



Article

Exploring the Thermophysical Properties of the Thermal Conductivity of Pigmented Polymer Matrix Composites with Barium Titanate: A Comparative Numerical and Experimental Study

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Abstract: This research paper focuses on investigating the thermal conductivity behavior of polymer matrix composite materials, specifically those composed of PSU and BaTiO₃, both experimentally and numerically. The thermal conductivity of composites has been studied using a variety of theoretical and semi-empirical methods. However, in cases where the filler concentration is minimal, these models provide a superior estimate. To numerically resolve the thermal heat transfer for an elementary cell, the finite element method is employed in this study. The impact of contact resistance, barium titanate percentage, and quenching temperature on the composite's effective thermal conductivity and dynamic behavior is given consideration. The results demonstrate that the suggested numerical model is in good agreement with experimental measurements as well as Hatta–Taya and Hashin–Shtrikman's analytical models. The results provide significant insight into the thermal conductivity behavior of composites, which can inform the development of more effective thermal management solutions for composite materials. Effective thermal management is critical for the successful application of polymer matrix composite materials in various engineering applications. Thermal conductivity is a key factor in thermal management and is influenced by factors such as the concentration of filler particles, their shape, size, and distribution, and the matrix material's properties.

Keywords: finite elements; composite materials; thermal conductivity; barium titanate; polymer



Citation: Belhaouzi, A.; Laaouidi, H.; Zyade, S.; Raji, Y.; Halimi, Y.; Tahiri, M. Exploring the Thermophysical Properties of the Thermal Conductivity of Pigmented Polymer Matrix Composites with Barium Titanate: A Comparative Numerical and Experimental Study. *J. Compos. Sci.* **2023**, *7*, 220. <https://doi.org/10.3390/jcs7060220>

Academic Editor: Francesco Tornabene

Received: 7 April 2023

Revised: 11 May 2023

Accepted: 22 May 2023

Published: 27 May 2023



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1. Introduction

The field of materials science has witnessed significant growth in recent years, driven by the discovery of new materials with better properties and characteristics. Among these materials, composite materials have emerged as a promising group with great potential for various technical applications [1,2]. Numerous studies have investigated the mechanical, chemical, and physical properties of composite materials, including those reinforced with carbon nanotubes, woven composites, thermoplastic polymer-based composites, natural fiber-based composites, and elastomers modified with multi-walled carbon nanotubes [3–6].

The unique composition and properties of composite materials make them increasingly popular in various industrial sectors, such as aerospace, automotive, building, and chemical industries [1,2]. However, to design composite materials for specific applications,

a thorough understanding of their thermal behavior is essential. Effective thermal conductivity is a crucial thermophysical characteristic used to measure the thermal behavior of composite materials [7], which is significantly influenced by the materials' composition and structure [8]. Various theoretical and semi-empirical methods have been developed to determine the thermal conductivity of composite materials [9], but modeling heat transfer in heterogeneous media remains a challenging task. Several studies have attempted to create models that closely approximate actual experimental findings [10,11]. To better understand the connection between material structure and thermal conductivity, investigating the effects of each variable individually is essential [12]. Obtaining accurate experimental measurements of thermal conductivity is often challenging, particularly for anisotropic materials, and manipulating the composition and structure of samples is extremely difficult [13]. Therefore, various numerical and analytical models have been proposed in the literature to predict the thermal conductivity of composite materials. For example, Hatta and Taya [14] developed a steady-state model based on the analogy with the mechanical properties of materials to study the equivalent thermal conductivity of composite materials. Fang et al. [15] used the multiple-relaxation-time Lattice Boltzmann method to numerically predict the thermal conductivity of needled C/C-SiC composite materials, and their results were validated using experimental data from the hot disk thermal constants analyzer. Determining the thermal conductivity of a material is crucial not only for optimizing processing conditions but also for understanding the heat transfer within the material in practical applications. Designing composites for various applications requires a thorough understanding of thermal conduction, which can be developed by formulating micromechanical models to accurately predict the thermal conductivity of multiphase composites. Numerical models need to be validated through experimental and theoretical work. Various theoretical and empirical models have been proposed to analyze the conduction equation response of heterogeneous materials. For more complex structures, it is beneficial to study the relationship between the structure of a material and its thermal conductivity.

In another study, Cruz [16] presented a numerical analysis of the effective thermal conductivity of unidirectional fibrous composite materials with an interfacial thermal resistance, which was verified using analytical treatments from the literature. Matt and Cruz [17] demonstrated a numerical approach based on an isoparametric finite element discretization of second order to determine the effective thermal conductivity of composite materials with interfacial and three-dimensional microstructures' thermal resistance. Their findings showed that the interfacial thermal resistance magnitude significantly affects the effective conductivity of composite materials, which was verified against analytical results for a simple cubic array of spheres. Additionally, the Hashin and Shtrikman [18] model provides a unique size distribution and higher thermal conductivity for dopants than matrix materials, which are arranged in a shell surrounding the orbits of dopants. Despite significant progress in developing models that closely approximate actual experimental findings, accurately predicting the thermal behavior of composite materials remains a challenging task. Improving the thermal conductivity (TC, hereafter) of polymers by incorporating organic or inorganic micro/nanoparticles having higher TC is now becoming more important in the production of advanced polymer composites. The potential applications of these composites comprise electronic packaging and encapsulations, satellite devices, and areas where high heat dissipation, small thermal expansion, and lightweight are required. It is known that the TC of polymers is generally very poor and has traditionally been improved by adding fillers (dispersed phase), such as graphite, carbon black, carbon fibers, and high TC ceramic or metal particles, into the matrix materials (continuous phase). It is clear that polymer composites with highly conductive fillers have advantages because of their ease of processing, lower cost, and corrosion resistance. Other reasons for the use of fillers can be found in.

Indeed, no theoretical model can accurately predict the thermal conductivity of composite materials, which makes numerical modeling an indispensable means to validate theoretical models. Recently, thanks to the rapid development of computer science and com-

putational techniques, most of the research has focused on numerical modeling via some numerical models that have been used to model the thermal conductivity of composite materials. The modeling of the thermal behavior of a composite consisting of two distinct phases (a discontinuous phase dispersed in a continuous phase) is the subject of much research, as evidenced by Maxwell's work on heat transfer in heterogeneous media. Based on the previous work [19,20] on the development of the composite material, we continue the study with the objective of investigating the relationship between the structure of the composite material and thermal conductivity. For this purpose, a numerical model based on the finite element method will be developed using the COMSOL Multiphysics software. The main advantage of COMSOL Multiphysics is its ability to simulate multiphysical systems in a fast and efficient way based on the finite element method, allowing us to deal with many problems thanks to its integrated library of very varied physical equations unlike conventional simulation software. The aim of our previous study [21] was to predict the thermal and mechanical properties of polystyrene (PS) polymer composites reinforced with nanoparticles of different sizes and volume fractions, compare numerical, analytical, and experimental results for effective thermal conductivity and mechanical properties, and create flowcharts for thermomechanical simulations process.

The objective of this study is to develop a numerical model for predicting the effective thermal conductivity of composite materials and investigate the impact of filler size on thermal conductivity. The model will be compared with experimental and theoretical results to verify its accuracy. The composite material will be synthesized by combining barium titanate with polymer, and the experimental procedure and materials used will be elaborated in detail. Previous research has indicated an improvement in the performance of the composite material created through this method [19,20]. This paper is divided into the four following sections: (1) an explanation of the experimental procedure and materials, (2) the development of a numerical model for predicting thermal conductivity, (3) a comparison of numerical results with experimental and theoretical findings, and (4) an examination of the effect of filler size on thermal conductivity.

2. Materials and Methods

2.1. Materials

In this investigation, the PSU/barium titanate composite was formulated using a pigmented PSU polymer, specifically the 2010 Ultrason S polysulfone from BASF located in Ludwigs-hafen, Germany. This high-performance polymer is extensively utilized in the production of power supply system components for contemporary civil aircraft. The polymer has a melt flow value of 7 g/10 min at 343 °C, a glass transition temperature of approximately 183 °C, and a polydispersity index of 1.9. The PSU polymer exhibits a thermal conductivity of 0.27 W/m·K.

Barium titanate has been demonstrated to be an effective additive for enhancing thermal and mechanical properties of PSU [22]. This ceramic material is widely recognized for its exceptional piezoelectric, ferroelectric, and dielectric properties. When incorporated into PSU as a pigment, barium titanate particles serve as reinforcement agents, elevating the stiffness and strength of the composite material. Additionally, the inclusion of barium titanate can enhance the thermal conductivity of the PSU composite, resulting in superior heat dissipation capabilities.

2.2. Sample Preparation

The preparation of thermally expanded graphite involved the use of 100 mL of concentrated sulfuric acid, which was continuously stirred for 3 h. Subsequently, 3 g of potassium permanganate and 5 g of graphite powder were added to the mixture, and the resulting mixture was vigorously stirred to ensure optimal exfoliation of the graphite. The oxidation process was allowed to continue for approximately 20 min, following which the solution was filtered and repeatedly rinsed with distilled water until the pH of the chemical approached that of natural water. Graphite flakes are characterized by their crystal structure,

which consists of layered planes of carbon atoms arranged in a hexagonal pattern. This structure confers unique physical and chemical properties to graphite. Using intercalation technology, we were able to produce thermally expanded graphite that exhibited enhanced properties, including increased thermal and electrical conductivity, improved mechanical strength, and greater surface area.

The composite material was fabricated using a solution casting technique that allowed for the incorporation of high concentrations of fillers. A polysulfone solution with barium titanate (Ba-TiO_3) as the filler was used and the polymer concentration was set at 20–40%. The mixture was manually stirred for 24 h using a magnetic stirrer before the filler was added. To ensure the homogeneity of the polymer–filler mixture, a head agitator was used to vigorously stir the solution. The concentration of fillers was varied in increments of 10% between 30–70% to achieve the desired properties.

2.3. Characterization

In this research, we employed a periodic measurement technique to investigate the thermo-physical properties of a composite material. This method involves subjecting the material to thermal excitation and measuring its temperature response. This non-destructive technique enables simultaneous determination of the material's thermal conductivity and diffusivity with high accuracy at room temperature [23,24]. The measurement sample used in this study had dimensions of $40 \times 40 \times 3 \text{ mm}^3$ and was positioned between two metal plates, as shown in Figure 1. The assembly was then placed in a secondary vacuum compartment, which was sealed to minimize heat loss. To generate a temperature gradient, a Peltier block was periodically used to excite the input plate's temperature. This gradient drives a heat flow, which is measured and utilized to determine the material's thermal conductivity and diffusivity.

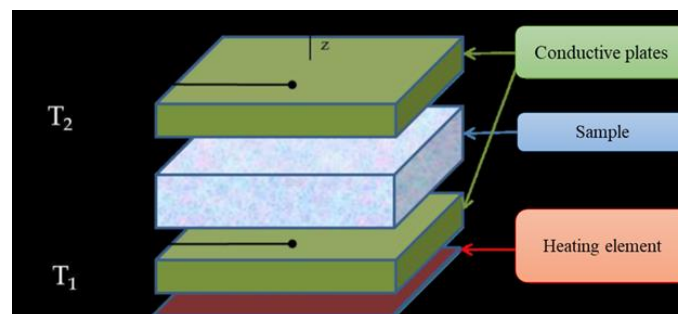


Figure 1. Schematic diagram of the measurement method [25].

3. Thermal Conductivity Models

In the heat transfer field, precise modeling of effective thermal conductivity for heterogeneous or composite materials is crucial for numerous applications. The intricate structure and composition of these materials can influence their effective thermal conductivity [9]. The thermal conductivity of composites has been the subject of extensive research, and several theoretical and empirical correlations have been established for predicting this parameter. Many analytical models of thermal conductivity are used and compared with experimental data. In addition, several references have been given to obtain more details about the types of models, their orders, and their application areas. Table 1 illustrates the primary analytical methods utilized for predicting thermal conductivity.

Table 1. List of frequently used classical theoretical models [26].

Models	Equation	Description
Maxwell	$\lambda_e = \lambda_m \times \frac{2\lambda_m + \lambda_p - 2v_p(\lambda_m - \lambda_p)}{2\lambda_m + \lambda_p + v_p(\lambda_m - \lambda_p)}$	1- Valid for spheres dispersed in a matrix 2- Not valid at finite particle concentrations 3- Interaction between particles is not taken into account
Bruggeman	$1 - v_p = \frac{\lambda_e - \lambda_p}{\lambda_m - \lambda_p} \left(\frac{\lambda_m}{\lambda_e} \right)^{\left(\frac{1}{1+x} \right)}$	1- d = 3 for spherical loads 2- d = 2 for cylindrical loads
Hatta and Taya	$\lambda_e = \lambda_m \left[1 - v_p \frac{2\lambda_m + \lambda_p - 2v_p(\lambda_m - \lambda_p)(\lambda_p - \lambda_m)(2S_{33} + S_{11}) + 3\lambda_m}{3(\lambda_p - \lambda_m)^2(1 - v_p)S_{11}S_{33} + \lambda_m(\lambda_p - \lambda_m)R + 3\lambda_m^2} \right]$ with: $R = 3(S_{33} + S_{11}) - v_p(2S_{11} + S_{33})$	1- $S_{11} + S_{22} + S_{33} = 1$ 2- Disk: $S_{11} = S_{22} = 0, S_{33} = 1$ 3- Sphere: $S_{11} = S_{22} = S_{33} = 1/3$ 4- Fibers, rods, or long cylinders: $S_{11} = S_{22} = 1/2, S_{33} = 0$ 5- Randomly oriented short fibers (length L and diameter D) $\begin{cases} S_{11} = \frac{\alpha}{2(\alpha^2 - 1)^2} \left[\alpha(\alpha^2 - 1)^{\frac{1}{2}} - \cosh^{-1} \alpha \right] \\ S_{33} = 1 - 2S_{11} \\ \alpha = \frac{L}{D} \end{cases}$ 6- Spheres randomly dispersed in a continuous matrix, $R = 2 - v_p$
Hashin and Shtrikman	$\lambda_e = \lambda_p \left(1 + \frac{v_p}{\frac{1-v_p}{3} + \frac{\lambda_p}{\lambda_p - \lambda_m}} \right)$	For spherical loads

4. Problem Formulation and Boundary Conditions

In previous studies, many models for the dependence of the thermal conductivity of particle-reinforced composites on filler content have been assumed. Several researchers attempted to validate their experimental and numerical results using these different models. The finite element method (FEM) is based on decomposing a domain into a finite number of subdomains (elements) for which an approximate solution to the system is created by applying the remainder method. Using the COMSOL finite element software, heat transfer occurs through conduction in the composite. For thermal analysis, a 2D circular lattice physical model is used to simulate the microstructure of the composite. In this study, we aimed to investigate the effective thermal conductivity of a PSU/BaTiO₃ composite material by considering the spatial arrangement of the particles within the matrix. To achieve this, we used a two-dimensional numerical simulation based on the finite element method to solve the steady-state conductive heat transfer problem. Our analysis shows that convection and radiation heat exchanges are negligible and can be ignored. The composite material under consideration consisted of a BaTiO₃ particle at the center of a square-shaped PSU matrix with a dimension of $L = 2 \mu\text{m}$. The radius r of the particle varied between 0.3 and 0.8 μm . We examined polymer matrices with different particle volume ratios. To determine the temperature distribution within the composite material, we solved the Laplace equation (Equation (1)) using a finite element formulation. We imposed specific boundary conditions to accurately simulate the heat transfer process.

- Two faces perpendicular to the direction of heat flow: isothermal with temperatures $T_1 = 298 \text{ K}$ and $T_2 = 323 \text{ K}$.
- Faces parallel to the direction of heat flow: adiabatic.
- Transfers by radiation and convection: negligible.
- Thermal contact resistance between matrices and loads: negligible.
- Dispersion of spherical particles in matrices: homogeneous.

The results of our study indicate that the effective conductivity of the PSU/BaTiO₃ composite material is impacted by the spatial distribution of particles within the matrix.

This can be attributed to variations in thermal resistance caused by the presence of particles. Our study contributes to the understanding of composite material behavior and can aid in the development of materials with desired thermal conductivity properties as shown in Figure 2.

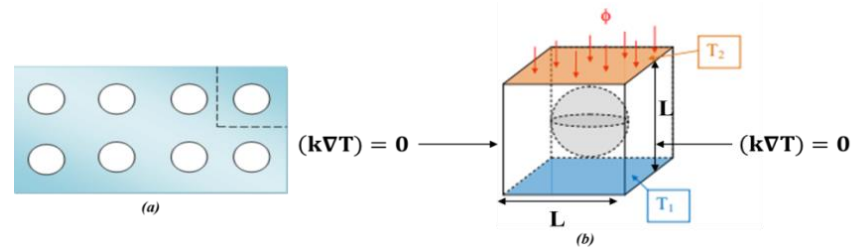


Figure 2. (a) Distribution of the PSU/BaTiO₃ composites and (b) the structural characteristics of the unit cell.

The phenomenon of conduction is represented by Equation (1), which is responsible for heat transfer in a substance as follows:

$$0 = \rho c p \times \frac{\partial T}{\partial t} - \nabla(k \nabla T). \quad (1)$$

Integrating Equation (1) over the boundaries of the elementary cell of the composite provides a computation of the heat transfer from the hot surface to the cold surface.

$$Q = \int_A k \frac{\partial T}{\partial z} dx dy, \quad (2)$$

in which the variables are defined as follows:

- Q is the heat flux; k is the thermal conductivity of the composite;
- $\frac{\partial T}{\partial z}$ is the temperature variation along the z direction;
- x and y represent the exchange surface.

To calculate the effective thermal conductivity, we utilized the flux value obtained from the COMSOL software and substituted it into Equation (3). With the flux known, we can calculate the effective thermal conductivity using Equation (3), which is expressed as follows:

$$k = \frac{Q}{A} \frac{L_z}{(T_1 - T_2)}, \quad (3)$$

where the exchange area $A = x \times y$ (m²); z is calculated; $\Delta T = T_1 - T_2$ (°K) is the temperature variation. The effective thermal conductivity of the composite is determined via the following formula [27]:

$$k = \frac{(Q \times Z)}{(x \times y \times (T_1 - T_2))}. \quad (4)$$

5. Results and Discussion

A complete thermal study is performed on the PSU/BaTiO₃ composite. For this purpose, the parameters used in the calculations are as follows:

- Thermal conductivity of BaTiO₃ is $k_{ch} = k(\text{BaTiO}_3) = 2.7\text{--}4.5$ W/m·K;
- Particle radius of BaTiO₃ is $r = 0.25\text{--}0.6$ μm;
- Thermal conductivity of PSU $k_m = k(\text{PSU}) = 0.17\text{--}0.22$ W/m·K;
- Pigment fraction is $\varphi = 3\%$.

5.1. Temperature Distribution in the Composite Material

After imposing boundary conditions, we analyzed the temperature distribution within a PSU/BaTiO₃ composite elementary cell. As shown in Figure 3, the propagation of heat flow from the hot boundary to the cold boundary illustrated that the composite's heat

transfer characteristics were affected by the boundary conditions. We found that the temperature within the composite's PSU matrix region decreased, while the temperature in the BaTiO₃ region remained relatively constant at around 310 K. These findings suggest that the BaTiO₃ phase acted as a thermal insulator, thereby hindering heat transfer.

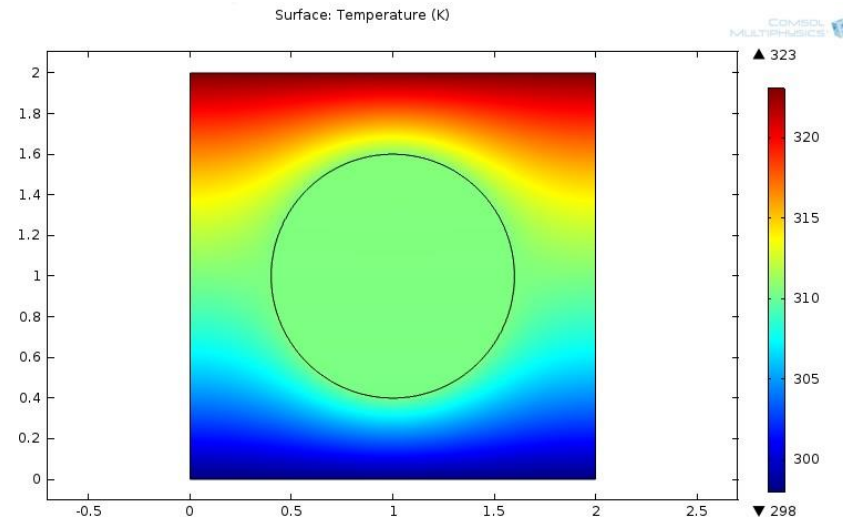


Figure 3. The temperature distribution in the PSU/BaTiO₃ composite.

To validate our findings, we analyzed the temperature distribution at three different positions ($X = 0.4$, $X = 0.8$, and $X = 1.4$) within the PSU/BaTiO₃ composite and plotted temperature variation curves against cell size, as shown in Figure 4. These curves confirmed that the BaTiO₃ phase exhibited superior thermal insulation properties compared to the PSU matrix, as observed in the temperature differences. This thermal insulation performance of BaTiO₃ is attributed to its low thermal conductivity and high specific heat capacity.

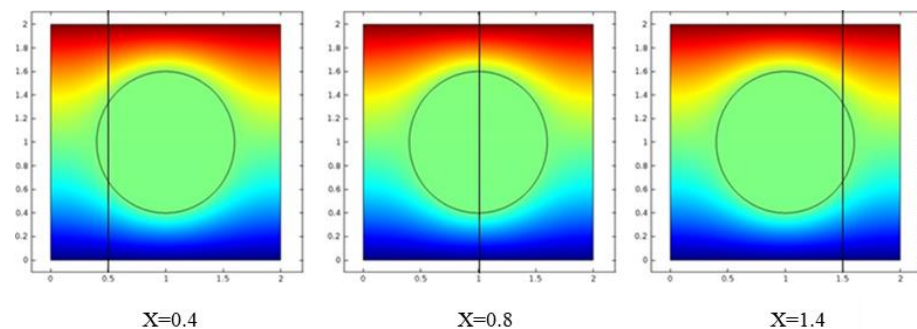


Figure 4. Schematic of a PSU/BaTiO₃ composite elemental cell with the 3 X positions.

This present study investigated the temperature distribution in a PSU/BaTiO₃ composite elementary cell, as presented in Figure 5. The results indicate that the temperature in the matrix domain decreased linearly with increasing cell dimension L , indicating the existence of a low thermal gradient within the composite. This behavior can be attributed to the low thermal conductivity of polysulfone. In the region where BaTiO₃ is present, the temperature remained constant, regardless of the Y position, suggesting that the position has minimal impact on the temperature. These results are consistent with previous research indicating that the low thermal conductivity of polymer composites is due to the presence of non-conductive polymer matrices, such as polysulfone. The use of high dielectric constant fillers, such as BaTiO₃, can result in a low thermal gradient due to the presence of interfacial layers between the filler and matrix.

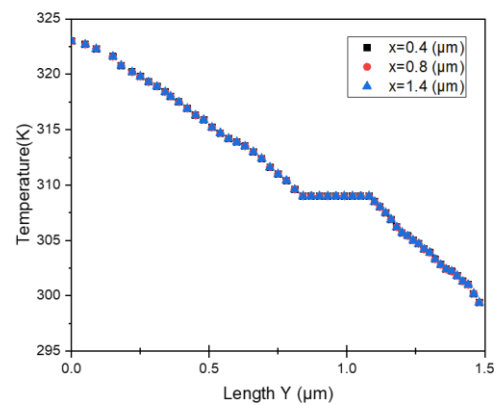


Figure 5. Temperature distribution for different positions X in the load as a function of length Y.

Figure 6 displays a comparison between several analytical models and the numerical results, showing the evolution of the effective thermal conductivity of the composite with varying loading rates.

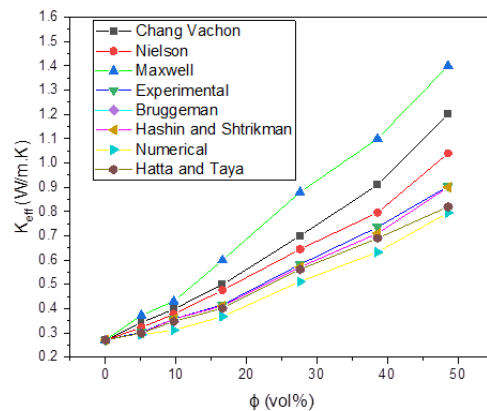


Figure 6. Comparison of the PSU/BaTiO₃ composite's thermal conductivity with theoretical models and experimental results.

Figure 6 compares the thermal conductivity of the PSU/BaTiO₃ composite to theoretical models and experimental results. The results show that for filler concentrations below 10%, all prediction algorithms provide a reliable estimation that matches well with the numerical findings. For concentrations between 10% and 30%, the models of Hata and Taya, Hashin and Shtrikman, and Bruggeman show a good match with the numerical results. However, for concentrations above 30%, the Bruggeman model provides the most accurate estimation of the composite's thermal conductivity compared to other predictions. At intermediate concentrations, the Bruggeman model is most applicable. It also follows that one should expect the complex permittivity and complex permeability to be equally well constrained by the Bergmann boundaries since the boundaries are derived from the same microstructure, and, therefore, their characteristic geometric functions are not independent.

5.2. Effect of Filler Volume Fraction and Thermal Contact Resistance

Figure 7 shows how the thermal conductivity of the composite changes when normalized to the matrix value for different contact resistances ranging from 1.10^{-5} to 1.10^{-1} [28].

Figure 7 shows that the effect of contact resistance on thermal conductivity is negligible when the filler content is below 20%. This is because there is no interaction between the fillers at concentrations below 20%. The influence of contact resistance becomes evident at 20% filler content. A significant change in thermal conductivity can be observed for a high contact resistance (above $10^{-2} \text{ m}^2 \cdot \text{K} \cdot \text{W}^{-1}$).

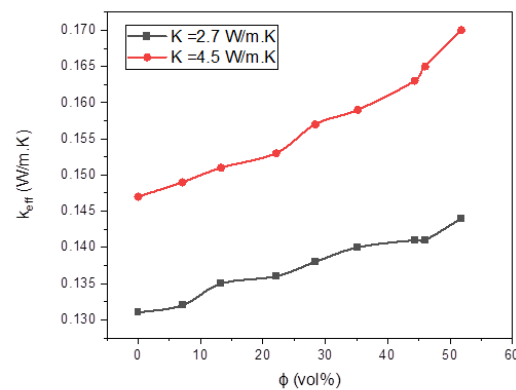


Figure 7. Influence of contact resistance on thermal conductivity.

5.3. The Effect of the Filler Thermal Conductivity

Figure 8 illustrates the correlation between the thermal conductivity of the filler and the effective thermal conductivity of the composite as a function of the filler concentration (k_{ch}). As k_{ch} is significantly larger than k_m , it is apparent that the effective thermal conductivity of the composite increases with increasing k_{ch} .

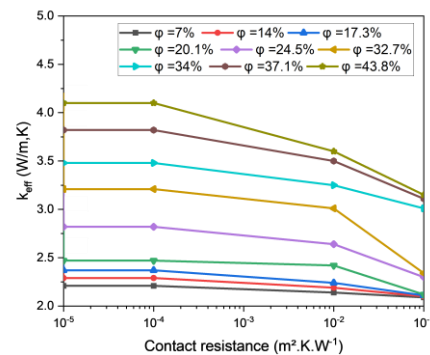


Figure 8. Effect of the thermal conductivity of the filler on the effective thermal conductivity of the composite.

5.4. Effect of Particle Size

Many authors have studied the effect of particle size on the thermal properties of composites. In order to study the influence of particle size of inclusions on the effective thermal conductivity, spherical inclusions with different thermal conductivity k_2 were arranged in the continuous thermal conductivity phase k_1 . Despite discrepancies between numerical simulations and experimental results at high filler volume contents, we investigated the effect of particle size on the thermal conductivity of composites using a 2D physical model; Figure 9 depicts the relationship between the effective thermal conductivity of composites and the size of BaTiO₃ particles at room temperature. The results show that the effective thermal conductivity of the composites increases with decreasing particle size. This behavior is due to the increase in the surface area-to-volume ratio of the particles, which leads to enhanced phonon scattering and hence improved thermal transport properties. The findings suggest that the optimization of the particle size can play a significant role in achieving high thermal conductivity in polymer composites. The thermal conductivity of reinforced composites can be impacted considerably by the size of the conductive particles. Smaller particles can create more efficient paths for the flow of heat through the material, resulting in increased thermal conductivity. Although decreasing the particle size results in an increase in the surface-to-volume ratio, this can lead to higher thermal resistance due to increased heat diffusion at the surface. In addition, the effect of particle size on thermal conductivity depends on the type of conductive material, the matrix, and the distribution of particles in the composite. Overall, the optimal particle size to maximize

thermal conductivity in a reinforced composite would depend on the balance between these factors.

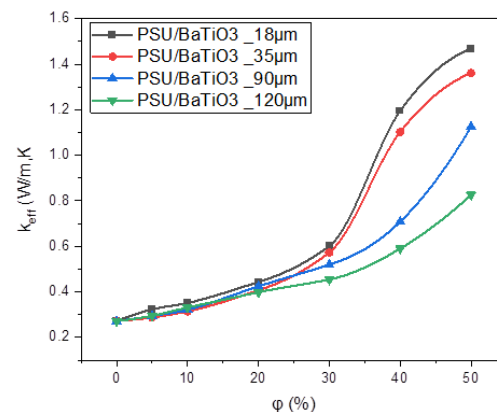


Figure 9. The thermal conductivity and volume fraction for various sizes.

Figure 9 demonstrates that the effective thermal conductivity of composites loaded with small $BaTiO_3$ particles is higher than those loaded with larger particles. It is noteworthy that the particle size effect becomes prominent for concentrations above 20%, which could be attributed to the contact between the matrix and fillers, where larger particles result in less perfect contact.

6. Conclusions

The thermal conductivity of a composite material composed of barium titanate ($BaTiO_3$) spheres filled into a polysulfide matrix was studied using finite element analysis. The effect of filler size and volume fraction on the effective thermal conductivity of the composite material was examined and compared with several theoretical models. The results showed that the Hatta–Taya and Hashin–Shtrikman models accurately predicted thermal conductivity values for filler volume fractions up to 30%. Furthermore, this study highlighted the impact of filler size and contact resistance on thermal conductivity, revealing that the thermal conductivity of the composite material could be improved by increasing the filler volume fraction and reducing the size of the filler particles.

Indeed, this study sheds light on the thermal conductivity of polymer composites, providing crucial knowledge for the design and optimization of their performance. Further research in this field could focus on developing novel models to accurately predict the thermal conductivity of these materials and exploring the impact of additional parameters, such as temperature and pressure, on their thermal behavior. These efforts would lead to the development of more efficient and effective composite materials for various applications in industries ranging from electronics to energy.

Author Contributions: Conceptualization, A.B. and S.Z.; Methodology, A.B.; Validation, S.Z. and Y.R.; Formal analysis, A.B.; Resources, H.L.; Data curation, A.B., S.Z. and M.T.; Writing-preparation of original version, A.B.; Software, A.B., Y.H. and Y.R.; Writing-reviewing and editing, H.L., S.Z., Y.R., Y.H. and M.T.; Supervision, M.T. and S.Z. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Data Availability Statement: Not applicable.

Conflicts of Interest: All authors certify that they have no affiliation or involvement with any organization or entity with a financial interest (such as honoraria, educational grants, participation in speakers' bureaus, membership, employment, consulting, stock ownership or other participation, and expert testimony or patent licensing agreements) or a nonfinancial interest (such as personal or professional relationships, affiliations, knowledge, or beliefs) in the subject matter or materials discussed in this manuscript.

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