Fabrication of composites via spouted bed granulation process and simulation of their micromechanical properties

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Abstract. In this contribution numerical simulation of Young's modulus of copper-polymer composites is presented. For the simulation of the composites the Bonded-Particle-Model was applied. The model allows representing of the structure of composite materials realistically. The polymer matrix, which surrounds the particles, was represented as network of solid bonds connecting copper particles. Simulation results were validated based on mechanical determination of modulus of elasticity. The modulus of elasticity was approximated in experiments as well as in simulation by four-point-bending tests. It was observed, that obtained simulation results are in good agreement with experimental results.

1 Introduction

Beside of one-component materials like pure metals there exist also materials, which consist of two or more components, which are arranged in a particular way. These materials are called composites. Naturally occurring composites consist mostly of ceramics and polymers, which are structured in several hierarchical levels [1-2]. The structure of the composites shows that ceramic particles are uniformly distributed in a polymer matrix and the content of ceramics is significantly higher than that of polymer. The high content of ceramic in polymer matrix is significant for reinforcement of the polymer. Polymer in the composites ensures the slight ductility of the materials. One of the best investigated natural materials is nacre. Nacre consists of small ceramic platelets. The platelets are surrounded by a polymer matrix (see fig. 1). The ceramic content in nacre is about 95 vol.%. Due to such a high amount of ceramic and further aspects like high aspect ratio of ceramic particles, nacre has very good mechanical properties. Strength of nacre reaches values up to 120 MPa and modulus of elasticity is up to 70 GPa.

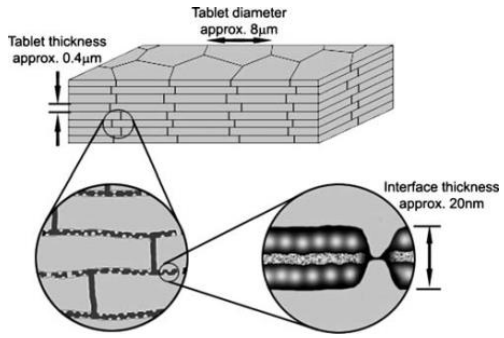

Fig. 1. Structure of nacre [2].

The good mechanical properties of natural materials gave an impulse for the reproducing of the natural structures. Therefore, in recently years, many attempts have been made to reconstruct the structure of the natural material and to reach a high ceramic content. One possible route for the reproducing of natural composite materials is the spouted bed spray granulation process. The application of the process has allowed reaching of very high ceramic contents in composite materials [3] and also reproducing of the hierarchical structure [4].

Spouted bed is a special form of fluidized bed with many applications in particle technology, such as drying, granulation, coating and sometimes also agglomeration. For processing of fine and cohesive particles a special spouted bed was designed. It could be successfully applied also for fabrication of ceramic-polymer composites [3, 4]. During processing, solid particles are covered with a polymer layer. Afterwards, coated agglomerates are pressed to get a bulk material. During warm pressing the melting of polymer occurs and it can fill the gaps between particles.

For the characterization of final material, its composition and its mechanical properties are evaluated. The determination of mechanical properties is carried out by four-point bending tests. Bending tests deliver strength and modulus of elasticity for investigated composites.

Numerical modelling of composite material plays important role for optimization of material structure and allow minimizing experimental effort. The mechanical properties of composites can be simulated by discrete element method (DEM). Initially, DEM was developed as a method for numerical simulation of granular materials consisting of ideally spherical particles. In

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DEM each particle is considered as a separate element and Newtonian equations of motion are solved for each particle [5]. As a consequence, motion and deformation behaviour, and stress state of single particle and of particle collectives can be calculated [6]. Many different extensions of DEM and additional contact models were developed in recent years. One of these extensions is the Bonded-Particle-Model (BPM). In the BPM materials are represented as particles, which are connected to each other by solid or liquid bonds. For the simulation of composite materials the BPM is probably one of the best suitable models, because composites consist of discrete particles, which are surrounded by a polymer matrix. The BPM represent the particles surrounded polymer by solid polymer bonds. This allows reflecting the real material structure with high precision. Nevertheless, the numerical model does not directly correspond to the real material. Therefore, the mechanical properties of polymer cannot be just taken from experimental tests to simulations and additional adjustment is needed.

In this contribution the BPM was used to simulate mechanical properties of copper-polymer composites. Solid bonds among particles were described with elastic solid beam model [7].

2 Methods

DEM simulations are carried out with an in-house developed software MUSEN. In the first stage, a bending beams consisting of primary particles and solids bonds were generated. The dimensions of simulated bending beams were approximately 10 times smaller than in the experiments. Dimensions of the beam are summarized in table 1.

Table 1: Dimensions of the bending beam.

Beam length (x)	Beam height (z)	Beam width (y)
4270 µm	235 µm	256 µm

In the experiments and simulations investigated composite material was consisting of copper primary particles and polyvinyl butyral polymer. The particles in experiments were not monodisperse therefore in simulation a particle size distribution was generated. Particles had a size between 6 and 40 µm and they were distributed in 4 fractions, so the mean diameter of particles was 30 µm as in experiments.

After generation of copper particle in the bending beam, they were connected to each other by polymer bonds. So the continuous polymer matrix surrounding the particles in experiments is represented by polymer bonds, which connect particles. During bending tests the relative motion of aggregated primary particles causes forces and moments in the solid bonds [8]. In each simulation step the forces and moments in the bonds are calculated as an increment to the corresponding values from previous time step [9]. The forces in tangential and normal direction are determined as:

$$\bar{F}_{b,t}^{i} = T \cdot \bar{F}_{b,t}^{i-1} - \bar{v}_{rel,t} \cdot \Delta t \cdot k_t \cdot A_b \qquad (1)$$

$$\bar{F}_{b,t}^{i} = k_n \cdot A_b (L_{init} - L_{cur}) \cdot \bar{r}_n \qquad (2)$$

where L_{init} and L_{cur} are the initial and current length of the bond, A_b is the bond cross-cut surface area, $\bar{r}_n$ is the unit vector between bonded particles, k_n and k_t are the normal and tangential stiffness of the bond, and Δt is the time step. For taking into account motion of the particles in the space, the values calculated during the previous iteration are multiplied by a transformation matrix T. The length of the bond L is calculated from distance between centres of particles L_{pp}, radii of the particles R_1, R_2 and radius of the bond R_b:

$$L = L_{pp} - \sqrt{R_1^2 - R_b^2} - \sqrt{R_2^2 - R_b^2} \qquad (3)$$

The moments in bonds in tangential and normal direction are calculated as:

$$\bar{M}_{b,t}^{i} = T \cdot \bar{M}_{b,t}^{i-1} - \bar{\omega}_{rel,t} \cdot \Delta t \cdot k_n \cdot I \qquad (4)$$

$$\bar{M}_{b,n}^{i} = T \cdot \bar{M}_{b,n}^{i-1} - \bar{\omega}_{rel,n} \cdot \Delta t \cdot k_t \cdot J \qquad (5)$$

where $\bar{\omega}_{rel,t}$ and $\bar{\omega}_{rel,n}$ are relative rotational velocities between two bonded particles in tangential and normal direction, respectively. I is the moment of inertia and J is the polar moment of inertia of the bond cross section [9].

The breakage criteria for bonds has following definition: if shear or tensile stress exceed the corresponding bond strength, then the bond breaks and is removed from the calculation procedure [9].

For the simulation of four-point-bending tests two upper and two lower supports were added to the simulation. The upper supports were moved down with a constant loading speed of 10 mm/s. Lower supports were stationary. Thereby beam was bended. A sketch of the bending beam is presented in fig. 2.

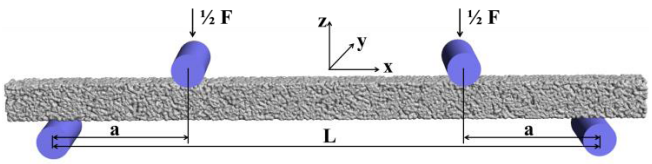

Fig. 2: Bending beam for DEM simulation. L=3.6 mm, a=0.25*L.

The modulus of elasticity for experiments and for simulations was calculated according to:

$$E = \frac{F * L^3}{8bh^3 d} \qquad (6)$$

Thereby F is bending force, L distance between lower supports, b width of sample, h height of sample and d deflection.

Properties of copper particles and steel supports were identical to experiments and are listed in table 2.

Table 2: Properties of copper particles and steel supports for DEM simulations.

Property	Unit	Copper	Steel
Density	kg/m^3	8930	7860
Modulus of elasticity	GPa	120	200
Poisson‘s ratio	-	0.34	0.3

In contrast to the fixed parameters of particles, properties for polymer bonds had to be adjusted to reproduce experimental results of four-point-bending tests [10]. This had to be done, because the presentation of continuous polymer matrix surrounding the copper particles by cylindrical polymer bonds, which connect two particles, deviates from reality slightly.

3 Results

For the adjusting of polymer properties, a composition with a copper content of 65.5 vol.% was used. The rest material in the sample was the polymer, whereby also the porosity of about 5 vol.% was taken into account. The modulus of elasticity at the composition was calculated by linear interpolation between two in experiments determined modulus of elasticity at compositions with copper contents of 60.6 and 69 vol.% respectively. Beside of modulus of elasticity also normal and tangential strength in polymer, and coordination number were adjusted. The coordination number describes an average number of solid bonds connected to single particle. By variation of the parameters in simulations best suitable parameters were determined. These parameters are presented in table 3.

Table 3: Coordination number of particles, modulus of elasticity, and normal and tangential strength of polymer.

Coordination number [-]	6
E_{PVB} [GPa]	6
$\sigma_{n,t}$ [MPa]	150

In further simulations these parameters were used. Figure 3 shows the force distribution in the bending beam at different loadings and deflections. The upper beam is unloaded, the beam in the middle is slightly loaded and the lower beam is fully loaded.

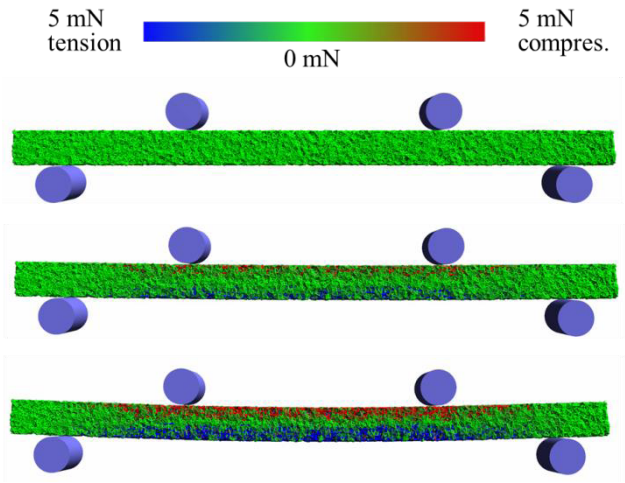

Fig. 3: Force distribution in polymer bridges of bending beam: unloaded (above), slightly loaded (middle) and loaded (below).

Especially at high loading three areas can be clearly differentiated. The upper part of the beam is compressed. This can be seen on the positive forces in the upper part. Negative forces in the lower part of the beam mean that there act tensile forces. The part in the middle of the beam stays unloaded also at high loadings. This loading distribution in a bending beam correlates to experimental and theoretical loading distribution.

After the adjustment of the parameters for the polymer, simulations at other compositions were carried out with the parameters. For further simulations four different compositions were chosen. For the simulations compositions with a copper content of 62, 64, 67 and 69 vol.% were used. Higher copper content led to a higher number of particles in the beam. For each simulation the porosity was set to 5 %. This corresponds to mean porosity in experiments. The rest in the beam was polymer. The parameters for simulations are summarized in table 4.

Table 4: Parameters for simulations of modulus of elastic in copper-polymer composites.

Parameters	**sample 1**	**sample 2**	**sample 3**	**sample 4**
Copper content [vol.%]	62	64	67	69
PVB content [vol.-%]	34	31	28	26
Porosity [%]	5	5	5	5
Number of particles	14570	15650	16190	17269

Modulus of elasticity determined by simulation and comparison between simulation and experimental results are presented in figure 4.

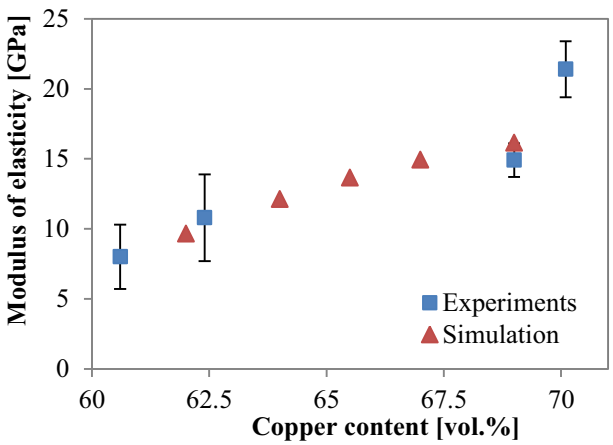

Fig. 4: Comparison between experimental [10] (blue) and DEM simulations (red) results. Here the dependency modulus of elasticity on copper content in copper-polymer composites is illustrated.

Figure 4 shows that the simulation could reproduce experimental values of modulus of elasticity for copper-polymer composites very well. The adjustment of simulation parameters was done only for one copper content. The moduli of elasticity at lower and higher copper contents could be reproduced automatically with high accuracy. This proves that the proposed modelling approach could be effectively applied for simulations of mechanical behaviour of composite materials.

4 Conclusions

In this contribution the applicability of the Bonded-Particle-Model for numerical modelling of mechanical properties of composite materials was investigated. The microscale model was validated on experimental results obtained from four-point-bending tests. Afterwards, it was successfully applied for composites with different higher and lower copper contents. The obtained results show that the model can be effectively applied for investigation of modulus of elasticity in composite materials.

We gratefully acknowledge financial support from the German Research Foundation (DFG) via the collaborative research center SFB986 "M3".

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