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**Numerical prediction of thermal conductivity in ZrB\(_2\)-particulate-reinforced epoxy composites based on finite element models**

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**Abstract:** Epoxy composites reinforced by Zirconium diboride (ZrB\(_2\)) particles were investigated by finite element models (FEMs). It helped to explore the relationship between the thermal conductivity of composites and the volume fraction, size, shape, orientation, and arrangement of the ZrB\(_2\) particles. The results showed that the thermal conductivity performance of composites was improved effectively when filled with ZrB\(_2\) particles. Specifically, epoxy composites filled with 50 vol% spherical ZrB\(_2\) particles had 12.05 times the thermal conductivity of epoxy resin. At the same volume fraction, the number of ZrB\(_2\) particles in the epoxy matrix has little influence on thermal conductivity due to the dimensionless models. At a high volume fraction, rectangular ZrB\(_2\) particles improved thermal conductivity more effectively than spherical particles. In the comparison of thermal conductivities among composites reinforced by rectangular fillers, the thermal conductivities of composites were clearly affected by the length-width ratios of fillers, and this effect was monotonically increasing. The vertical orientations of particles could conduct heat most effectively compared with slant and parallel orientations. The agglomerate distribution of ZrB\(_2\) particles has the negative effect of thermal diffusion in a certain direction compared with homogeneous distribution.

**Keywords:** finite element analysis (FEA); polymer-matrix composites; thermal properties.

1 **Introduction**

Over decades various types of composites have been reinforced by ceramic materials due to their outstanding hardness, corrosion resistance, and especially in thermal performances [1, 2]. Among these ceramic reinforcements, Zirconium diboride (ZrB\(_2\)) shows great performances in improving thermal conductivity. However, most of the studies fabricated composites by adding ZrB\(_2\) reinforcements to improve the thermal performances only in the fields of ceramic matrix composites (CMCs) and metallic matrix composites (MMCs) [3–7]. In order to expand the applications of ZrB\(_2\) reinforcements in improving the thermal conductivity of the composites, polymeric matrix composites (PMCs) reinforced by ZrB\(_2\) should be investigated. As a conventional polymeric matrix material, epoxy has been applied effectively in many areas in need of improvement of thermal conductivity. In this work, the thermal conductivity performance of epoxy composites reinforced by ZrB\(_2\) particles will be studied, and the results can be used as the basis for ZrB\(_2\) applied in PMCs.

In order to deal with a large number of combinations and conditions of composites, more and more researchers use numerical methods to estimate the experimental results theoretically. In PMC systems, Maxwell’s model, Bruggeman’s model, and Agari’s model etc., have been used widely. In reality, the actual properties of the materials are not as good as their assumed properties. In order to solve this problem, FEM will be an efficient way. Naya et al. used FEM to discuss pine wood dust reinforcing epoxy from 6.5 vol% to 36 vol%, and the best volume fraction in improving the coefficient of thermal conductivity is 11.3% [8]. Zain-ul-Abdein et al. discovered different packing densities affecting thermal conductivity in graphite/polymer systems by finite element analysis software [9]. Mu et al. improved thermal conductivity and friction coefficient by comparing experimental data and finite element simulation data and found the best situation in polyimide composites [10].

In our previous work [11], the FEMs in original ideal condition have been used to estimate the thermal conductivities
of epoxy composites filled with ZrB₂ particles. The results of FEM are in good agreement with experimental results and self-consistent models. Meanwhile, as a result of the developments in numerical research on epoxy/inorganic particle composites, many numerical models have been reported and they are in great agreement with experimental results. However, more research should be done to understand the relationship between thermal conductivity performances and structure of composites. Thus, in this work, epoxy composites reinforced by ZrB₂ were studied, and meanwhile, the probable factors influencing effective thermal conductivity: the size, shape, orientation, and arrangement of ZrB₂ particles were investigated and compared with the original ideal condition by the practical finite element analysis software, ANSYS provided by ANSYS Ltd., USA.

2 Numerical simulations

2.1 Finite element models in original ideal conditions

For ideal conditions of the composites, it was assumed that the ZrB₂ particles were dispersed randomly in the matrix of the composites in finite element models (FEMs). As an existing classical numerical model, Maxwell’s model which assumes that the particles are spherical without any interaction between them [12] is the basic reference for this FEM in this work. Specifically, the composites were set as square, and the ZrB₂ particles were set as spherical. The distribution of the ZrB₂ particles was set as random. The random distribution is realized by the function of Excel. Each simulated result was obtained from the average of 10 calculations. In original ideal condition, the size of ZrB₂ particles was invariant and the volume fractions of ZrB₂ were changed by adjusting the numbers of particles.

In order to obtain the general thermal conductivity of composites, it is required that the parameters of thermal conductivity corresponding to each material should be given beforehand. Herein, all materials were assumed to be isotropic, homogeneous, and independent, and specifically, the thermal conductivity of epoxy was set as 0.204 W/mK and that of ZrB₂ was set as 57.9 W/mK. For 2-D thermal entities, PLANE55 was used as the plane element type. In this type, the elements have three nodes in triangular option with one degree of freedom, temperature, at each node. Due to the irregular shape of an epoxy matrix, the three-noded triangular option was chosen to mesh the models uniformly. The model is shown schematically in Figure 1. The mesh level in each model is kept at about 70,000 nodes and 140,000 elements, respectively. In order to make the heat transfer happen for the observation, in the boundary conditions of the FEMs, the heat loads were set that there were the fixed temperature difference on the both sides of the epoxy composite. The temperature of the bottom surface was set to be higher than that of the top surface. In order to prevent the deviation caused by the particles which are too close to the boundaries, the boundaries were set at a periodic boundary condition. This setting allows that the simulated results of the bounding surfaces to repeat. The boundary conditions are shown in Figure 2 and specifically T₁ was set as 20°C and T₂ was set as 100°C.

2.2 Influences of effective thermal conductivity

Most reports about numerical simulations applied in the thermal conductivity of composites use such ideal assumptions as spherical particles and uniform dispersions, and it is inevitable that these assumptions cause systemic errors [8–10]. In order to reduce the systemic errors caused by these assumptions, the actual status of
ZrB₂ particles should be discussed and compared with the original ideal conditions, respectively. For the investigation of particle size dependence, the number and radius of the ZrB₂ particles were varied and the volume fraction was kept constant. The relationship between the number and radius of particles is shown in Eq. 1:

\[
\frac{V_f}{\pi} = \frac{nr^2}{S},
\]

where \(V_f\) is the volume fraction, \(n\) is the number, \(r\) is the radius of the particles, and \(S\) is the area of the composites. Note that this area of the composites was set as 100. For the investigation of particle shape and orientation dependence, ZrB₂ particles were set as a rectangle instead of sphere as a typical contrast. For the 2-D model, the length-width ratios of particles were set as 1(square), 1.2, 1.4, 1.6, 1.8, and 2 for comparison, respectively. In order to keep the volume fraction of ZrB₂ fillers invariant, the area of fillers follows the relationship shown in Eq. 2:

\[
r_a l^2 = \pi r^2,
\]

where \(r_a\) is the length-width ratio and \(l\) is the length of fillers.

3 Results and discussion

In FEM, the effective thermal conductivities of composites are not obtained directly as laser flash analysis; instead, they are calculated by Fourier’s law of heat conduction. According to the conditions in this study, the equation is expressed as:

\[
q'' = -k_{\text{eff}} \nabla T,
\]

where \(q''\) is the thermal flux, \(k_{\text{eff}}\) is the effective thermal conductivity, and \(\nabla T\) is the thermal gradient. The thermal flux and thermal gradient can be exported from FEM; thus, the effective thermal conductivity is available.

For the investigation of particle size dependence, the simulation results are shown in Figure 3. It was obvious that the effective thermal conductivities of composites in original ideal condition increased effectively and evenly. Specifically, the epoxy composites filled with 50 vol% ZrB₂ particles had 12.05 times the thermal conductivity of neat epoxy resin. By the comparison of the results of different numbers of particles, it is observed that the differences in thermal conductivities of epoxy composites filled with homogeneous condition, the layout of the ZrB₂ particles follows the ideal distribution which makes the separation distance of ZrB₂ particles equal to each other in the epoxy matrix for the enhancement of the different effects of homogeneous and agglomerate distribution. In the agglomerate condition, the random distribution of particles is limited in the small range which results in a crowded dispersion. The random simulated results were calculated 10 times for obtaining the averaged thermal conductivities.
different numbers of ZrB\(_2\) particles are small at the same volume fraction in FEM. In FEMs, all scales are dimensionless. It determines that the variation of the number affects only the dispersion of particles at the same volume fraction. It will not have the particle size effect in reality. The small differences of thermal conductivities in Figure 3 could be caused by random errors.

For the investigation of particle shape dependence, the results are shown in Figure 4. In this simulation, the orientations of particles in these composites were set as random. Compared with the results of thermal conductivity of composites filled with spherical and rectangular ZrB\(_2\) particles, respectively, it is observed that at 20 vol% of ZrB\(_2\) particles, the epoxy composites filled with spherical particles have higher effective thermal conductivities than composites reinforced by rectangular particles, but when the volume fractions of particles are beyond 20 vol%, epoxy composites filled with spherical particles have higher effective thermal conductivities than composites reinforced by rectangular particles. This result indicates that the effects of length-width ratios of fillers are monotonically increasing. When the length-width ratio becomes large, the particles with such length-width ratio can be considered as the short fibers. This shape contributes to improving thermal conductive performances of epoxy composites by the effects of both volume fractions of high thermal conductivity fillers and the orientations which form thermal conductive paths. With the length-width ratio increasing, the effect of particle orientation becomes significant. Once the shape of ZrB\(_2\) particles is rectangular instead of spherical, it becomes important to discuss the thermal conductivities of composites which could be affected by the orientations

\[\text{Figure 4: Investigation of the influence from the dependence on the shape and the length-width ratio of rectangular particles.}\]

\[\text{Figure 5: Comparison of the different length-width ratios of rectangular particles.}\]

\[\text{Figure 6: Investigation of the influence from the dependence on particle orientation.}\]
of particles. In the discussion for particle orientation dependence, epoxy composites filled with ZrB₂ particles of a length-width ratio of 2.0 were chosen due to their best thermal conductivity among other composites filled with rectangular particles, and the results are shown in Figure 6. According to Figure 6, the vertical type composites have the best effective thermal conductivity and meanwhile the parallel type composites have the poorest effective thermal conductivity in these comparisons. The results of slant type composites and original ideal condition type composites show almost no difference up to about 25 vol%. Because the circular fillers are beneficial to conduct heat through all directions, including vertical, parallel, and slant directions, it is inferred that the comprehensive thermal conductive effect of circular and slant rectangular fillers is similar at low volume fraction. The differences in thermal conductivity caused by the orientations of particles become more obvious at high volume fraction. In order to clarify the effect of the orientation of high-aspect-ratio particles, the vector diagrams of thermal flux are shown in Figure 7A–C. According to these Figures, it is observed that the thermal flow is directed by ZrB₂ particles; thus, the vertical type composites conduct heat more effectively, and yet the parallel type composites block the thermal flow causing the differences in thermal conductivity of composites. With the volume fraction of ZrB₂ particles increasing, the effects of orientations of particles are magnified. At high volume fraction, inorganic fillers could tend to be agglomerated and it could influence the effective thermal conductivity of composites. In FEM, the distribution of ZrB₂ particles has little influence on some thermal conductivities of epoxy composites due to the ideal models. However, the distribution still affects the efficiency of thermal diffusion in a certain direction. For the investigation of particle arrangement dependence, the results are shown in Figure 8. The variation of thermal conductive performances due to different distribution is characterized by y-axis thermal conductivities of epoxy composites. It is clear that composites with good dispersion have better effective thermal conductivity than ones with agglomerate particles, and as the volume fraction of particles increases, this difference in thermal conductivity becomes larger.

### 4 Conclusions

The effective thermal conductivities of epoxy composites reinforced by multi-status ZrB₂ particles from 5 vol% to 50 vol% were investigated by FEM and the factors affecting the thermal conductivity of the whole composites were discovered. The thermal conductivity performance of composites was improved effectively by filling ZrB₂ particles and specifically epoxy composites filled with 50 vol% ZrB₂ particles had 12.05 times the thermal conductivity of epoxy resin. At the same volume fraction, the variation of the number of ZrB₂ particles in epoxy matrix does not affect the thermal conductivities of composites due to the dimensionless unit in FEM.

As a result of the comparisons of different shapes of ZrB₂ particles, the composites filled with spherical particles have higher effective thermal conductivity than composites...
filled with rectangular particles at 20 vol%, yet rectangular particles have a better effect in improvement of the thermal conductivities beyond 20 vol%. The effects of rectangular fillers improving effective thermal conductivities of composites are obvious and monotonically increasing. The epoxy composites filled with particles of a length-width ratio of 2.0 obtained the highest thermal conductivities among other length-width ratio rectangular particles. In the discussion of orientations of particles, the thermal flux in composites was directed by ZrB₂ particles. Meanwhile, the vertical type particles lead the thermal flux more effectively than parallel type particles; thus, the vertical type particles improve effective thermal conductivity more remarkably. For the investigation of the arrangement of particles, composites reinforced by homogeneous dispersion have higher thermal conductivity than agglomerate dispersion.

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References