Microstructure-property relationships in alumina trihydrate filled poly (methyl methacrylate) composite materials

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Microstructure-property relationships in alumina trihydrate filled poly (methyl methacrylate) composite materials

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Abstract. The mechanical properties (Young's modulus and fracture toughness) of composite made from a poly (methyl methacrylate) (PMMA) matrix filled with alumina trihydrate (ATH) are reported. The experiments were performed using flexural tests and single edge notched bend (SENB) tests. The composites samples were tested at a range of filler volume fractions (34.7%, 39.4% and 44.4%) and mean filler diameters (8 μm, 15 μm and 25 μm). The data of Young's modulus agreed well with the results of Lielens model and finite element analysis (FEA) model.

1. Introduction

Particle-filled polymer composites are important among engineering materials due to their ability to provide both economic and technical advantages. The work here is concerned with ATH filled PMMA composites at high filler volume fractions. These composites have been widely studied [1-3]. One of the major concerns is the relationship between the microstructure and the mechanical properties of these composites. The elastic behavior of particle-filled polymers is well understood and several models have been developed [4, 5] to describe the relationship between Young's modulus and the addition of filler. However, the effects of filler on the fracture toughness are not well understood. Generally, as the modulus increases, the fracture toughness decreases. In this work, fracture toughness tests were performed on composites with different filler volume fractions (34.7%, 39.4% and 44.4%) and mean particle diameters (8 μm, 15 μm and 25 μm). In addition, by comparing with the experimental results, the reliability of FEA modelling results is verified.

2. Experiments and methods

2.1. Materials

ATH/PMMA composites used in this work were provided by E.I. DuPont Nemours & Co. (Inc.) in the form of sheets (250 mm x250 mm x12 mm). The particles were mixed with the matrix using a four-blade marine-type propeller and the sheets were manufactured using casting process. These samples were all prepared using a proprietary adhesion promoting agent as part of the matrix composition. Each composite is formulated using the same type and amount of adhesion promoter and crosslinker. Table 1 shows the manufacturer's (DuPont) specifications. Particles with a 15 μm mean diameter were

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used for the manufacturing of composites with codes 1, 3 and 5. For these composites, the filler volume fractions were 34.7%, 39.4% and 44.4%, respectively. Keeping the same filler volume fraction as 3 (i.e. 39.4%), 8 µm particles were used for manufacturing composite 2, while 25 µm particles were used in composite 4.

Table 1. Particle size and volume fraction of the ATH/PMMA composites.

Composite Code	Volume Fraction ATH Filler (%)	Mean Particle Size of ATH Filler
1	34.7	15
2	39.4	8
3	39.4	15
4	39.4	25
5	44.4	15

2.2. Experimental

The flexural tests were performed according to ASTM D790M-10 [6]. The specimens used were simple rectangular beam specimens, with dimensions of 12x12x200 (mm^3) in accordance with the standards. The loading span L was 160 mm. The crosshead rate was 4.5 mm/min.

The SENB tests were performed according to ISO 13586 [7]. The specimens were rectangular beams, of dimensions 10x20x100 (mm^3). A 10mm notch was machined in each specimen and then sharpened using a razor blade. The tests were performed at a cross-head motion rate of 10mm/min, while the compliance correction test was performed at 1mm/min.

2.3. Analytical model

The results of Lielens model were used to verify the experimental results. Like most analytical models, Lielens model is based on the following equation [4]:

$$\boldsymbol{C} = \boldsymbol{C_m} + \emptyset_f(\boldsymbol{C_f} - \boldsymbol{C_m})\boldsymbol{A} \tag{1}$$

where $\boldsymbol{C}$ are the elastic stiffness tensors, while the subscripts m and f represent the matrix and fillers, respectively. $\boldsymbol{A}$ is the strain concentration factor [4] and $\emptyset_f$ is the particle volume fraction. $\boldsymbol{C}^{-1}$, or compliance tensor, $\boldsymbol{D}$, can be expressed as:

$$\boldsymbol{D} = \begin{bmatrix} \frac{1}{E_{11}} & -\frac{v_{12}}{E_{11}} & -\frac{v_{12}}{E_{11}} & 0 & 0 & 0 \\ -\frac{v_{12}}{E_{11}} & \frac{1}{E_{11}} & -\frac{v_{23}}{E_{22}} & 0 & 0 & 0 \\ -\frac{v_{12}}{E_{11}} & -\frac{v_{23}}{E_{22}} & \frac{1}{E_{22}} & 0 & 0 & 0 \\ 0 & 0 & 0 & \frac{11v_{23}}{E_{22}} & 0 & 0 \\ 0 & 0 & 0 & 0 & \frac{1}{2\mu_{12}} & 0 \\ 0 & 0 & 0 & 0 & 0 & \frac{1}{2\mu_{12}} \end{bmatrix}$$

where E_{11} is longitudinal Young's modulus, E_{22} is transverse Young's modulus, μ_{12} longitudinal shear modulus, v_{12} is longitudinal Poisson's ratio and v_{23} is transverse Poisson's ratio. Hence, if $\boldsymbol{A}$ is obtained, the mechanical properties of the composite can be calculated accordingly.

Lielens model is a bounding method, in which the reference materials for upper and lower bound are filler and matrix, respectively. Using the rule of mixtures between the upper and lower bound, Lielens et al. proposed a 'convenient' strain-concentration factor $\boldsymbol{M_L}$, which can be calculated according to:

$$\boldsymbol{M_L} = \left[(1 - F_L)(\boldsymbol{M_{lower}})^{-1} + F_L\left(\boldsymbol{M_{upper}}\right)^{-1}\right]^{-1} \quad (2)$$

where $\boldsymbol{M}_{lower} = \left[\boldsymbol{I} + \boldsymbol{S_m}\boldsymbol{C_m^{-1}}\left(\boldsymbol{C_m} - \boldsymbol{C_f}\right)\right]^{-1}$, $\boldsymbol{M}_{upper} = \left[\boldsymbol{I} + \boldsymbol{S_f}\boldsymbol{C_f^{-1}}\left(\boldsymbol{C_m} - \boldsymbol{C_f}\right)\right]^{-1}$ and $F_L = \emptyset_f + \emptyset_f^2/2$. $\boldsymbol{S_m}$ and $\boldsymbol{S_f}$ are the Eshelby's tensors of the matrix and the particle, respectively.
The strain concentration factor of Lielens' model can then be calculated according to:

$$\boldsymbol{A_L} = \boldsymbol{M_L}\left[\left(\boldsymbol{1} - \emptyset_f\right)\boldsymbol{I} + \emptyset_f\boldsymbol{M_L}\right] \quad (3)$$

2.4. Finite element analysis

The FEA modelling was performed using Abaqus [8], and the microstructure geometries of the composites were converted from SEM images using a Matlab python proposed by Tarleton et al. [9]. In a binary image, the position of a pixel can be defined by its coordinate (*i, j*). By defining a black pixel (matrix) as f_{ij}=0 and a white one (filler) as f_{ij}=1, the position of a white pixel can be identified according to its four neighbours. By calculating a value, $k_{ij}=f_{i+1j}+f_{i-1j}+f_{ij+1}+f_{ij-1}$ (see figure 1), if k_{ij}=4, the pixel lies in the bulk of a particle. If k_{ij}=3, the particle lies on a vertical or horizontal boundary of a particle. If k_{ij}=2, the pixel lies on a diagonal boundary or a corner. If $k_{ij}=1$ or $k_{ij}=0$, the pixel will be removed from the microstructure to simplify the microstructure. The position of each pixel is then input into Abaqus for the creation of the 2-D FEA microstructure.

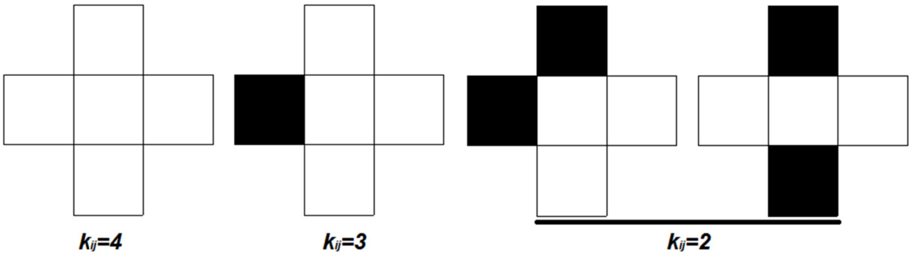


Figure 1. The identification of the position of a white pixel.

3. Results and discussion

Figure 2 shows the G_{IC} values as a function of filler volume fraction (A) and particle diameter (B). It can be seen that higher filler content lead to lower fracture .This can be explained by that as more particles were added, the deformation of the matrix around the crack tip was more limited and hence, less energy can be absorbed during the crack growth. In addition, due to the high filler volume fractions of the composites, further increase in the filler content can lead to the agglomeration of the particles. The agglomerated particles induce stress concentrations, which can initiate cracks and make

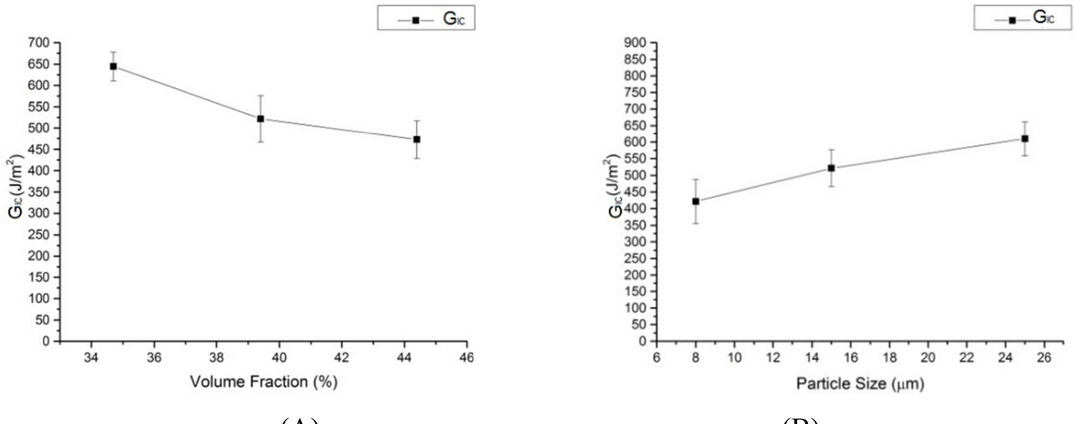
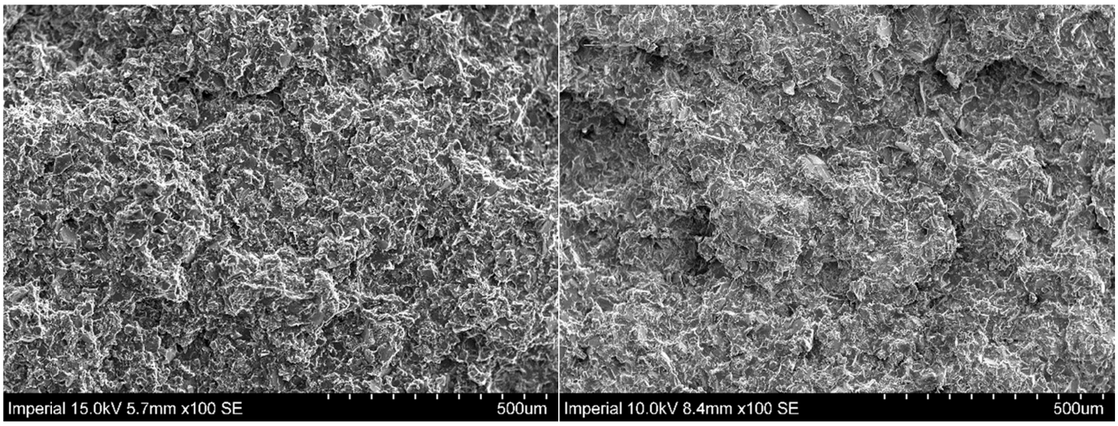


Figure 2. GIC values as function of (A) filler volume fraction and (B) particle diameter.

the crack extend until the cracks are larger than the critical size that cause failure, and hence, reduce the fracture toughness of the composites.

On the other hand, larger particles lead to higher G_{IC} values. This is because that in the composites filled with smaller particles, the crack propagated in the matrix was deflected by the particles slightly, while in the composites filled with larger particles, the crack propagated through both the matrix and the particles. This conclusion is supported by the images shown in figure 3, in which a smoother fracture surface can be observed in the composite filled with 8μm particles than in the one filled with 25 μm particles.



(A) (B)

Figure 3. Fracture surface of composite filled with (A) 8 μm particles and (B) 25 μm particles.

An image-converted FEA microstructure is shown in figure 4. A tensile simulation was performed on the microstructure of each composite to get the Young's modulus. The results of FEA modelling agreed well with the results of Lielens model (figure 5), which means the image based FEA geometry can represent the microstructure of the composites and hence, could be useful in the future work of fracture toughness modelling.

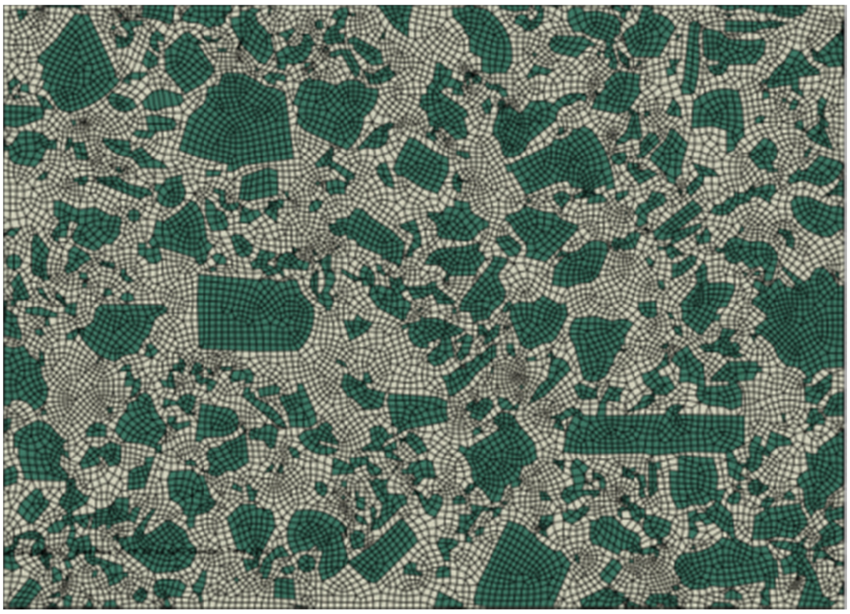


Figure 4. SEM image converted FEA microstructure of ATH/PMMA composite (8 μm, 39.4%).

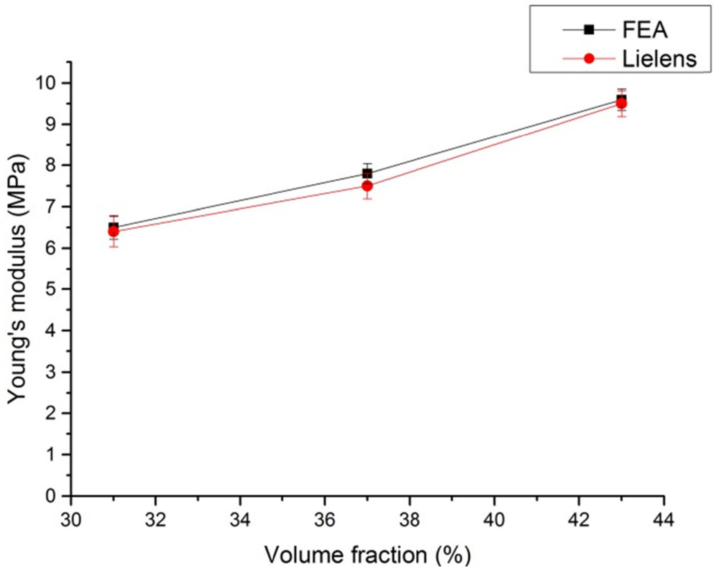


Figure 5. The results of FEA and Lielens modelling of Young's modulus.

4. Conclusion and future work

G_{IC} values of composites increase with the increasing particle size, while higher filler content means lower fracture toughness. The results of FEA modeling and Lielens modeling agreed well with each other, which means that the image converted FEA geometries can represent the microstructure of the composites. Therefore, in the future, this method will be used in the modelling of fracture toughness.

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Corrigendum: Microstructure-property relationships in alumina trihydrate filled poly (methyl methacrylate) composite materials

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