Experimental study on thermal conductivity of polyurethane resin filled with modified nanoparticles

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Abstract

Most of the energy loss in building is through the walls. For this reason, insulating materials are used to reduce the loss of energy in buildings. Common insulations are high thickness thermally resistant material while thin coatings are usually used less. The purpose of this research is to introduce nanocomposite thin polymer coatings and its effect on thermal conductivity.In order to prepare the insulating layers with nanocomposite coatings, chemically modified nano zirconium oxide and nano aluminum oxide in three different weight percentages (1, 3, and 5%) were used in polyurethane matrix. To study thermal conductivity, the metallic plates are coated with prepared nanocomposites and the thermal conductivity of the samples was measured. The results show that adding zirconium oxide and aluminum oxide nanoparticles in polyurethane matrix increases the thermal resistance of the plates in all three weight percentages, compared to the coating without nanoparticles. The lowest thermal conductivity was found for 5% nano aluminum oxide composition, which, compared to the conductivity of the pure polyurethane resin, has decreased about 40%.

Keywords:

Nanoparticles, Thermal conductivity, Thermal barrier coating, Polyurethane, Insulation.

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1. Introduction

Regarding the increasing demand of energy in the world, conservation of energy is the major topic of numerous studies in recent years [1-3]. Decreasing energy loss in building envelope is one of the issues that have attracted the attention of material and mechanical experts, leading to the development of various materials and systems to achieve this goal. Using thermally resistant materials such as fiberglass, stone wool, polymeric foams and other insulating materials [4] is a straight forward solution. In most cases, insulated walls are thick, and insulated walls with less thickness may be a better choice. For this reason, attention has been drawn to low-thickness composite coating materials. Compared to ceramic and metallic materials, polymeric coatings innately have much lower thermal conductivity and are considered as better thermal insulators. If the polymers can be reinforced with suitable materials, advanced polymeric composites with low thickness, low thermal conductivity, and suitable thermal insulation properties can be developed [5, 6].Hollow ceramic particles are one of the items used in the coating of buildings. It has been reported that the use of these hollow particles makes 20%improvement in energy saving [7], moreover, application of the hollow particles is not limited by weather conditions, and they can be used in different areas with different climates [8].By choosing the right filler material for reinforcing polymeric composites, we can achieve coatings with low absorption or high reflection. In areas with high exposure to sunlight, these coatings can help to prevent buildings from warming and, hence, contribute to energy saving [9].These types of coatings can be made by using metal oxides[10].Various studies have been carried out to identify and select materials with low thermal conductivity and highly stable thermodynamic properties [11-13].In recent decades, the use of nano materials has made it possible to create special features, for which various applications have found in the industry. Nanofluids, for example, to which attention is drawn nowadays in heat transfer issue, have been the subject of a lot of research activities. The use of nanoparticles with ceramic and polymeric materials, have created ceramic and polymeric nanocomposites materials with special properties [14, 15]. The use of metallic nano oxides such as nano aluminum oxide and nano zirconium oxide as thermal protection coating in a ceramic matrix has shown satisfactory results [16, 17]. Besides, polymeric nanocomposite made with nanoparticles creates materials with different properties.

This research investigates the thermal conductivity of low-thickness polymeric nanocomposites. These nanocomposites may be used as thermal barrier coating to coat the exterior metallic surfaces of buildings. For this purpose, polyurethane resin (with high resistance to weather conditions) has been used as polymeric matrix, and aluminum oxide and zirconium oxide nanoparticles used to enhance thermal insulation properties. An experimental setup has been developed to investigate the heat transfer through the coatings and thermal properties.

2. Experimental setup
2.1 Materials

The polymer base coating was polyurethane 9016 WU18AX and supplied by Peka Chimie Company (Iran). Synthesize grade of isopropyl alcohol, acetone and vinyltrimethoxysilane (as coupling agent) were prepared from Merck Co. (Germany). Nano zirconium oxide and nano aluminum oxide with average of 40 nm diameters was from US Research Nanomaterials Inc. (USA). All the materials were used without any purification.

2.2 Method

Nanoparticles were modified by vinyltrimethoxysilane [17-19] and dried in vacuum oven for 12 hours at 60 ℃. Nanoparticles were dispersed in acetone by ultrasound and mixed with polyurethane resin by twin screw extruder. Blank composite was prepared with the same method without nanoparticles. Prepared materials were coated on clean 10×15 cm

galvanized plate and cured at 180 ℃ for 15 minutes.

2.3 Instruments and test methods

Ultrasound, Hielscher UP400S (Germany) and twin screw extruder Yantai Donghui Powder Processing Equipment Company (China) were used for dispersing of nanoparticles in polyurethane resin. Vacuum oven, WiseVen, WOV-30 -60 cmHg (South Korea) was used for drying of materials. Morphology of nanocomposites have studied by scanning electron microscopy (SEM), LEO 1455VP, Zeiss (Germany). Temperature of coatings surfaces was measured by thermography camera Testo 875-2 (United Kingdom).

To measure thermal diffusivity, various methods are used such as hot wire method, thermal wave method, and laser flash technique [20].The laser flash technique, which is used in this study, is the most common method because of its speed and applicability to small samples [20-22].

Laser flash is a technique for measuring thermal diffusivity and specific heat capacity by the flash method. Measurement is provided inside a vacuum insulated system with adiabatic condition to avoid convection effects and the laser beam is driven on the test sample. This test allows the comparison of thermal properties between substrate coating and coat coating. In this study, the German-made laser flash analyzer LFA 1000/1000 HT is used. The sample is put on a place located in a furnace at a definite temperature (-125 to 500 °C). At this temperature the sample surface is then irradiated with laser or xenon flash. This energy pulse results in a homogeneous temperature rise at the sample surface which is measured by a high speed infrared detector and thermal diffusivity values are computed from the temperature rise versus time data. The resulting measuring signal computes the thermal diffusivity, and in most cases the specific heat (C_p) data. If the density (ρ) is identified, the thermal conductivity can be calculated [22]. For this purpose, we used copper surfaces with diameter 1.25 cm coated with thin polymeric nanocomposites. The specific heat capacity is measured using Mettler Toledo calibrated at five points with Zn, Al, Au and Pd in samples according to ASTM 1269 standard. The density is measured (ASTM B 923) with Helium Pycnometry calibrated with stainless steel sphere. The thermal diffusivity is measured according to ASTM E1461 standard using the laser flash analyzer LFA 1000/1000 HT calibrated with Graphite. This device measures thermal diffusivity and specific heat capacity within a temperature range of -125 to 500 °C using an absolute measurement technique and covers the widest measuring range from 0.1 to 2000 W/(m.K) for thermal conductivity and from 0.01 to 1000 mm^2/s for thermal diffusivity. Equation 1gives the value of thermal conductivity using the results obtained according to ASTM E1461.

$$K(T) = a(T) \cdot C_p(T) \cdot \rho(T) \qquad (1)$$

where, a(T) is the thermal diffusivity, $C_p(T)$ is the specific heat capacity, and $\rho(T)$ is the density.

3. Results and discussion
3.1. Morphology of nanocomposites

Figure 1 shows the images of scanning electron microscopy for aluminum oxide and zirconium oxide urethane nanocomposites. As we can see, nanoparticles are well distributed in the urethane matrix. For both 1 and 3% samples, a homogeneous distribution of nanoparticles is observed. In 5% samples, nanoparticles begin to agglomerate. Therefore, we can predict that above 5%, due to the adhesion and aggregation of nanoparticles and non-homogeneous distribution of them, the properties will degrade.
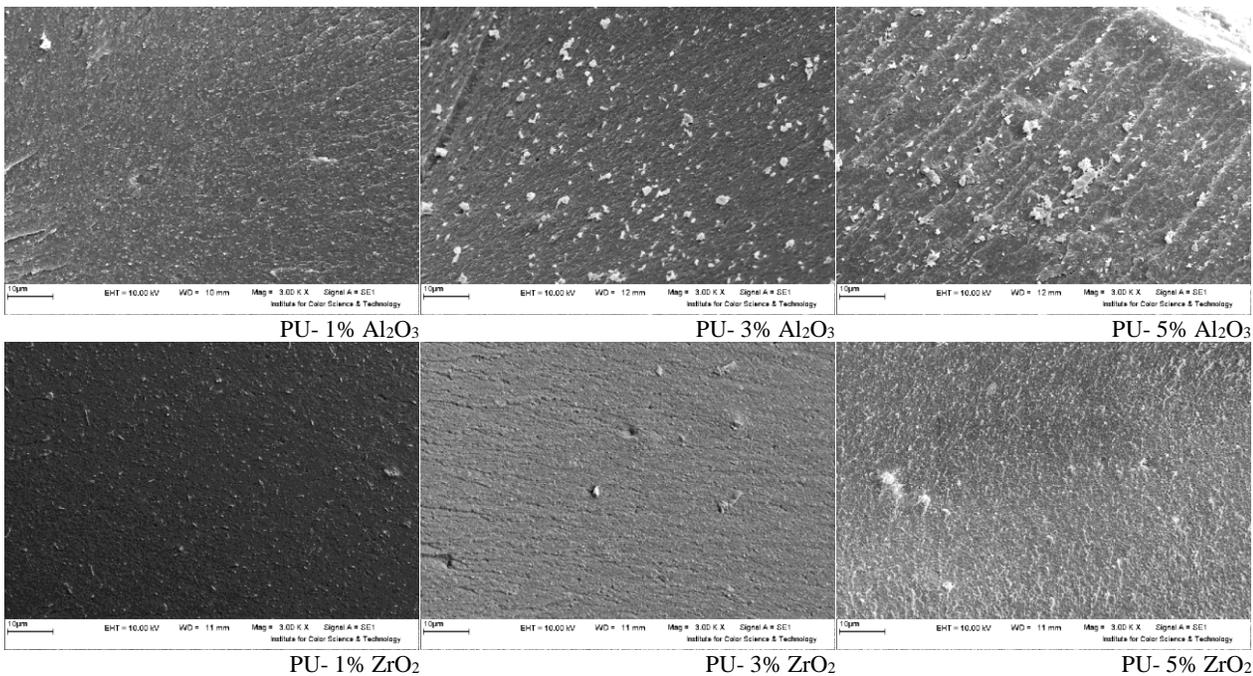
PU- 1% Al$_2$O$_3$	PU- 3% Al$_2$O$_3$	PU- 5% Al$_2$O$_3$
PU- 1% ZrO$_2$	PU- 3% ZrO$_2$	PU- 5% ZrO$_2$

Fig. 1. SEM images of urethane nanocomposites with various nanoparticles.

3.2. Specific heat capacity of nanocomposites

To measure and calculate the thermal conductivity, we need to measure the specific heat capacity of the samples. The specific heat capacity depends on the intermolecular forces, kinetic motions in the polymer chain, and its physical state. The type of movements in a polymer chain affects both the heat transfer rate through the polymer chain and the heat capacity of the substance. The temperature of a substance increases with the movements of its components; the faster the movements of components, the faster the temperature rise, and hence, the lower the heat capacity. Furthermore, if the intermolecular forces are weak, the temperature will again rise rapidly since the particles escape from the forces of attraction; this leads to an increase in their movements and, thus, in the temperature of the substance. As a result, the weaker the intermolecular forces, the lower the specific heat capacity [23]. Besides, the specific heat capacity of nanocomposites depends on the type, size, weight content, distribution, and crystalline structure of nanoparticles. Since the

particles for all coatings are the same size, the effect of size is not taken into account. For nano aluminum oxide, more adhesion of particles (compared to nano zirconium oxides) was observed. Figures 2 and 3 are shown the specific heat capacity of samples with nano aluminum and zirconium oxides.
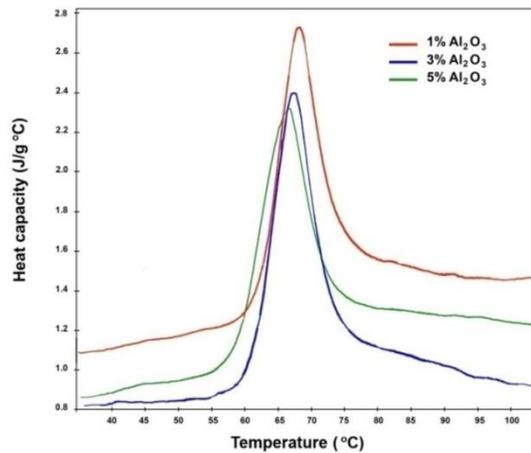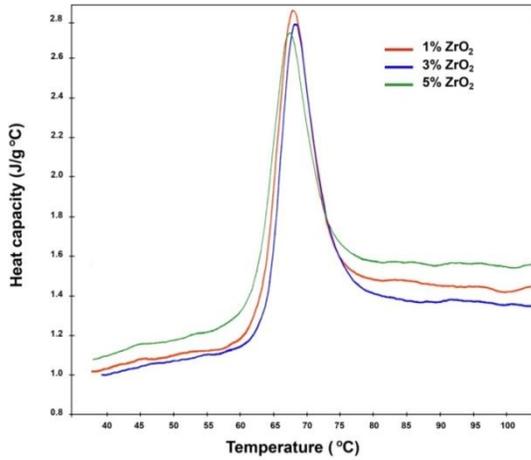

Fig. 2. Specific heat capacity of urethane nanocomposites containing 1, 3, and 5wt% nano aluminum oxide.

Fig. 3. Specific heat capacity of urethane nanocomposites containing 1, 3, and 5-wt%nanozirconium oxide.

Since the specific heat capacity of nano aluminum oxide (0.95 J/(g.K)) is higher than that of nano zirconium oxide (0.42 J/(g.K)), by adding nano aluminum oxide to the polymeric matrix we expect higher heat capacity of the coating than that with zirconium oxide nanoparticles. We also expect the heat capacity of the coatings to increase with the weight content, but such factors as their dispersion and their crystalline structure are important in increasing or decreasing the specific heat capacity and have given rise to different behavior of the heat capacity of the coatings. As shown in Figures 2 and 3, the specific heat capacity of nano aluminum oxide samples increases at 1, 3, and 5wt%, respectively, whereas nano zirconium oxide samples increases at 3, 5, and 1wt%, respectively. As can be seen, the specific heat capacity of the coatings increases with the weight content of the nanoparticles of zirconium oxide, which regarding the results in Figure 1, can be due to their good dispersion. Moreover, the minimum value of specific heat capacity is for 3wt% nano aluminum oxide; this, based on the results obtained from the scanning electron microscopy images (Figure 1), is attributable to the quality of the nanoparticles dispersion in the polymeric matrix.

3.3. Thermal conductivity of nanocomposites

Thermal conductivity of nanocomposite coatings depends on thermal diffusivity, specific heat capacity, and density of the coatings. It also depends, like the specific heat capacity, on the size, weight content, type and chemical behavior of the nanoparticles [24]. In this study, as mentioned previously, the size of particles in the coatings is the same; hence, its effect is not considered in the analyses. According to the rule of mixtures (Equation 2), the thermal conductivity of nanocomposites is a function of the volume fraction of particles and their mesh, and increases with the weight content of the nanoparticles [25-28].

$$k_c = k_p \phi_p + k_m \phi_m \tag{2}$$

where, k_c, k_p, and k_m, are the thermal conductivities of the composite, particles, and matrix, respectively. Φ_p and Φ_m are the volume fractions of the particles and mesh, respectively. The thermal conductivity of the nanocomposite coatings depends on the thermal properties of the nanoparticles. An increase in the weight content of the nanoparticles leads to an increase in the heat transfer area of the nanoparticles in the polymeric matrix and, thereby, an increase in the thermal conductivity coefficient [29].

Table 1 presents the results, which is obtained for the thermal conductivity, thermal diffusion, specific heat capacity, and density of the elaborated coatings. As given in table 1, the thermal conductivity does not follow an orderly manner with the changes in the weight content of the nanoparticles. With reference to Figure 1, these changes can be due to disperse situation of nanoparticles in polymeric matrix. Taking into account the possibility of the accumulation of the nanoparticles at higher weight percentages and, thereby, the reduction of the heat transfer area of the nanoparticles in the polymeric matrix, the thermal conductivity of the samples will reduce.

As can be seen in Table 1, the measured density of the coatings is approximately the same; therefore it has no effect on their thermal conductivity. It is also observed that, compared to nano zirconium oxide, the lowest thermal conductivity coefficients are related to aluminum oxide nanocomposite coatings, which may be due to the properties of nano aluminum oxide and its

chemical performance after the surface modification of the nanoparticles in the polymeric matrix. These changes can enhance interfacial forces and consequently reduce the thermal resistance in the polymeric matrix [30]. Also, the minimum thermal conductivity coefficient is related to the 5 wt% aluminum oxide nanocomposite coating, which may be regarding to increasing the amount of the nanoparticles.

This minimum conductivity coefficient is about 40% lower than pure polyurethane sample, which leads to a decrease in the heat flux through the surface. According to Fourier's law, heat flux is directly related to thermal conductivity. Therefore, regarding the temperature difference and area to be constant, the heat flux through the wall is directly proportional to the thermal conductivity.

In order to verify the results, the measurements were performed 10 times for each sample. Then, the effects of weight content and type of the nanoparticles on thermal conductivity were studied and also the standard deviation of the samples was analyzed by SPSS software, as presented in Table 2.

Table 1. Thermal conductivity, thermal diffusivity, specific heat capacity and density of samples.

Sample	Density ρ(g/cm^3)	Specific Heat Capacity Cp (J/(g.K))	Thermal Diffusivity a (cm^2/s)	Thermal Conductivity K (W/(m.K))
Blank - 0%	1.470	1.02	0.00342	0.513
PU-1% nano ZrO$_2$	1.420	0.98	0.00400	0.556
PU-3% nano ZrO$_2$	1.455	1.00	0.00292	0.425
PU-5% nano ZrO$_2$	1.520	1.04	0.00308	0.486
PU-1% nano Al$_2$O$_3$	1.460	1.10	0.00230	0.370
PU-3% nano Al$_2$O$_3$	1.42	0.80	0.00341	0.388
PU-5% nano Al$_2$O$_3$	1.468	0.90	0.00221	0.293

Table 2. The effect of various parameters on thermal conductivity.

| Source	Type of Calculation (Type III Sum of Squares)	Degree of Freedom (df)	Error (Mean Square)	Ratio (F)	Level of Confidence (Sig)	Size Effect (Partial Eta Squared)
Intercept	8.780	1	8.780	320.168	0.000	0.988
VAR00001	0.292	2	0.146	5.318	0.045	0.727
Error	0.110	4	0.027	-	-	-

As shown in Table 2, the material and weight content of the nanoparticles changes the thermal conductivity of the coatings by 72.7%, whereas other factors such as dispersion of nanoparticles contribute to about 27% of the changes in thermal conductivity. Also, at 95% confidence level, it

can be concluded that the thermal conductivity of nanocomposite coatings is best improved with nano aluminum oxide compared to the other nanoparticles. Figure 4 shows the values of the average and the standard deviation of the thermal conductivity of samples. As can be seen, the low value of standard deviation demonstrates a high accuracy in the measurements.

Figure 5 shows the averages and standard deviations of the thermal conductivity of the samples, which are coated on copper plates. Similar to Figure 4, the results in Figure 5 are highly accurate in the measurements. The left bar in Figure 5 indicates the pure copper sample. As can be seen, the use of these coatings leads to a significant reduction in the thermal conductivity of all of the samples.
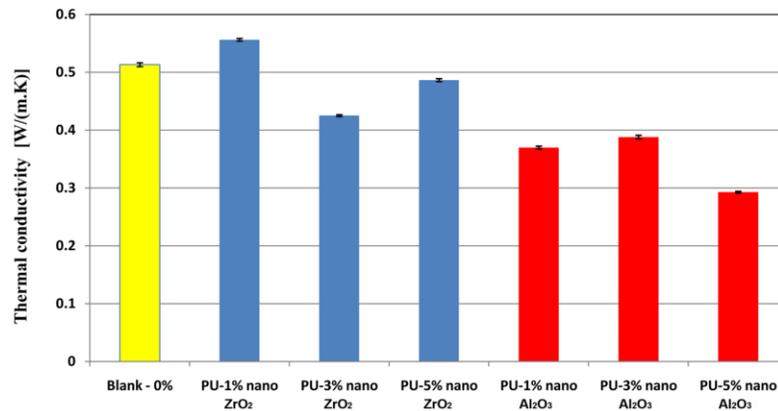
Fig. 4. Standard deviation of thermal conductivity of nanocomposite coatings with 0, 1, 3 and 5% of zirconium oxide and aluminum oxide nanoparticles.
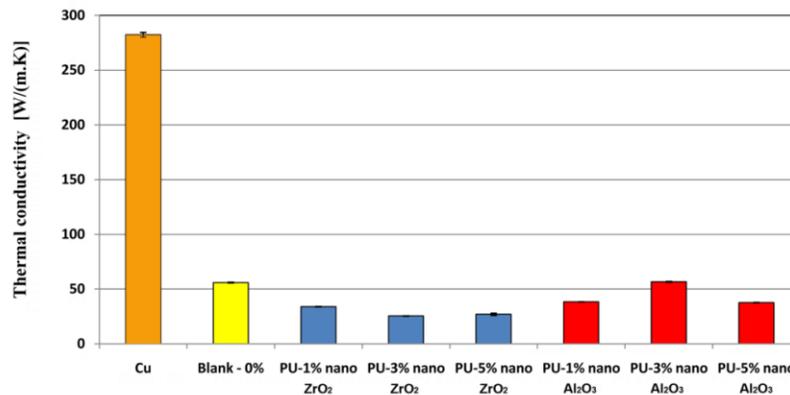
Fig. 5. Standard deviation of thermal conductivity of nanocomposite coating on copper plate with 0, 1,3 and 5% of zirconium oxide and aluminum oxide nanoparticles.
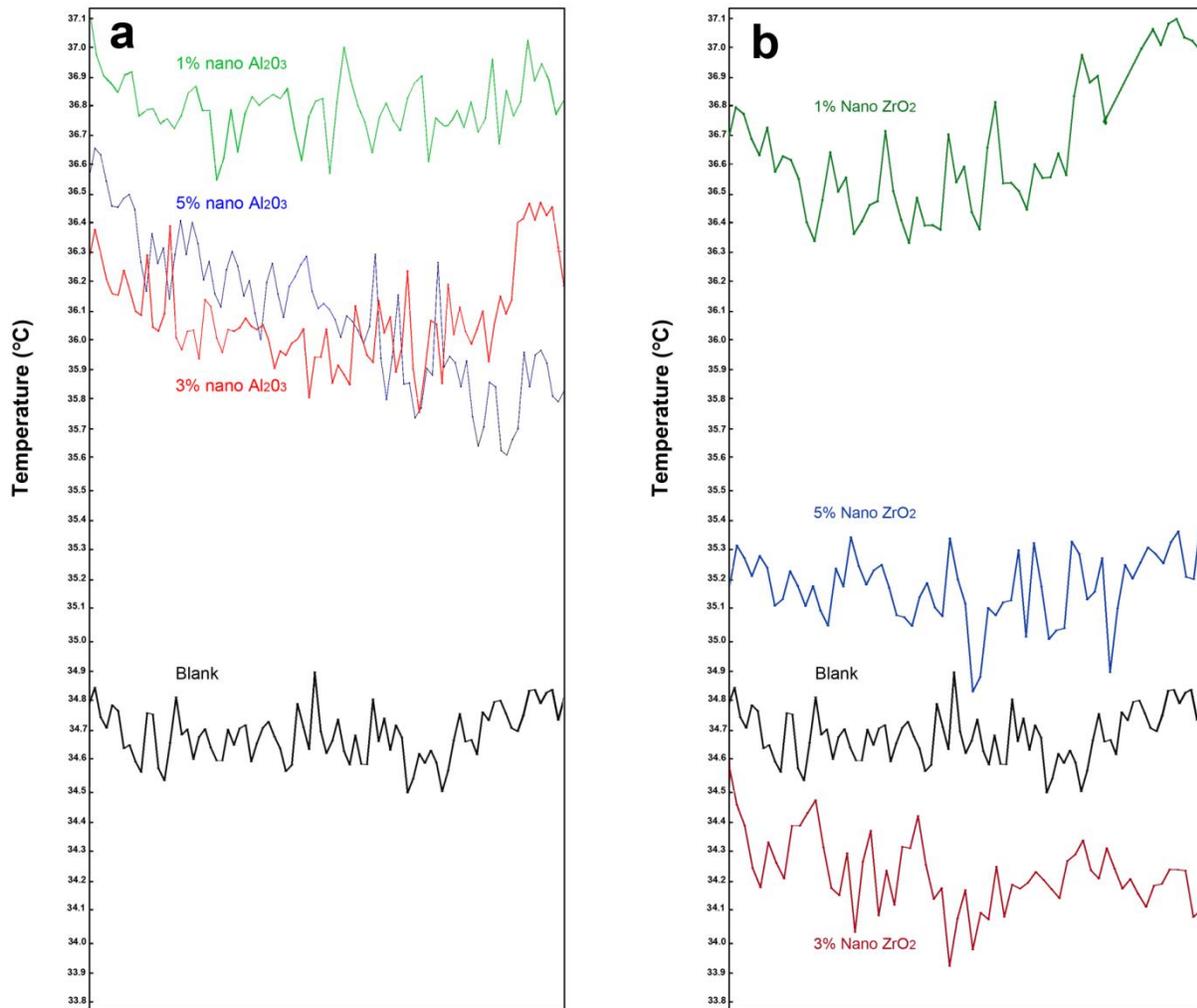
Fig. 6. Thermography results of nanocomposite coating on copper plate. a) Samples with 0, 1, 3 and 5% of aluminum oxide nanoparticles and b) Samples with 0, 1, 3 and 5% of zirconium oxide nanoparticles.

To investigate heat transfer accurately, temperature variations on the surface of the samples are measured using the thermography camera. The samples of pure polyurethane and polyurethane with different weight percentage of nano aluminum oxide and nano zirconium oxide are coated on copper plates. As shown in Figure 6, coating with 3 wt% nano zirconium oxide has the lowest surface temperature. Also, regarding the results of Figure 5, the lowest thermal conductivity coefficient is related to the coating contain 3 wt% nano zirconium oxide. Therefore, when the surface temperature and thermal

conductivity coefficient of coating are low, the surface heat flux will decrease and, hence, the temperature of the other side of the sample is decreased, too [31, 32]. As a result, the prepared coating acts effectively as insulation, reducing significantly the energy consumption.

4. Conclusion

Nanoparticles can alter the thermal conductivity behavior of polymeric coatings. This may be due to the behavior and nature of the particles, their

shape and their amount. The present study investigates the effect of adding zirconium oxide and aluminum oxide nanoparticles in polymeric coatings on conductive heat transfer through these coatings. For this purpose, coatings with composition of polyurethane resin and chemically modified nano zirconium oxide and nano aluminum oxide were prepared and coated on metallic plates. In order to study thermal conductivity, thermal diffusivity, specific heat capacity and density of the samples were measured and then, the thermal conductivity is calculated. Besides, the effects of various parameters such as the type and weight content of the nanoparticles on the thermal conductivity of the samples are analyzed. Based on results, it is concluded that, by adding modified zirconium oxide and aluminum oxide nanoparticles to polyurethane resin, the thermal conductivity of the samples is improved in all weight percentages compared to that of the pure polyurethane samples. The lowest thermal conductivity coefficients are related to the aluminum oxide samples, which can be due to its nature. Also, the sample with 5wt% nano aluminum oxide has the minimum thermal conductivity coefficient which is about 40% lower than pure polyurethane resin. On the other hand, the lowest surface temperature and thermal conductivity coefficient are observed in sample with 3wt% zirconium oxide. When the surface temperature and thermal conductivity of the coating are low, the surface heat flux and the temperature of the other side of the sample are decreased.

References

[1]   H. Zhou, M. Rao, and K. T. Chuang, "Knowledge-based automation for energy conservation and indoor air quality control in HVAC processes", *Engineering Applications of Artificial Intelligence*, Vol. 6, Issue 2, pp. 131-144, (1993).

[2]   R. Parameshwaran, and S. Kalaiselvam, "Energy efficient hybrid nanocomposite-based cool thermal storage air conditioning system for sustainable buildings", *Energy*, Vol. 59, pp. 194-214, (2013).

[3]   R. Parameshwaran, S. Kalaiselvam, S. Harikrishnan, and A. Elayaperumal, "Sustainable thermal energy storage technologies for buildings: A review", *Renewable and Sustainable Energy Reviews*, Vol. 16, Issue 5, pp. 2394-2433, (2012).

[4]   L. Aditya, T.M.I. Mahlia, B. Rismanchi, H.M. Ng, M.H. Hasan, H.S.C. Metselaar, O.Muraza, and H.B. Aditiya, "A review on insulation materials for energy conservation in buildings", *Renewable and Sustainable Energy Reviews*, Vol. 73, pp. 1352-1365, (2017).

[5]   M. Zhao, W. Pan, Ch. Wan, Zh. Qu, Zh. Li, and J. Yang, "Defect engineering in development of low thermal conductivity materials: A review", *Journal of the European Ceramic Society*, Vol. 37, Issue 1, pp. 1-13, (2017).

[6]   Y. Lin, Y.Jia, G. Alva, and G. Fang, "Review on thermal conductivity enhancement, thermal properties and applications of phase change materials in thermal energy storage", *Renewable and Sustainable Energy Reviews*, ISSN 1364-0321, https://doi.org/10.1016/j.rser.2017.10.002, (2017).

[7]   A. A. Azemati, B. Shirkavand Hadavand, H. Hosseini, A. and SalemiTajarrod, "Thermal modeling of mineral insulator in paints for energy saving", *Energy and Buildings*, Vol.56, pp. 109-114, 2013.

[8]   G.A. Sheikhzadeh, A.A. Azemati, H. Khorasanizadeh, B. Shirkavand Hadavand, and A. Saraei, "The effect of mineral micro particle in coating on energy consumption reduction and thermal comfort in a room with a radiation coolingpanel in different climates", *Energy and Buildings*, Vol.82, pp. 644-650, (2014).

[9]   S. W. Lee, Ch. H. Lim, and E. B.Salleh, "Reflective thermal insulation systems in building: A review on radiant barrier and reflective insulation", *Renewable and*

Sustainable Energy Reviews, Vol. 65, pp. 643-661, (2016).

[10]   A.A. Azemati, H. Khorasanizadeh, B. Shirkavand Hadavand, and G.A. Sheikhzadeh, "Study on radiation properties of polyurethane/nano zirconium oxide nanocomposite coatings", *Materials Science Forum*, Vol. 894, pp. 109-112, (2017).

[11]   D. R. Clarke, "Materials selection guidelines for low thermal conductivity thermal barrier coatings",*Surface and Coatings Technology*, Vol. 163–164, pp. 67-74, (2003).

[12]   W. W. Li, W. L. Cheng, B.Xie, N. Liu, and L. S. Zhang, "Thermal sensitive flexible phase change materials with high thermal conductivity for thermal energy storage", *Energy Conversion and Management*, Vol. 149, pp. 1-12, (2017).

[13]   K.S. Reddy, and S. Jayachandran, "Investigations on design and construction of a square guarded hot plate (SGHP) apparatus for thermal conductivity measurement of insulation materials", *International Journal of Thermal Sciences*, Vol. 120, pp. 136-147, (2017).

[14]   H. Porwal, and R. Saggar, "Ceramic Matrix Nanocomposites", *Comprehensive Composite Materials II*, Elsevier, Oxford, pp. 138-161, (2018).

[15]   J. Zhao, L. Wu, Ch. Zhan, Q. Shao, Zh.Guo, and L. Zhang, "Overview of polymer nanocomposites: Computer simulation understanding of physical properties", *Polymer*, available online 6 November 2017, ISSN 0032-3861, https://doi.org/10.1016/j.polymer.2017.1 0.035.

[16]   V. Kumar, and B. Kandasubramanian, "Processing and design methodologies for advanced and novel thermal barrier coatings for engineering applications", *Particuology*, Vol. 27, pp. 1-28, (2016).

[17]   H. Farzad, F. Najafi, M. Bengisu, E. Yilmaz, and B. Shirkavand Hadavand, "Synthesis and characterization of aliphatic tri-functional oligomeric urethane methacrylate used for UV-curable aluminum pigmented coatings″, *Journal of Macromolecular Science, Part A*, Vol. 50, pp. 504-512, (2013).

[18]   B. Shirkavand Hadavand, M. Ataeefard, and H. Fakharizadeh Bafghi, "Preparation of modified nanoZnO/polyester/TGIC powder coating nanocomposite and evaluation of its antibacterial activity", *Composites Part B Engineering*, Vol. 82, pp. 190-195, (2015).

[19]   A. Madhi, B. Shirkavand Hadavand, and A. Amoozadeh, "Synthesis, characterization and study on thermal stability, mechanical properties and thermal conductivity of UV-curable urethane acrylate-Clay (MMT) nanocomposites", *Journal of Applied Chemistry*, Vol. 12, No. 45, pp. 91-98, (2018).

[20]   W. Nunes dos Santos, "Thermal properties of polymers by non-steady-state techniques", *Polymer Testing*, Vol.26, pp. 556-566, (2007).

[21]   W. Nunes dos Santos, P. Mummery, and A. Wallwork, "Thermal diffusivity of polymers by the laser flash technique",*Polymer Testing*, Vol.24, pp. 628–634, (2005).

[22]   P.S. Gaal, M.A. Thermitus, and D.E. Stroe, "Thermal conductivity measurements using the flash method",*Journal of Thermal Analysis and Calorimetry*, Vol.78, pp. 185-189, (2004).

[23]   I. Tavman, Y. Aydogdu, M. Kök, A. Turgut, and A. Ezan, "Measurement of heat capacity and thermal conductivity of HDPE/expanded graphite nanocomposites by differential scanning calorimetry", *Archives of Materials Science and Engineering*, Vol. 50, pp. 56-60, (2011).

[24]   I. H. Tavman, "Thermal and mechanical properties of aluminum powder-filled high-density polyethylene composites",*Journal of Applied Polymer Science*, Vol. 62, pp. 2161-2167, (1996).

[25]   D.M. Bigg, "Thermal conductivity of heterophase polymer compositions",*Advanced Polymer Science*, Vol.119, pp. 1–30, (1995).

[26]   H. Zhou, S. Zhang, and M. Yang, "The effect of heat-transfer passages on the effective thermal conductivity of high filler loading composite materials",*Composites Science and Technology*, Vol.67, pp. 1035-1040, (2007).

[27]   S. Okamoto, and H. Ishida, "A new theoretical equation for thermal conductivity of two-phase systems",*Journal of Applied Polymer Science*, Vol.72, pp. 1689-1697, (1999).

[28]   H. Ebadi-Dehaghani, M. Reiszadeh, A. Chavoshi, M. Nazempour, and M. H. Vakili, "The effect of zinc oxide and calcium carbonate nanoparticles on the thermal conductivity of polypropylene", *Journal of Macromolecular Science, Part B*, Vol. 53, pp. 93-107, (2014).

[29]   L. Frormann, A. Iqbal, and S.A. Abdullah, "Thermo-viscoelastic behavior of PCNF-filled polypropylene nanocomposites",*Journal of Applied Polymer Science*, Vol. 107, pp. 2695-2703, (2008).

[30]   A. Li, C. Zhang, and Y. F. Zhang, "Thermal conductivity of graphene-polymer composites: Mechanisms, properties, and applications",*Polymers*, Vol. 9, pp. 420-437, (2017).

[31]   W. Zhou, Sh. Qi, Ch. Tu, H. Zhao, C. Wang, and J. Kou, "Effect of the particle size of Al_2O_3on the properties of filled heat-conductive silicone rubber", *Journal of Applied Polymer Science,* Vol.104, pp. 1312-1318, (2007).

[32]   A Ch. Liu, M. Chen, D. Zhou, D. Wu, and W. Yu, "Effect of filler shape on the thermal conductivity of thermal functional composites", *Journal of Nanomaterials*, Vol. 2017, pp. 1-15, Article ID 6375135, 15 pages, https://doi.org/10.1155/2017/6375135, (2017).