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INFLUENCE OF MINERAL FILLER ON THE THERMAL CONDUCTIVITY OF POLYMER COMPOSITES

The study focuses on the development of polymer composites based on the Latex 2012 aqueous dispersion with mineral fillers of volcanic (andesite) and technogenic (fly ash from Burshtyn TPP and Kurakhove TPP) origin, aimed at achieving optimal thermal insulation properties. The main problem addressed was determining the influence of the type, concentration, and combination of fillers on the thermal conductivity of composites. High thermal conductivity of polymeric materials significantly limits their application in thermal insulation systems, making it crucial to investigate the mechanisms of interfacial interactions between fillers and the matrix for creating effective compositions.

The sizes of filler crystallites were determined using the Scherrer method: for andesite – 110 nm, fly ash B – 100.4 nm, and fly ash K – 113 nm. These data indicate the fillers' ability to affect phonon scattering in the material, reducing overall thermal conductivity. The thermal conductivity of the fillers is as follows: fly ash B – 0.2072 W/m·K, fly ash K – 0.2241 W/m·K, and andesite – 0.2118 W/m·K. Fly ash B demonstrated the best results due to its low thermal conductivity and high surface energy, which contributes to better interaction with the polymer matrix.

An analysis of the dependence of composite thermal conductivity on temperature and filler concentration showed that increasing filler concentration increases thermal conductivity due to the formation of thermal bridges between particles. However, combining different fillers in optimal proportions can mitigate this effect. Based on the Nielsen model, the composite compositions were optimized to achieve minimum thermal conductivity. The best results were obtained for a binary filler system of fly ash B and andesite in a 53:35 mass ratio, providing the lowest effective thermal conductivity of the composite – 0.173 W/m·K. Other successful combinations include fly ash B with fly ash K (60:40 wt. %) and andesite with fly ash K (45:55 wt. %), which also demonstrate significant improvements in thermal insulation properties.

The proposed compositions can be applied in the field of energy-efficient construction, thermal regulation systems, thermal insulation materials for industrial equipment, and other areas requiring low thermal conductivity. The research results are also valuable for developing materials that operate under significant temperature variations (from –125 °C to +100 °C), providing stable thermal insulation properties.

Keywords: polymer composites, composite thermal conductivity, mineral fillers, fly ash, andesite, Nielsen model.

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1. Introduction

Polymer composites with mineral fillers are increasingly utilized in advanced applications due to their ability to balance mechanical strength, thermal insulation, and cost efficiency. Thermal conductivity is a crucial property of these materials, determining their suitability for thermal management systems, building construction, and energy-efficient applications [1]. Integrating volcanic fillers (andesite) and anthropogenic waste, such as fly ash from thermal power plants, into polymer matrices offers a promising approach to enhancing the thermal properties of composites [2]. This approach leverages the unique physicochemical characteristics of these fillers, such as low thermal conductivity, high porosity, and specific structural features, to create materials with specialized thermal insulation capabilities [3, 4].

Among volcanic fillers, andesite stands out for its micro- and nanostructured porosity, high density, and silica-rich composition, contributing to its low thermal conductivity and suitability for thermal insulation applications [5]. Similarly, fly ash, a by-product of coal combustion, combines fine particle size and amorphous content, making it an effective supplementary filler in polymer composites [6].

It is noteworthy that the composition and structure of fly ash vary depending on the type of coal burned and the collection method used. For instance, fly ash from Burshtyn TPP (Ash B) and Kurakhove TPP (Ash K) differ significantly in their silica-to-alumina ratios, iron oxide content, and alkali oxide concentrations, affecting their efficiency as fillers [7].

The thermal behavior of composites is influenced by the properties of individual fillers, their ratios, and their interactions with the polymer matrix [8]. Studies on similar

systems have shown that the inclusion of micro- and nano-structured fillers disrupts heat transfer pathways, thereby reducing thermal conductivity. While volcanic fillers and fly ash have been studied independently, their combined effect within a single composite system remains insufficiently explored. This presents an opportunity to optimize the composition of polymer composites to achieve specific thermal properties, serving applications in energy-efficient construction and thermal management systems.

The primary objective of this research is to analyze the impact of andesite and fly ash fillers on the thermal conductivity of polymer composites based on the Latex 2012 matrix. This involves assessing the influence of filler ratios, nanostructural characteristics, and porosity on the thermal properties of composites. Special attention is given to determining the optimal combination of andesite and fly ash to achieve desirable thermal characteristics for specific applications.

The aim of this research is to establish a comprehensive understanding of the relationship between the composition of polymer composites with volcanic and anthropogenic fillers and their thermal conductivity. The research focuses on identifying optimal filler compositions for both maximum and minimum thermal conductivity, providing a foundation for the development of advanced thermal insulation materials.

2. Materials and Methods

2.1. Materials

The study utilized a polymer matrix based on the aqueous dispersion Latex 2012, which is a copolymer of styrene and butadiene with a solid content of 51 %, an average particle size of 140 nm, and a viscosity of 200 mPa·s. Mineral fillers included andesite from the Khust deposit (Zakarpattia region) and fly ash from the Burshtyn and Kurakhove TPPs (Ukraine). Fly ash from Burshtyn TPP (Ash B) was obtained by the wet collection method and is characterized by low thermal conductivity (0.2072 W/m·K) and a SiO₂:Al₂O₃ ratio of 2.6. Fly ash from Kurakhove TPP (Ash K), obtained by the dry collection method, had a thermal conductivity of 0.2241 W/m·K and a SiO₂:Al₂O₃ ratio of 3.2. Andesite was distinguished by its high specific surface area (11.91 m²/g) and thermal conductivity of 0.2118 W/m·K.

Sample Preparation. Composite samples were prepared using a binary system of polymer binder and filler. The fillers were mechanically activated in a planetary ball mill for 20 minutes to enhance their surface activity and improve interaction with the polymer matrix.

After milling, the fillers were sieved to ensure a uniform particle size distribution of less than 100 μm, providing stable dispersion in the composites.

The polymer-filler mixtures were homogenized, molded, and cured under the following conditions:

1. The homogenized mixture was poured into cylindrical molds with a diameter of 15 mm and left to cure at room temperature for 48 hours.

2. The formed blanks were subjected to heat treatment by gradually heating them to 80 °C and maintaining this temperature for 1 hour to ensure proper structural development.

3. After cooling to room temperature, the samples were cold-pressed to achieve the required density and mechanical integrity, using a pressing pressure of 5–10 MPa.

2.2. Methods

Structural analysis was performed using a DRON-3M diffractometer (Cu Kα radiation, 40 kV, 20 mA). Based on diffraction data from our previous studies [9], the crystallite size of the fillers was calculated using the Scherrer equation [10] to evaluate their nanoscale properties:

$$D = \frac{K \cdot \lambda}{\beta \cdot \cos \theta}, \quad (1)$$

where D – crystallite size (nm), $K=0.9$ – shape factor, $\lambda=1.5406 \text{ \AA}$ – wavelength of the X-ray radiation, β – FWHM (full width at half maximum) in radians, θ – Bragg angle.

This detailed methodology provides a foundation for comparing the thermal properties of polymer composites with the studied fillers (andesite, Ash B, and Ash K), enabling the investigation of their combined effect on thermal conductivity.

The morphology and dispersion of fillers within the polymer matrix were examined using a high-resolution SEM. Specific surface areas of the fillers were measured using the BET method via nitrogen adsorption at –195 °C. These data were derived from our previous studies [11], and this research relies on those results.

Thermal conductivity was measured using the IT-λ-400 thermal conductivity analyzer, operating on the principle of dynamic calorimetry, suitable for temperatures ranging from –100 °C to +100 °C. Sample surfaces were polished to achieve a roughness of $R_z < 0.63 \text{ μm}$ ensuring optimal thermal contact with the measurement plates. A thermal grease was applied to the contact surfaces to minimize thermal resistance. Samples were subjected to a steady heat flow, and the temperature gradient was recorded using a chromel-alumel thermocouple.

Thermal conductivity (λ) was calculated using the formula:

$$\lambda = \frac{h}{P}, \quad (2)$$

where λ – thermal conductivity (W/m·K), h – sample height (m), P – thermal resistance of the sample ((m²·K)/W).

The average temperature was calculated as the arithmetic mean of the temperatures on the upper and lower surfaces of the sample. The method provides an error of no more than ±10 %, which allows for reliable data to be stored for assessing the thermal conductivity of composites in a wide temperature range.

3. Results and Discussion

The structural properties of fillers play a crucial role in determining the thermal conductivity of polymer composites. One of the key structural parameters influencing thermal properties is the crystallite size, which reflects the average size of coherently diffracting domains in the material. Crystallite size is particularly important for materials with micro- and nanostructures, as it directly affects interfacial interactions and phonon scattering mechanisms within the composite matrix.

To evaluate the crystallite size, X-ray diffraction (XRD) analysis was conducted for the selected fillers: andesite, Ash B, and Ash K. Crystallite size was calculated using the Scherrer equation, and the results for two diffraction peaks for each filler are presented in Table 1. The average crystallite size was determined to provide a more representative parameter for comparison.

Table 1
Calculation of crystallite size based on XRD data

Material	2θ, °	FWHM, β, rad	Crystallite size, D, nm	Average crystallite size, nm
Andesite	26.5	0.7	116.59	110.0
	32.0	0.8	103.31	
Ash B	26.0	0.8	111.3	100.4
	30.0	1.0	89.5	
Ash K	25.0	0.7	127.0	113.0
	28.5	0.9	98.6	

As shown in Table 1, clear differences in the crystallite sizes of the studied fillers are observed. Andesite exhibits a relatively large average crystallite size of 110.0 nm, with individual diffraction peaks corresponding to crystallite sizes of 116.6 nm and 103.3 nm. This indicates a consistent, well-ordered crystalline structure in the material, characteristic of volcanic-origin minerals with high silica content.

In contrast, Ash B shows a somewhat smaller average crystallite size of 100.4 nm. The crystallite sizes for the two peaks are 111.3 nm and 89.5 nm, reflecting certain heterogeneity in its structure, likely due to its production process (wet collection) and higher iron oxide content. This heterogeneity may affect its interaction with the polymer matrix.

Ash K, on the other hand, has the largest average crystallite size of 113.0 nm, with individual peak values of 127.0 nm and 98.6 nm. The dry collection method and the higher silica-to-alumina ratio in Ash K contribute to its more homogeneous crystalline structure compared to Ash B.

From the analysis of the obtained data, it can be concluded that andesite, as a volcanic-origin mineral, has a stable crystalline structure, making it a reliable filler for thermal insulation applications. Ash B, with its heterogeneous structure, may induce greater phonon scattering due to its variability in crystallite size, potentially reducing its thermal conductivity. Conversely, Ash K, with its larger and more uniform crystallite size, may offer improved thermal properties due to reduced surface scattering compared to Ash B.

The results obtained provide a foundation for understanding the structural contribution of fillers to the thermal properties of composites. Variations in crystallite size among the fillers emphasize the importance of their origin and processing methods, which must be considered when optimizing composite compositions for specific thermal applications.

A summary table (Table 2) of the studied filler properties offers a comprehensive evaluation for predicting their behavior within composite materials.

Table 2
Physico-mechanical properties of the fillers

Material	Surface area, m ² /g	Average pore diameter, nm	Contact angle, °	Surface energy	Thermal conductivity, W/m-K
Ash B	3.45	2.459	69	51.67	0.2072
Ash K	0.68	2.934	76	39.58	0.2241
Andesite	11.91	2.34	67	53.14	0.2118

Andesite exhibits the highest specific surface area (11.91 m²/g) among the studied fillers, reflecting its well-developed micro- and nanostructure. This characteristic aligns

with its smaller average pore diameter (2.34 nm), which enhances its ability to scatter phonons and reduce thermal conductivity. The surface energy of andesite (53.14 mJ/m²) also indicates strong interactions with the polymer matrix, contributing to improved dispersion and interfacial bonding. These properties correlate well with its average crystallite size of 110.0 nm, indicating a relatively uniform and stable crystalline structure.

In contrast, Ash B shows a significantly lower specific surface area of 3.45 m²/g compared to andesite (approximately 3.5 times less) and a slightly larger average pore diameter (2.459 nm). This suggests that Ash B provides a less effective mechanism for phonon scattering. However, its contact angle (69°) and surface energy (51.67 mJ/m²) indicate good compatibility with the polymer matrix. The thermal conductivity of Ash B (0.2072 W/m-K) is the lowest among the studied fillers, which can be attributed to its moderate crystallite size (100.4 nm) and balanced structural properties. The smaller crystallite size compared to andesite may enhance phonon scattering, further reducing thermal conductivity.

On the other hand, Ash K has the lowest specific surface area (0.68 m²/g) and the largest average pore diameter (2.934 nm). This combination reduces the filler's ability to disrupt heat flow within the polymer matrix, resulting in the highest thermal conductivity (0.2241 W/m-K) among the studied fillers. The contact angle of Ash K (76°) and its relatively low surface energy (39.58 mJ/m²) suggest weaker interactions with the polymer matrix, potentially leading to less effective dispersion. The average crystallite size of Ash K (113.0 nm) aligns with its structural characteristics, indicating a more uniform crystalline structure that may facilitate heat transfer compared to Ash B.

The analysis reveals that andesite and Ash B are more effective fillers for reducing the thermal conductivity of polymer composites due to their lower thermal conductivity and structural properties that promote phonon scattering. The larger surface area and smaller pore diameter of andesite make it a suitable candidate for applications requiring strong interfacial bonding and thermal insulation. Conversely, Ash K, despite its larger crystallite size and uniform structure, exhibits higher thermal conductivity due to its smaller surface area and larger pore diameter, limiting its effectiveness as a thermal insulation filler. These findings underscore the importance of considering both structural and thermal parameters when selecting fillers for polymer composites.

The relationship between crystallite size and properties, summarized in Table 2, highlights the critical role of structural characteristics in determining the overall thermal performance of composites. Further optimization of filler ratios can leverage this knowledge to achieve specific thermal properties tailored to different applications.

The observed differences in the properties of the fillers under study will also influence the thermal behavior of the composites incorporating them. To evaluate the impact of these fillers and their structural characteristics on thermal conductivity, polymer composites with varying filler concentrations were fabricated, and their thermal conductivity was investigated (Fig. 1). This allowed the relationship between filler type, concentration, and the thermal characteristics of the composite to be understood.

As shown in Fig. 1, the overall trend observed on the graph indicates that the thermal conductivity for all composites

does not exceed 3.0 W/m·K within the studied temperature range, highlighting moderate insulation properties for these systems. Most of the composites exhibit relatively stable thermal conductivity values with minor fluctuations, demonstrating consistent heat transfer mechanisms.

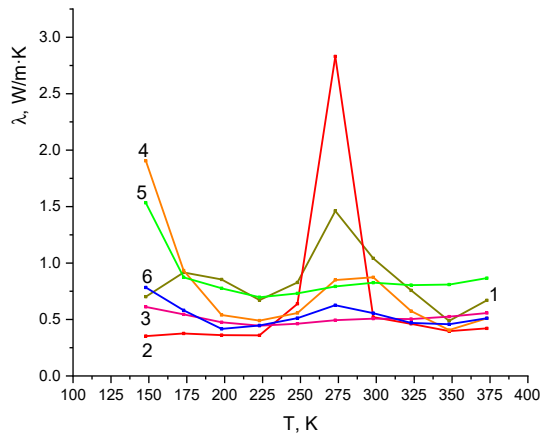


Fig. 1. Dependence of thermal conductivity on temperature for Latex 2012-based systems with different fillers: 1 – Ash B 90 wt. %, 2 – Ash B 65 wt. %, 3 – Ash K 90 wt. %, 4 – Ash K 65 wt. %, 5 – Andesite fines 90 wt. %, 6 – Andesite fines 65 wt. %

A sharp peak is observed for Ash B at 65 wt. % (curve 2), indicating an anomaly likely caused by structural changes or variations in filler dispersion at certain temperatures. Ash B at 90 wt. % (curve 1) shows more stable behavior, while composites with Ash K (curves 3 and 4) exhibit higher thermal conductivity. Composites based on andesite (curves 5 and 6) at 90 wt. % demonstrate the highest thermal conductivity, but a noticeable improvement in stability and a reduction in conductivity are observed at 65 wt. %. These trends underscore the critical role of filler type and concentration in determining the thermal characteristics of composites.

When comparing the influence of filler concentration on thermal conductivity (Fig. 2) at different temperatures (25 °C and 100 °C), the overall trend indicates that thermal conductivity increases with filler concentration. At 25 °C, Ash B (curve 1) consistently demonstrates the lowest thermal conductivity among all fillers, indicating excellent thermal insulation properties. This is followed by Ash K (curve 2) and andesite screenings (curve 3), with andesite showing the highest thermal conductivity, especially at 90 wt. %.

At 100 °C, a similar trend is observed, with Ash B again exhibiting the lowest thermal conductivity, while andesite shows the highest. However, thermal conductivity values increase slightly at higher temperatures for all composites, reflecting enhanced heat transfer due to the temperature's influence on the filler-matrix interface.

Considering the above, it should be noted that Ash B provides the best insulation properties at both temperatures, while higher filler concentrations and elevated temperatures generally result in increased thermal conductivity and reduced insulating performance of the developed composites.

Thus, to further enhance the thermal insulation properties of the composites, it is necessary to optimize the filler composition. By selecting the appropriate combination and proportions of fillers, let's aim to minimize thermal conductivity while maintaining the mechanical integrity and processability of the composite materials.

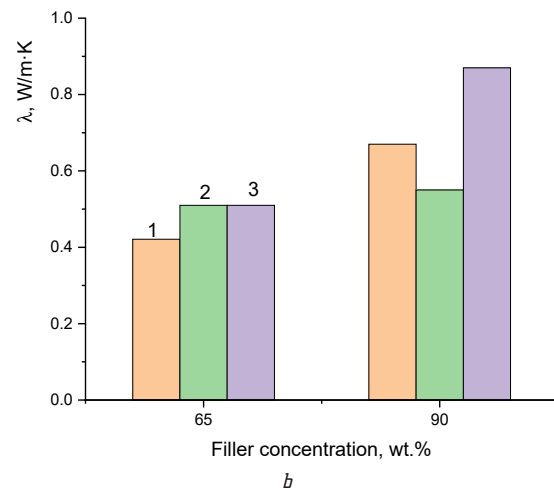
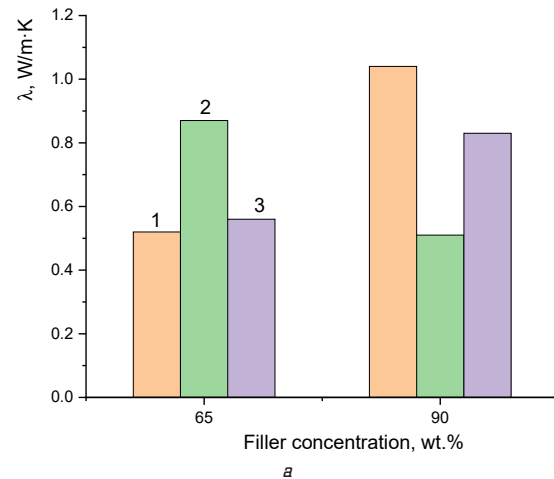


Fig. 2. Dependence of thermal conductivity on filler concentration (at different temperatures) for systems based on Latex 2012 with various fillers: 1 – Ash B, 2 – Ash K, 3 – Andesite screenings; a – 298 K, b – 373 K

Advanced modeling approaches were employed to identify the most effective binary filler combinations. Among the available models, the parallel (rule of mixtures) and series models were considered as basic approaches, but they lack the flexibility to account for the complex interactions and packing effects observed at high filler loadings.

The Nielsen model incorporates the maximum packing fraction of fillers and accounts for their orientation and distribution within the polymer matrix. This makes it particularly suitable for systems with high filler content (up to 90 vol. %), where particle interactions and agglomeration significantly influence thermal conductivity. Alternative models, such as the Maxwell model, are more appropriate for low filler concentrations, while advanced models, such as Hashin-Shtrikman or Agari-Nielsen, require additional parameters not available in this study.

Thus, the Nielsen model [12] was chosen as the most appropriate tool for optimizing the binary filler compositions to achieve the lowest thermal conductivity and the best thermal insulation properties:

$$\frac{\lambda_{CM}}{\lambda_h} = \frac{1 + (A-1)B\phi}{(1 - \psi B\phi)},$$

$$\psi = 1 + \frac{1 + (1 - \phi_{max})\phi}{\phi_{max}^2}, \quad B = \frac{\lambda_f / \lambda_p - 1}{\lambda_f / \lambda_p + A - 1}, \quad (3)$$

where λ_{CM} – effective thermal conductivity of the composite (material with filler); λ_h – thermal conductivity of the matrix (base material without filler); ϕ – volume fraction of the filler in the composite; ψ – coefficient accounting for the effect of the maximum volumetric content of the filler; ϕ_{max} – maximum possible volume fraction of the filler that can be incorporated into the matrix; B – dimensionless coefficient that accounts for the thermal conductivity of the filler and the matrix; λ_f – thermal conductivity of the filler; λ_p – thermal conductivity of the polymer matrix; A – coefficient depends on the geometry and orientation of the filler particles.

The results of the calculations according to this model are presented in Table 3.

Table 3

Optimized binary filler compositions for determining minimum thermal conductivity

Filler 1	Filler 2	Mass fraction of filler 1 (%)	Mass fraction of filler 2 (%)	Effective thermal conductivity (W/m·K)
Ash B	Andesite	53	35	0.173
Ash B	Ash K	56	34	0.176
Andesite	Ash K	44	46	0.179
Ash B	Andesite	36	52	0.179
Ash B	Ash K	60	30	0.180

As shown in Table 3, among the studied mixtures, the best thermal insulation properties with the lowest effective thermal conductivity (0.1732 W/m·K) are demonstrated by the combination of Ash B and andesite in a mass fraction ratio of 53:35, respectively. Other effective combinations include Ash B and Ash K (ratio 56:34) and andesite with Ash K (ratio 44:46), which also provide relatively low thermal conductivity. These results emphasize the importance of balancing filler types and concentrations to enhance their individual properties and achieve a synergistic effect for optimal thermal insulation performance.

Despite the obtained results, the implementation of the developed materials requires additional research focused on studying their durability and optimizing production processes to ensure economic feasibility.

The conditions of martial law in Ukraine have partially impacted the research, particularly due to limited access to laboratory equipment, difficulties in obtaining consumables, and the need to adapt to remote work. Despite these challenges, the main experiments were successfully completed, and the research findings can serve as a foundation for further scientific and practical developments.

The prospects for further research include a detailed study of the durability of the developed composites under prolonged exposure to temperature fluctuations, mechanical loads, and aggressive environments. Special attention should be given to exploring the possibilities of using other types of fillers and polymer matrices to expand the functional properties of the materials, which will be the subject of our future studies.

4. Conclusions

The study established that the combination of volcanic and anthropogenic fillers in polymer composites based on

Latex 2012 significantly alters the thermal properties of the materials. The main findings are as follows:

- The crystallite size calculated using the Scherrer equation revealed that andesite has the largest average crystallite size (110 nm), while Ash B exhibits a smaller average size (100.4 nm), which promotes more effective phonon scattering and reduces thermal conductivity. Ash K has the largest average crystallite size (113 nm), which may increase thermal conductivity due to fewer scattering interfaces.
- Andesite and Ash B demonstrated the lowest thermal conductivity (0.2118 W/m·K and 0.2072 W/m·K, respectively), making them effective fillers for creating thermal insulation materials. Ash K exhibited the highest thermal conductivity (0.2241 W/m·K) due to its larger average pore and crystallite sizes.
- Graphical dependencies showed that the thermal conductivity of composites increases with higher filler concentrations and depends on the filler type and properties. Ash B exhibited the lowest thermal conductivity among all composites, confirming its effectiveness as a thermal insulation filler.
- Based on the Nielsen model, optimal filler ratios were calculated to achieve minimal thermal conductivity. The best results were obtained for the combination of Ash B and andesite in a ratio of 53:35 mass. %, achieving the lowest effective thermal conductivity (0.173 W/m·K). Other effective combinations included Ash B with Ash K and andesite with Ash K.

Conflict of interest

The author declares that she has no conflict of interest regarding this study, including financial, personal, authorship or other nature, which could affect the study and its results presented in this article.

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The study was conducted without financial support.

Data availability

There are no related data in the manuscript.

Use of artificial intelligence

The author confirms that she did not use artificial intelligence technologies in creating the presented work.

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