/s)	Final Temperature (°C)
Epoxy/0 min	1585 ± 3	25*
Epoxy/5 min	957 ± 7	35
Epoxy/10 min	887 ± 9	56
Epoxy/15 min	767 ± 6	68
Epoxy/30 min	709 ± 3	74
Epoxy/60 min	631 ± 8	83

* Room temperature

The effects of sonication time on the mechanical properties of epoxy resin are shown in Fig. 2. Regarding impact properties the values remain almost unaltered, demonstrating that the sonication time did not affect these properties. On the other hand, the samples sonicated for 5, 10, 15 and 30 minutes showed higher values for tensile strength than the non-sonicated sample (epoxy/0 minutes). A clear trend was observed, once increasing sonication time, the tensile strength shows an ascending trend, reaching a peak and then falls. Similar behavior was reported by Montazeri et al [8]

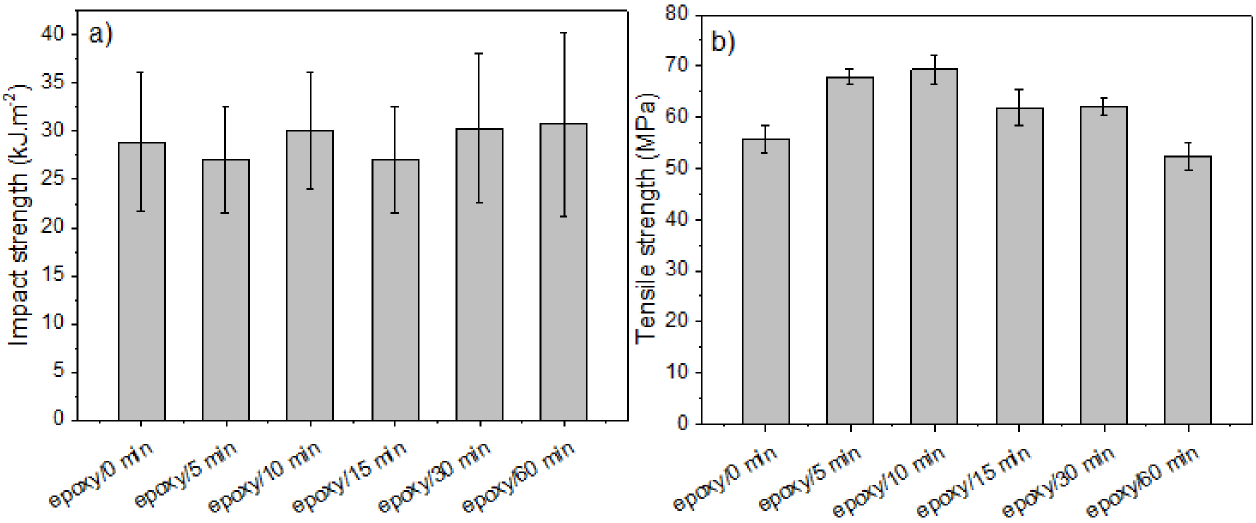

Figure 2– a) Impact and b) Tensile strength for different sonication times applied on the epoxy resin

Figure 3 displays all the dynamic-mechanical curves for different sonication times applied on the epoxy.

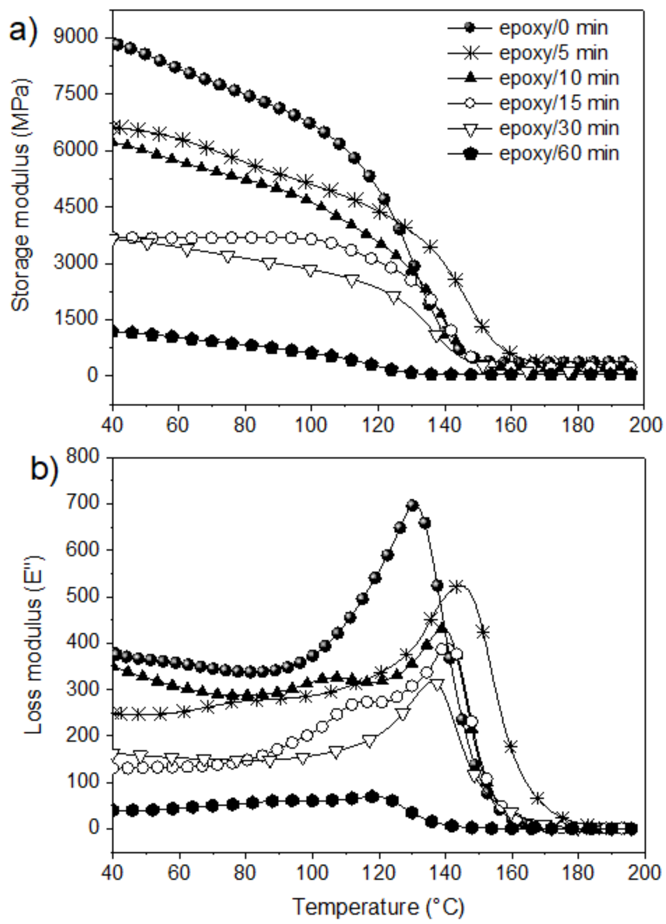

Figure 3– Dynamic-mechanical curves for different sonication times applied on the epoxy resin a) Storage modulus (E') b) Loss modulus (E'')

The storage modulus (E’) (Fig. 3a) shows a clear trend among the samples. Increasing sonication time a decrease in the E’ happened. This behavior has the same trend in both states: glassy (35-100 °C) and rubbery (above 150 °C). Comparing with the non-sonicated sample, a decrease in the modulus around 20% and 200% was observed for epoxy/30 min and epoxy/60 min in the glassy state. In the rubbery state, a decrease of around 70% and 480% was verified. Regarding the loss modulus (E”) curves, it was possible to observe that higher sonication times lead to low peak intensity, suggesting that the epoxy resin lost a part of its energy-releasing property. On the other hand, the intermediate sonication times (5, 10, 15, and 30 minutes) lead to a dislocation of the E " peak to higher temperatures, indicating an increase in the Tg of these samples compared to the non-sonicated resin [9].

Conclusions

Epoxy resin sonicated using different times were studies. The influence of time was more pronounced in the viscosity values and E’ curves than the mechanical properties. Higher time of sonication on the epoxy resin led to lower viscosity and higher temperature during the process. Regarding impact strength, the values remain almost unaltered. On the other hand, tensile strength showed higher values for epoxy/5 min and epoxy/10 min. The dynamic-mechanical complied data suggests that the sonication process might lead the epoxy resin to a lower capacity to store and release energy. Considering the results reported here, lower sonication times seem to be the most adequate conditions for epoxy resin, without fillers.

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