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Influence of particles-matrix interphase on stress distribution in particulate composite with polymer matrix

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Abstract

In this paper fracture behaviour of particulate composite ($CaCO_3$ – PP) is studied. Attention is focused mainly to the influence of interphase between particles and matrix on stress distribution and on micro-crack propagation in composite matrix. The composite was modeled as three-phase continuum and numerically simulated on a microscopic scale using the finite element program ANSYS. Simplified two-dimensional model is used for estimation of hypothetical micro-cracks path prediction. The influence of interphase properties on fracture toughness for particle reinforced polymer composite is discussed.

Keywords: particulate composite, crack propagation, interphase, $CaCO_3$ – PP composite

1. Introduction

Polymeric particulate composites are frequently used in engineering applications. The properties of the particles themselves (size, shape, material properties) can have a significant effect on the global behaviour of the composite. The addition of rigid particles to a polymer matrix usually has an embrittling effect on the composite. Most studies on modification of thermoplastic composites with rigid mineral fillers report a significant decrease in fracture toughness compared with the neat polymer [4]. The presence of particles significantly influences the cure reaction, resulting in the formation of the third phase known as the interphase, which possesses property distinct from those of the matrix and the particles. In the studied case the size of the interphase is between 20 and 50 nanometres. The interphase has a microscopic scale and controls the adhesion between particles and matrix. Thus it essentially contributes to the ability of the matrix to transfer macro-load and plays a deciding role in the evaluation of the driving force of micro-cracks [1].
Rigid particles as toughness must fulfill certain requirements: the particles have to be of small size (less then 5 µm), the aspect ratio should be close to 1 to avoid high stress concentration, the particles must debond prior to the yield strain of matrix in order to change the stress state of the matrix and the particles should be dispersed homogeneously in the matrix [3].

The main goal of the present paper is to estimate the influence of interphase on micro-crack propagation in the particulate composite. In the contribution the particle-filled polymer composite is modelled as three-phase continuum represented by infinite matrix with homogeneously dispersed identical coated stiff spherical particles. The studied composite corresponds to calcium carbonate (CaCO3) filled polypropylene.

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2. Determination of crack propagation direction

A propagation of a micro-crack in the matrix of particulate composite is controlled by its interaction with particles. To describe the interaction the micro-crack propagation direction has to be known. Generally a crack propagates in direction leading to zero values of K_{II} . For determination of crack propagation direction numbers of criterions exist in the literature. In this paper maximum tangential stress (MTS) criterion [2] has been used. Determination of crack propagation direction Ω_s can then be expressed by the following equation:

$$\Omega_S = \arccos\left(\frac{3K_{II}^2 + K_I \sqrt{K_I^2 + 8K_{II}^2}}{K_I^2 + 9K_{II}^2} \right), \tag{1}$$

where K_I and K_{II} are corresponding values of the stress intensity factors for normal and shear mode of loading.

3. Numerical model

To estimate the crack propagation direction the values of stress intensity factors for mode I and II have to be numerically calculated. To this aim the stress strain distribution of the three-phase composite with homogeneously distributed coated particles was numerically simulated on a microscopic scale using the finite element program ANSYS. A simplified 2D model has been used in the present contribution. The geometry of the model is shown in fig. 1.

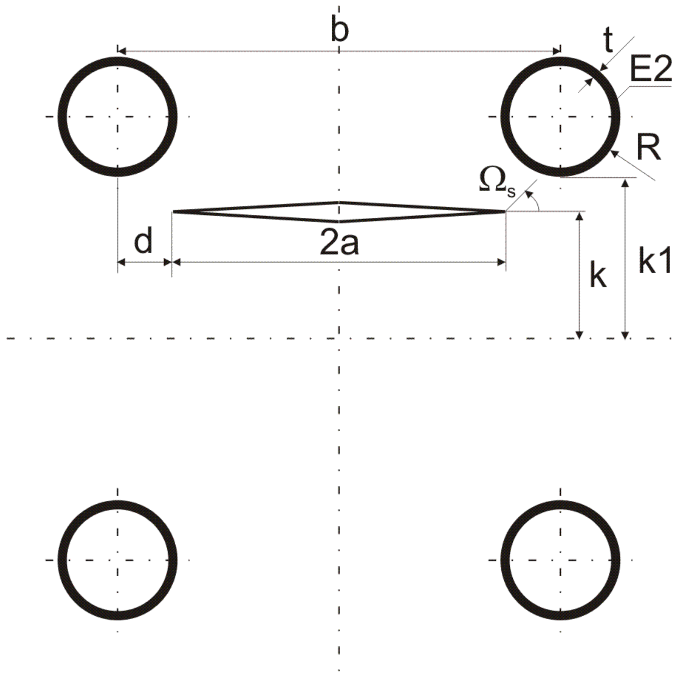

Fig. 1. The geometry of the 2D model used for estimation of micro crack behaviour in the studied particulate composite.

Tensile load was applied via a prescribed force F. The finite element model with symmetry boundary conditions is shown in fig.2. For calculations plane stress conditions were assumed. Two dimensional isoparametric elements (PLANE82) were non-homogenously distributed, because of the material inhomogenity and high stress concentration in the crack tip. The typical finite element model has about 40 000 elements.

The material properties characterizing the composite corresponding to calcium carbonate (CaCO3) - filled polypropylene (PP) at room temperature are used. The calculations have been performed for rigid particle dimension (given by radius of the $CaCO_3$ particles R = 5 μm) and wide interval of particulate filler volume fraction between 10 – 35 %. The Young´s modulus of the particles E = 72 GPa, and the value of Poisson´s ratio ν = 0.29. The corresponding parameters of the neat polymer matrix (PP material) are E = 1.8 GPa, ν = 0.36. The thickness t of the interphase 50 nm is considered here only. The perfect adhesion between particles, interphase and matrix was assumed. The stress and strain distributions in the matrix have been determined for a variety of the interphase properties. The value of Young´s modulus of the interphase ranges from 0.05 to 1.80 GPa. It is assumed that Young's modulus of the interphase is constant through its thickness. A micro-crack of length corresponding approximately to the distance between the particles was modelled and the corresponding values of the stress intensity factors K_I and K_{II} were calculated for different micro-cracks configurations.

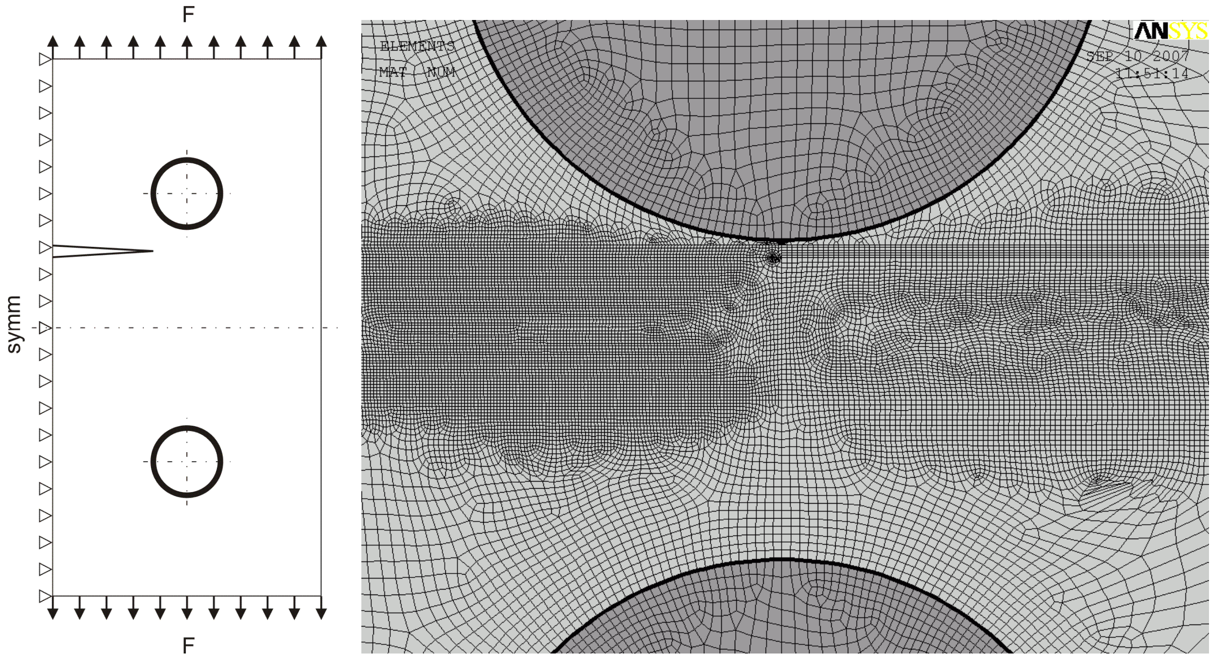

Fig. 2. Schematic representation of FEM model used for calculation of the stress intensity factor.

4. Numerical results

The corresponding values of stress intensity factors K_I and K_{II} were estimated using the standard KCALC procedure as implemented in ANSYS. The mesh around the crack tip has to be refined because of high stress concentration. Special "crack" finite elements with shifted mid-nodes and modeling the near tip stress singularity were applied. Obtained values K_I and K_{II} were used for estimation of further crack propagation direction Ω_s using eq. (1), see fig. 3.

The influence of the volume fraction of the composite on the crack located close to the particle, i.e. for $k/k1$ = 0.9 and $2a/b \rightarrow 1$ has been studied. Six configurations were modeled: for three values of filler volume fraction and for two limiting values of interphase moduli. The curves accordance with value of Young´s modulus 0.05 GPa are marked by quads and curves accordance with value of Young´s modulus 1.8 GPa are marked by circles. It has to be mentioned, that Young´s modulus of the interphase 1.8 GPa corresponds to Young´s modulus of the matrix and behavior of this configuration corresponds to the two-phase composite without interphase. Strong decrease of the angle of crack propagation Ω_s corresponds to Young´s modulus 1.8 GPa for all mentioned filler volume fractions. In this case the micro-crack propagates purely in the matrix and has a tendency to deflect to rigid particles. For this material configurations the direct interaction between particle and crack is rare a have no influence on fracture toughness of the composite.

Contrary to it for interphase with Young´s modulus 0.05 GPa the influence of rigid particles is shielded by a soft interface and even for a small volume fraction of the interphase, the behaviour of the micro-crack can be changed. The crack deflection is much smaller and in some cases crack cannot avoid the particle and is attracted to it. Final configuration corresponds then to a micro crack with its tip on the interface between matrix and interphase. Due to existing high stress concentration matrix and particle are debonded and as a consequence, the crack is blunted. This is connected with strong decrease of the stress near the crack tip and the singular stress field is changed to regular one. The crack is transformed to a notch and arrested near the particle. Stronger influence for crack behavior was observed for volume fraction 35%. Therefore only this special case is discussed here in the details.

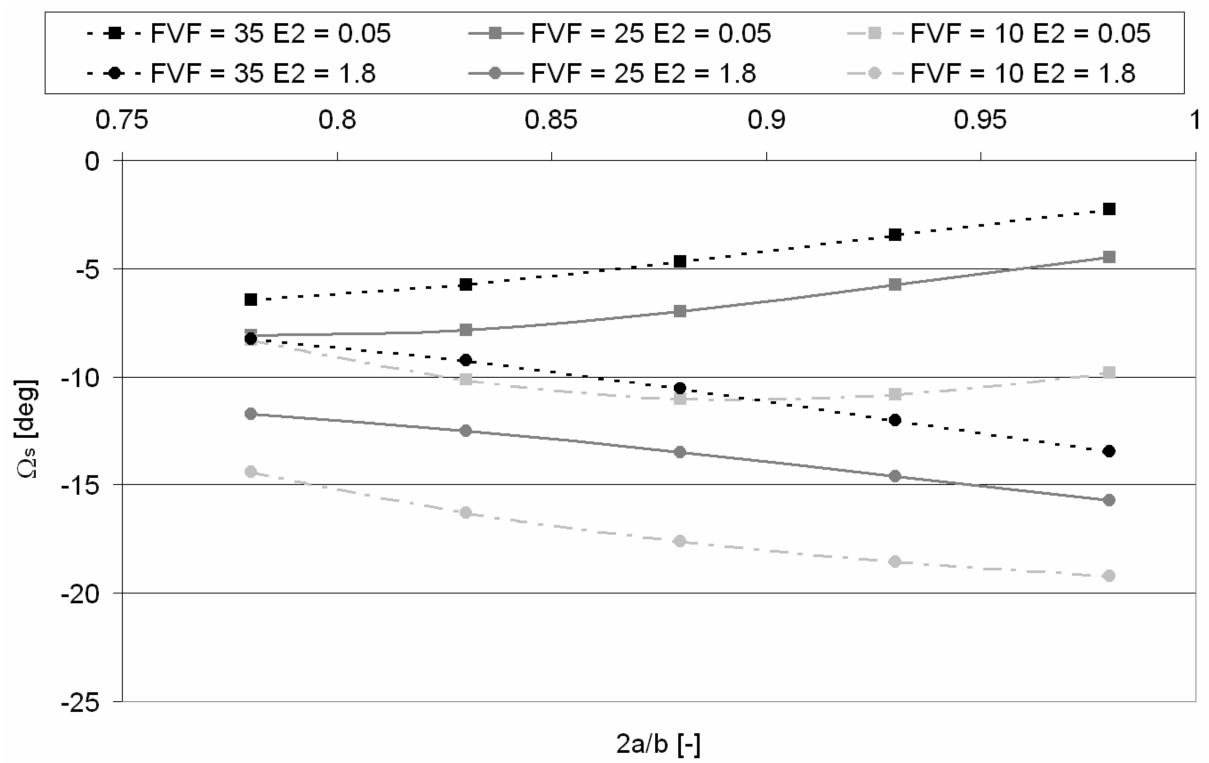

Fig. 3. Dependence of crack propagation direction Ω_s on ratio *2a/b* for variety of filler volume fraction (FVF) and elastic moduli *E2* of interphase.

Again six configurations with filler volume fraction 35% was modeled, three for variety of ratio of *k/k1* and for two different moduli of interphase, see in fig. 4. For small ratio of *k/k1* (0.1 in our case) the crack is close to symmetrical position between two particles, therefore the change of crack propagation direction is minimal. The influence of the particles and interphase is visible mainly for the crack with k/k_1 = 0.5 -0.9, where the influence of the interphase started to be important.

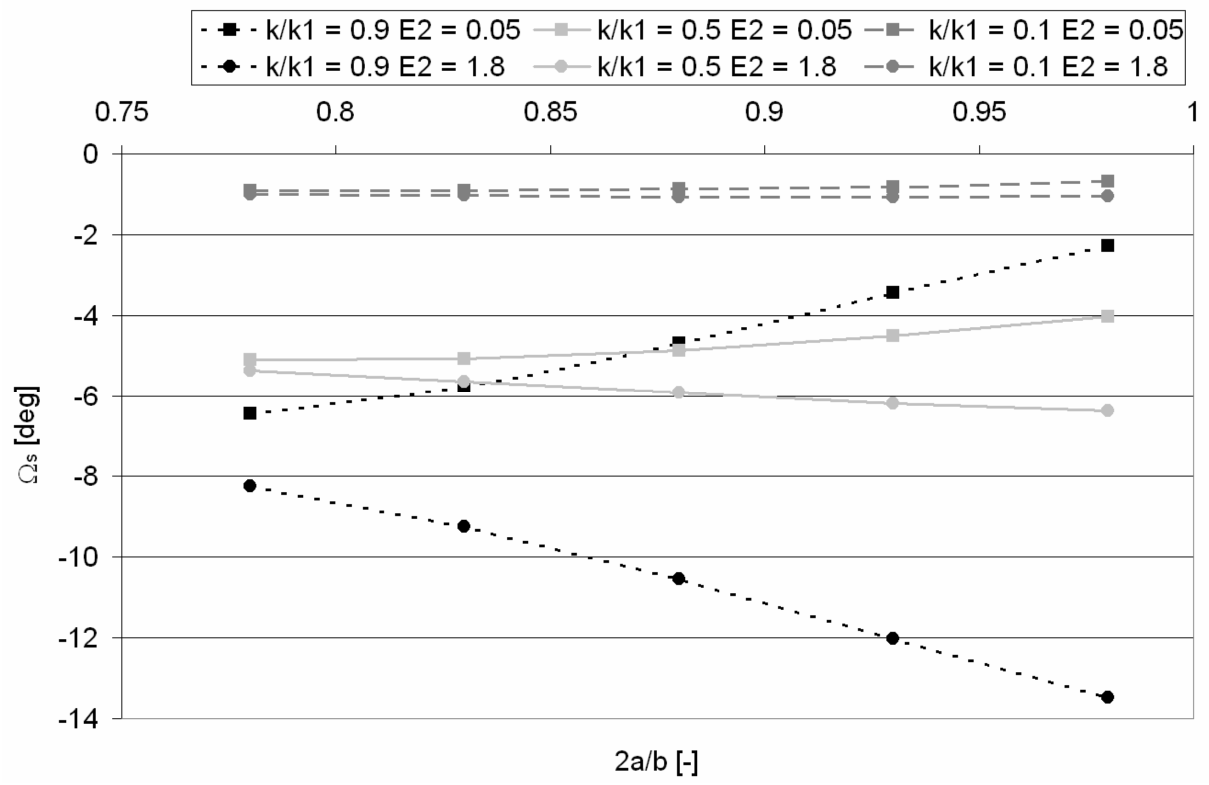

Fig. 4. Dependence of crack propagation direction Ω_s on ratio *2a/b* for filler volume fraction 35% for variety of ratio *k/k1* and elastic moduli *E2* of interphase.

5. Conclusion

The addition of mineral fillers to polypropylene (PP) can profoundly change the mechanical properties of a polymer system. The properties of the particles themselves (size, shape, material properties) can have a significant effect on the global behaviour of the composite. Computation of the internal stress and strain fields in the body with particles is the first step towards to understanding of the composite in macro- and micro levels. In the contribution finite element simulations based on the microstructure of polymer composite filled by coated particles are conducted in order to transfer the information from micro- to macro-scale. To this end finite element analyses were carried out to study the influence of the interphase on the fracture toughness of the composite. The stress distribution was determined for a variety of thickness and the material properties of the interphase as well as the corresponding values of the stress intensity factors for a micro-crack crack lying in the matrix were evaluated. It is suggested that the interaction between particles and micro-cracks plays a deciding role. The simplified 2D model of a micro-crack interacting with the nearest particle was used. The influence of the interphase between rigid particle and matrix on toughening mechanism was investigated. The basic mechanism of the composite toughening due to micro-crack propagation consists in shielding of rigid particles by soft interphase followed by debonding of the particle and the matrix. As a consequence, the crack is blunted and can be arrested on the particle. The intensity of this effect depends mainly on the size and quality of the interphase.

Presented computational methodology give us powerful tool to quantified effects of particular micro structural properties on macroscopic material response. Consequently, numerical calculations can help design polypropylene based composite with specifically requested material properties.

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