



Mechanical properties of epoxy/boehmite nanocomposites in dependency of mass fraction and surface modification - An experimental and numerical approach

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ABSTRACT

Boehmite nanoparticles show great potential in improving the matrix dominated mechanical properties of fiber reinforced polymers. For the material design process and the prediction of the nanocomposite's properties, knowledge about the material behavior of the constituent phases and their interactions is crucial. Since the chemical composition of the particle surface can strongly affect the interaction between particle and matrix, the influence of particle surface modification and mass fraction on mechanical properties (tensile modulus, tensile strength, fracture toughness) and failure mechanisms of nanoparticle reinforced epoxy resins is investigated using experimental and numerical methods. In the present work, unmodified and thus chemically reactive boehmite particles are compared to acetic acid modified particles with supposedly lower chemical reactivity and thus worse particle-matrix bonding. A linear relationship between particle mass fraction and tensile modulus/fracture toughness is experimentally found with a maximum increase of 26% in tensile modulus and 62% in critical energy release rate for a particle content of 15 wt%. Furthermore, the experiments indicate that the acetic acid surface modification increases the tensile modulus (up to 6% compared to the unmodified boehmite particles), but at the same time not significantly affects the tensile strength or the fracture toughness. Molecular Dynamic Finite Element Method (MDFEM) simulations are conducted to identify and understand the mechanisms induced by nanoparticles. The material properties of both, modified and unmodified, nanoparticle systems are discussed in relation to the change of particle-matrix bonding strength and interphase morphology. While simulation results of the unmodified system show an outstanding agreement with the experiments, the acetic acid modified system deviates significantly. In conclusion, it seems that additional effects have to be considered to completely understand the material behavior.

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1. Introduction

Fiber reinforced composites are regarded as one of the most important future lightweight materials. Major weaknesses are their matrix-dominated properties. These properties can be improved by adding filler materials. Various studies [1–4] show that in particular nanoscaled fillers, such as nanoparticles and nanotubes, possess desirable properties as matrix additives in composite materials. The reinforcement effect depends on their shape, size, content, and particle-polymer interaction. The work of Kango et al. [5] reviews in detail the production of nanocomposites and their

mechanical properties with a specific focus on surface modified, inorganic particles.

The incorporation of nanoparticles in polymers can generally improve selected matrix dominated properties of fiber reinforced composites, like shear strength, shear modulus, compressive strength, fracture toughness or impact resistance. For example, Arlt [6] reports that a boehmite particle content of 15 wt% leads to an increase in these properties up to 25% for a carbon fiber/Bisphenol-A-diglycidylether composite compared to the reference composite without particle reinforcement. Other studies confirm these results. Uddin & Sun [7] report a significant increase in compressive strength and transverse tensile strength for silica particle reinforced glass/epoxy composites. Subramaniyan & Sun [8] obtain similar results for nanoclay/glass/epoxy composites.

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In contrast, other studies report opposite results. Shahid et al. [9], alike Arlt [6], investigate boehmite/carbon fiber/epoxy composites with different particle surface modifications, i.e. lysine and para-hydroxybenzoate. For small particle weight fractions, tensile and flexural properties improve. Fractions above 5 wt% show deteriorate properties compared to the unloaded composite. In addition to this, Siddiqui et al. [10] report an ambivalent behavior of nanoclay/epoxy composites, as flexural modulus and fracture toughness increase, while flexural strength and impact toughness decrease. Apparently, the same particle/matrix system affects the mechanical properties differently. In particular, it appears that the surface modification is a decisive factor for the effect of the particle filler on the mechanical properties. Thus, the research on boehmite nanoparticles modified with acetic acid may provide an opportunity to generate further knowledge concerning the effects of modified nanofillers on the mechanical properties of boehmite/epoxy composites.

The comparison of the results of Arlt [6] and Shahid et al. [9] demonstrates that the particle/matrix bonding and the resulting interphase are of vital importance to the mechanical properties of the composite materials. Following the argumentation of Arlt, nanoparticles and their surface modifications strongly affect the morphology of the surrounding matrix by impeding the cross-linking reaction and thus forming a soft interphase around the particles. It is assumed that hydroxyl or other functional groups at the surface of the particle or at the end of a molecule, acting as surface modification, are able to react with epoxy groups of the matrix during the curing reaction. As a result, the particle-matrix interphase becomes softer due to a higher density of particle-matrix bonds. This hypothesis is substantiated by previous numerical atomic force microscopy simulation studies [11]. Generally, the particles can have different effects on the curing behavior of the matrix. Putz et al. [12] systematically varied the cross-link density in an epoxy/carbon nanotube composite to characterize the effect on the formation of interphases. Based on their results, Putz et al. proposed three different mechanisms. The first mechanism is the restriction of the mobility of the polymer chains due to attractive forces between the particle and the matrix. Secondly, the nanoparticles lead to an altered thermal conductivity that changes the curing behavior. The third mechanism is a disruption of the polymer network near the particle-matrix interface because of steric limitations or phase segregation.

The present study is an attempt to contribute to the understanding of the effect of surface modifications of nanoscaled matrix additives on the mechanical properties of epoxy/boehmite nanocomposites. Therefore, the mechanical properties of epoxy resins, filled with either unmodified or acetic acid modified boehmite particles, are investigated experimentally and numerically. Unmodified boehmite particles are assumed to chemically interact with the polymer matrix. The hydroxyl groups of the unmodified boehmite might lead to an adsorption or covalent bonding to reactive groups of the matrix. These effects lead to a change in cross-link density in the vicinity of the particles. In contrast, the acetic acid modification, covalently bonded to the boehmite particles utilizing the carboxyl group of the acetic acid molecule, is chemically non-reactive on the opposite molecule end. The chemically non-reactive molecule end is assumed to reduce or prevent particle-matrix bonding, resulting in a lower total surface reactivity of the particles.

By surface reactivity, the authors refer to attractive and repulsive forces affecting the particle-matrix interactions. The surface reactivity and wetting properties of the nanoparticle surface change due to different surface modifications, having an influence on the particle-matrix interphase by changing the morphological properties (degree of cross-linking, chemical composition). It is

expected that the macroscopic mechanical properties of the nanocomposites are directly influenced by the surface configuration of the boehmite particles. Regarding the mechanisms stated above, four hypotheses are formulated. The authors refer back to them in Section 5 to discuss their adequacy to explain experimental and numerical results regarding the chemical structure, the mechanical stiffness, the strength at fracture under tensile loading and the fracture toughness.

Hypothesis 1. The acetic acid weakens the particle-matrix bonding and reduces the development of a particle-matrix interphase.

Reasoning: The methyl group of the acetic acid is non-reactive and thus no reaction with the polymer occurs. Depending on the degree of surface loading, the particle-matrix bonding is reduced. This leads to a reduced influence on the epoxy cross-linking and thus to an undisturbed epoxy matrix in the vicinity of the particles (no or only a small interphase region).

Hypothesis 2. Due to the presence of the acetic acid modification, particle-matrix bonding is weakened, resulting in lower overall stiffness.

Reasoning: A weaker particle-matrix bonding reduces the elastic stiffness. However, a less disturbed epoxy network has the opposite effect. Previous simulations [11] indicate that the influence of reduced bonding is dominating the influence of a less disturbed epoxy network in the interphase. Thus, in total a reduction of the Young's modulus is expected due to acetic acid modification.

Hypothesis 3. The acetic acid reduces the tensile strength.

Reasoning: Weaker particle-matrix bonding caused by acetic acid leads to decreased interfacial stress transfer. In this case, the particle essentially acts as a defect. Thus, there are high local stress concentrations around the particles leading to a decrease in maximum tensile strength.

Hypothesis 4. Due to the weakened particle-matrix bonding caused by the acetic acid, the particles have a higher tendency to de-bond during the fracture.

Reasoning: The particle induced toughening mechanisms depend on particle size and stiffness and the morphology of the particle-matrix interphase. Crack deflection, particle fracture and crack pinning occur for strong particle-matrix bonding. Weak particle-matrix bonding leads to de-bonding and subsequently to plastic void growth.

2. Materials

2.1. Epoxy

The material used in this study is an epoxy resin, diglycidyl ether of bisphenol A (DGEBA) (Araldite LY556; Huntsman), cured with an anhydride curing agent, 4-methyl-1,2-cyclohexanedicarboxylic anhydride (Aradur HY917; Huntsman) and accelerated by an amine, 1-methyl-imidazole (DY070; Huntsman). The matrix components are mixed in the proportion of 100:90:1 parts per weight (epoxy:hardener:accelerator).

2.2. Boehmite

Boehmite (γ -AlO(OH)) is an oxide-hydroxide mineral. It consists of double layers of oxygen octahedrons bonded to a central aluminum atom. The out-facing oxygen atoms are connected to the hydroxyl groups of the obverse octahedral layer via hydrogen bonds. Boehmite exhibits an orthorhombic unit cell (see Fig. 1a), as reported by Bokhimi et al. [13] and experimentally confirmed through X-ray diffraction carried out at the Institute for Particle

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