

EXPERIMENTAL CHARACTERIZATION AND MODELLING OF MECHANICAL BEHAVIOR OF MICROCAPSULES IN COMPOSITES

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1 Introduction

Microcapsules are applied in many advanced materials and composites [1, 2]. The concept of the use of microcapsules as sensors of damage indication for high-performance polymer composites is one of the most relevant. This concept is based on the integration of microcapsules into different layers of the composite. If mechanical load brings to a capsule rupture, a dye is released which is color contrast in relation to the state material. Especially actual this is for types of damages referred as barely visible impact damage, where small indentation on the surface may lead to significant delamination or other internal damages. Experimentally developed polymer composite with an inherent function of damage visualization and evaluation of the mechanical properties of the system is a first step in the non-destructive approach realization [3-5]. Making predictions of the mechanical behavior and response of these self-sensing structures with microencapsulated active materials embedded into polymer matrices by numerical simulations and analytical analysis based on experimental data is a good possibility to understand a real phenomenon that occurs at the micro level of the composites [6]. Due to the fact that microcapsules placed into a polymer matrix effect on mechanical properties of the material, the theoretical evaluation and modelling of the properties of microcapsules are important for the prediction of the behavior of the entire self-sensing structure.

The aim of the research is to predict mechanical properties of smart polymer materials containing microcapsules.

2 Materials

Microcapsules with leuco dye (MCD) and microcapsules with dye activator (MCA), as water based dispersions were supplied by *Papierfabrik August Koehler Ag., Germany*. The shell of microcapsule is made from melamine-formaldehyde (MF) resin. The nominal concentration of the microcapsules in the dispersion is $\approx 40\%$ (data from the manufacturer).

3 Experimental characterization of a single microcapsule

Experimental characterization of a single microcapsule was performed by optical and electronic microscopy, light scattering particle size analyzer. Average hydrodynamic diameter of MCD capsules was measured as $D = 7.0 \pm 0.5 \mu\text{m}$ while for MCA was $D = 2.0 \pm 0.2 \mu\text{m}$. The average wall thickness of the microcapsules was defined $h = 0.10 \pm 0.01 \mu\text{m}$.

Mechanical properties of microcapsules were investigated by AFM and nanoindentation. The value of the elastic modulus was calculated based on force-displacement diagrams according thin shell theory applying equation found for shallow spheres by Reissner [7]. Interrelation between the force F and the normal displacement of the pole d under point loading is given by Eq. (1):

$$F = \frac{h^2}{R} \cdot \frac{4E}{\sqrt{3(1-\nu^2)}} \cdot d \quad (1)$$

where E is elastic modulus of microcapsules shell material, ν the Poisson ratio, h is the shell thickness, R is the radius of capsule. Elastic modulus of shell

material for the individual microcapsule with the diameter $D = 1.5 \text{ } \mu\text{m}$ was estimated as $E = 3.5 \pm 0.3 \text{ GPa}$.

4 Modelling of mechanical behavior of a single microcapsule

Modelling of mechanical behavior of a single microcapsule was performed both analytically and numerically. Buckling of a thin-walled spherical shell under external pressure was considered firstly. Analytical solutions allow to calculate the critical buckling stress, critical pressure, half-wave length of buckling, and the number of buckling waves that was in a good agreement with numerically calculated by using the finite element method (FEM).

FEM model was verified on two demonstrator shells - table tennis ball (TTB) and closed cylindrical shell (CCS). Compression behavior of both demonstrator shells between two rigid plates for empty shells and ones filled with incompressible liquid (water) was in a good agreement with experiments.

5 Mechanical behavior of microcapsules in composites

Mechanical behavior of polymer composite films based on epoxy resins and polyvinyl acetate (PVA) matrixes filled with microcapsules (concentrations up to 20 wt. %) was investigated. The mechanical properties (elastic modulus, tensile strength and elongation at break) of filled polymer specimens were obtained from tensile tests performed by Zwick 2.5 testing machine. Applying *Voigt* and *Reuss* approximations for the experimental results, the effective elastic modulus of microcapsules in epoxy matrix was estimated as $E = 0.2 \text{ GPa}$ and $E = 1.0 \text{ GPa}$ for microcapsules in PVA matrix.

For modelling of microcapsules in PVA matrix, one eighth symmetry model representative volume element (RVE) was created. Multilinear Isotropic Hardening material model was used both for MF resin and PVA. Nonlinear structural static analysis was performed to evaluate the influence of microcapsules diameter (volume fraction) to the stress-strain state of microcapsules and matrix. The constant small displacement on the top surface of RVE was used for loading. Such deformation usually does not initiate the plastic strain in matrix, but the stress concentrations between the PVA and microcapsule surface can influence the stress-strain state changes in some areas. The results show that very small microcapsules or in other words small amount of microcapsules (ca. 1 %) can cause the increase of plastic strains both in the matrix and in the

microcapsules shells. In this case microcapsules act only as the stress concentrators and can drastic decrease the mechanical properties of composite. As the diameter of microcapsule increases the plastic strains in the matrix and microcapsule decrease. The experimental testing also shows strength properties increasing as the volume fraction of microcapsules increases.

Conclusions

Mechanical behavior of single microcapsule with MF shell and microcapsules embedded in polymer matrixes was investigated experimentally and modelled analytically and numerically. Elastic approach was successfully used for prediction of small strain of both microcapsules and composites. Liquid in microcapsules and plastic strains of capsule shell and polymer matrix play essential role for large strains.

References

- [1] Y.-K. Song, et al., "A microcapsule-type fluorescent probe for the detection of microcracks in cementitious materials". *Sensors and Actuators B: Chemical*, 2015. 222: p. 1159-1165.
- [2] W. Li, et al., "Autonomous indication of mechanical damage in polymeric coatings". *Advanced Materials*, 2016. 28(11): p. 2189-2194.
- [3] O. Bulderberga, A. Aniskevich, and S. Vidinejevs, "A glass-fiber-reinforced composite with a damage indication function". *Mechanics of Composite Materials*, 2016. 52(2): p. 155-162.
- [4] S. Vidinejevs, et al., "Development of a composite with an inherent function of visualization of a mechanical action". *Mechanics of Composite Materials*, 2013. 49(1): p. 77-84.
- [5] O. Bulderberga and A. Aniskevich, "Polymer composite material with inherent function of damage visual indication". *21st International Conference on Composite Materials, ICCM 2017*; Xi'an; China, 20-25 August, 2017. , 2017.
- [6] D. Zeleniakiene, et al. "A numerical simulation of mechanical properties of smart polymer composite with microcapsules for damage sensing applications". *ECCM17 - 17th European Conference on Composite Materials*. 2016. Munich, Germany.
- [7] A. Fery, and R. Weinkamer, "Mechanical properties of micro- and nanocapsules: Single-capsule measurements". *Polymer*, 2007. 48(25): p. 7221-7235.