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#### Review

## Elaboration and characterization of composite material based on epoxy resin and clay fillers

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#### Abstract

The present work aims at investigating the effect of locally produced clay (Algeria), along with the effect of their size and rate on physical and mechanical properties of the composite material. This study is divided into two parts: The first one is devoted to the study of the composite material based on epoxy resin with kaolin, using different size fractions at rates ranging from 2% to 20%. The second part examines epoxy resin-based composite with calcined kaolin (metakaolin) with regard to the influence of the structure, the particle size and the charge rate on the properties of the material. It is shown that the clay fillers give the epoxy resin different properties compared to the epoxy resin alone and, additionally, reduce the cost of materials. It was also observed that the fillers enhance the mechanical properties by increasing the rigidity of the material. There is a maximum value of 2.4 GPa to 18% kaolin, or more than 325% increase in the modulus of elasticity with respect to unfilled resin for the finer particle size. It was also found that the modulus of elasticity increases with increasing the loading rate. Indeed, the rigidity increases with increasing the filler rate. Moreover, for both fillers, lower fraction yields better results.

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Keywords: Epoxy resin; Clay fillers; Physical properties; Mechanical properties

#### 1. Introduction

Performance optimization of materials is very important in providing a competitive advantage, which basically means improving properties while reducing cost (Bardonnet, 1992; Cho, Joshi, & Sun, 2006). Many modern technologies make use of new materials with properties that out perform traditional ones (metals, ceramics and polymers). The search for better alternatives to those materials has led to continuous improvement to ward either less expensive or more efficient materials. Yet, combining both remains the most desirable outcome.

A composite is a multiphase material that combines the properties of its constituents to acquire better properties than its parent components (Berthelot, 1996; Glossaire des matériaux, 2006). It consists of a reinforcement which contributes to the mechanical properties and a binder called matrix. The matrix

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ensures the cohesion between the elements of the reinforcement, transfers the strain and makes the material resistant to environmental elements (corrosion, aging) and temperature (Berthelot, 1996).

The properties of a composite material depend on the nature (Kornmann, Lindberg, & Berglund, 2001), the shape of the reinforcement, the input amounts, the quality of the matrix/fillers interface and finally, on the synthesis process. The integration of reinforcement into matrix to obtain new specific functions is a widely investigated topic (Antoon, Koenig, & Serafini, 1981; Bondioli, Cannillo, Fabbri, & Messori, 2005; Gerdinand, Budde, & Kurrat, 2004; Janssen, Seifert, & Karner, 1999).

Epoxy resins are flexible polymeric materials characterized by the presence of two epoxide groups or more in their molecular structure (Freddy, 2000).

Epoxy resins are thermosetting polymers, widely used in high performance composites in many industrial applications, such as high performance adhesives in airplanes, pipe coatings in the oil industry, printed circuit boards, and many other applications in construction, automotive and aerospace industries (Fu, Feng, Lauke, & Mai, 2008; Solihin, Tongamp, & Saito, 2011).

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Their insulating electrical properties and their relatively high thermal resistance (Du & Yang, 2012; Galán & Martín Vivaldi, 1972) are the main reasons for using those materials in such applications. However, from a mechanical perspective, epoxy polymers show a brittle fracture mode.

Therefore, they usually fail by brittle fracture mechanisms under normal conditions of use. To address this issue and increase the properties of the brittle polymer, several approaches are possible. This can be achieved by firstly adding rubber particles (Castrillo, Olmos, Torkelson, & González-Benito, 2010; Du & Yang, 2012; Galán & Martín Vivaldi, 1972; Lee & Yee, 2001; Sue & Yee, 1993) and then a thermoplastic polymer in the epoxy matrix (Boo, Liu, & Sue, 2006; Boo et al., 2007). Yet another alternative is to modify epoxy resins by adding particles or inorganic fillers (silica, kaolin, montmorillonite) (Castrillo, Olmos, Sue, & González-Benito, 2015), which generally lead to lower costs without deteriorating the properties of the material, and sometimes it can also bring further improvements in the final material, such as barrier properties, flammability resistance, thermal stability and solvent absorbance (Fellahi, Chikhi, & Bakar, 2001; Garg & Mai, 1988).

The integration of inorganic fillers in polymer matrices is a subject extensively investigated (Irekti, 2011; Wang, Tian, Wang, & Han, 2005; Zapata-Massot, 2004).

The inorganic fillers are used as reinforcement in epoxy resin systems. They significantly increase the mechanical, electrical and thermal properties. These inorganic fillers are chosen for their availability. They are highly sought after as powders in several scientific fields as a way improving some of the features mentioned above and also to reduce the cost of the desired materials. The final properties of the composite depend on the intrinsic characteristics of each component, quantity, shape and size of loads and the nature of the interfaces many studies have been devoted to the study of loaded composite materials and several types of inorganic fillers were used in this work to improve certain characteristics of the matrix.

The role of charges is multiple, it can more cost effectively fill a volume if the load is much cheaper than the polymer, and it can modify the macroscopic properties as well.

The main objective of this work is to develop composite materials based on epoxy resin and inorganic fillers, in this case, treated (physical treatment) Tamazert kaolin (Algeria) and untreated kaolin (metakaolin) at different rates and with different particle sizes. The purpose is hence to highlight the influence of the structure, the rate and size of the fillers on the specific properties of the material (mechanical strength, thermal stability, dimensional stability).

#### 2. Experimental procedure

#### 2.1. Materials used characterization

#### 2.1.1. Matrices characterization

The matrix consists of two elements, element A the monomer is the resin injected medapoxy 812 A, and element B called hardener is the hardener of the resin injected medapoxy 812 (aromatic amine) gives the mixture a cross linked resin.

In the present work, we used MEDAPOXY 812INJ from Granitex (ALGERIA), which is an efficient product with numerous qualities utilized in many advanced technology applications. The mass ratio (MR) between the monomer and the hardener provided by Granitex is MR = 2.

The resin characteristics MEDAPOXY 812INJ according to the supplier's data sheet are shown in Table 1.

#### 2.1.2. Clay fillers characterization

The untreated kaolin and kaolin physically treated (metakaolin) were used as fillers in the present study. The powders were mechanically milled through a ball mill process. We obtained three ratios of different granulometry for both fillers.

The kaolin

Pre-treated Tamazert kaolin was used (deposit of Tamazert near MILA Algeria, 200 km east of Constantine).

Metakaolin

The metakaolin was obtained by calcination of kaolin at high temperatures (500–550 °C) for 5 h. Calcination causes dehydroxylation and destruction of the initial crystalline structure of the kaolinite (Brindley & Nakahira, 1959; Brindley, Sharp, Patterson, & Narahari, 1967; Horvath & Kranz, 1980).

The obtained fillers were characterized using granulometry, X-ray diffraction (XRD), with X-ray fluorescence.

2.1.2.1. Granulometriy. The particle size is crucial in mechanical and esthetic properties of composite resins and a key parameter in different classifications; the most commonly adopted classifications are indeed based on the shape and size of particles. The granulometry of kaolin and metakaolin was done by means of a Mastersizer 2000 laser particle type of URMPE research unit of Boumerdes Algeria.

The results are shown in Figure 1 (K1). From these results we can see that the particle size distribution K1 (kaolin particles have the biggest size) varies between  $0.1 \, \mu m$  and  $300 \, \mu m$ .

The kaolin sample (K1) shows a multimodal distribution made up of 4 medium size fractions, (1) is <1  $\mu m$  with  $\sim\!20\%$  of volume, (2) is 4  $\mu m$  with >40% of volume, (3) is 30  $\mu m$  with 20% of volume and (4) is 200  $\mu m$  with >15% of the volume. The whole volume is filled with particles <300  $\mu m$ .

This particle size curve shows that the particle size distribution of the load K2 (kaolin particles have an average particle size) is between 0.1  $\mu$ m and 100  $\mu$ m, the results are presented in Figure 1 (K2).

The kaolin sample K2 has a multi-modal distribution which is composed of 3fractions particle size, (1) a fraction of average size less than 1  $\mu$ m with about 25%, (2) a fraction of average size of 4  $\mu$ m with more than 40% by volume, and (3) a fraction of average size of 30  $\mu$ m with more than 20% by volume. The total volume is occupied by particles <100  $\mu$ m.

The results are shown in Figure 1 (K3 (kaolin particles have a fine particle size)). From these results we can see that the particle size distribution K3 varies between  $0.1~\mu m$  and  $50~\mu m$ .

The kaolin sample K3 has a bimodal distribution consisting of two size fractions, (1) a fraction of average size less than 1  $\mu$ m

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