Heat insulation effect in solar radiation of polyurethane powder coating nanocomposite

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This study aims to improve polyurethane-based coating by modified zirconium oxide and aluminum oxide nanoparticles for preparing thin polymeric heat insulation coatings. In the first step, the nanoparticles were chemically modified with the silane coupling agent. Then, three different weight percent of modified nanoparticles (1, 3, and 5% w/w) were mixed with polyurethane, to prepare the nanocomposites, which were coated on metallic plate samples. Then, these plates are used to measure the radiation heat transfer coefficients, absorption coefficient in a region of short wavelengths (UV/VIS/NIR), the emissivity coefficient, and thermography of the samples in a region of long wavelengths (IR). Results showed that by adding the modified nanoparticles to the polyurethane matrix, absorption was decreased and the emissivity coefficient was increased. According to the thermography results, it was observed that the surface temperature of both samples with 3% w/w of nanoparticles had the minimum temperature compare to others. Minimum heat surface observed for 3% w/w of modified nano zirconium oxide.

Energy conservation has gained importance in many advanced industries as a part of eco-efficiency. In this regard, the use of advanced nanocomposites and structural coating has become one of the innovative approaches in energy conservation applications1-2. Reinforced composites using microfibers and ceramic particles in the form of thin layer and coatings, are effectively used to enhance the mechanical features3-4 while reducing the emissivity coefficient and heat transfer that results in reducing the thermal energy and annual electricity consumption5-8, while in buildings under different climate changes. The absorption and emission of the coating material features are known to be important in surface heating, which could be controlled to produce a coating with cooling features. The cooling effect is measured by the reflection of the surface solar radiation, as well as the surface emissivity, as reflected back into the atmosphere. Such a concept was used for white materials in construction applications (e.g. roof surface) where is subjected to large solar radiation and has a high reflectance9-12.

A testing method has been developed to evaluate the energy performance and sustainability of innovative new products, which are known as cool colors13. These products were the component of selective materials (high absorption coefficient in the visible light region and high reflectance in the infrared light region), and consequently, they had both the aesthetics and cooling capabilities parameters in construction application.

Joudi14 studied the radiation properties of the surface, including the emissivity coefficient of interior surfaces, modeling methods, and its results on building energy performance and the thermal environment of buildings. This study indicated that surface with low emissivity can increase indoor air temperature vertical gradient, which is dependent on the time of day and outside weather conditions.

In other research by Ascione et al.15, a new method was implemented, in which thermo-physical properties of a building and coatings were optimized by changing the thermal resistance, capacity, and radiation properties of the surfaces exposed to radiation. In summer, the majority of cooling load faced on the building as a result of solar radiation is on the external walls of buildings. By reducing the amount of solar radiation absorbed by the external walls, the cooling load can be highly reduced. Although, the facade of the building and constructional

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of 55 μm. The nanocomposite was electrostatically coated on 10 × 15 cm² galvanized plate and cured at 180 °C.

Preparation of polyurethane nanocomposite. Nano ZrO₂ and Al₂O₃ particles were chemically modified by a silane coupling agent. Then, polyurethane resin and different percentages (1, 3, and 5% w/w) of modified nano ZrO₂ and Al₂O₃ particles were extruded in the twin screw extruder (Yantai Donghui Powder Processing Equipment Company, China). The prepared chips were powdered and sieved to the average particle size.

Table 1. Polyurethane power coatings nanocomposite samples.

| Samples   | Nano ZrO₂ (%) | Nano Al₂O₃ (%) |
|-----------|---------------|----------------|
| Blank     | 0             | 0              |
| PU-1% nano ZrO₂ | 1             | 0              |
| PU-3% nano ZrO₂ | 3             | 0              |
| PU-5% nano ZrO₂ | 5             | 0              |
| PU-1% nano Al₂O₃ | 0             | 1              |
| PU-3% nano Al₂O₃ | 0             | 3              |
| PU-5% nano Al₂O₃ | 0             | 5              |

As a thermal barrier, polymer coatings have to be made with specifically large thicknesses as foams or porous composites, which is the main disadvantage of such coating and limit its application. Therefore, to achieve a thin and durable coating with thermal barrier properties, a special material is required. One of the ways is to use suitable additives such as nanoparticles in the coating structure. In recent years, the use of powder coatings to coat metal products that are exposed to direct sunlight, become a common choice to prevent them from the transfer of radiant heat energy of the sun. For example, in the insulation of metal canopies and air conditioners exposed to sunlight, a thin insulating coating with a thermal insulating property is required in addition to creating a suitable appearance. Since the use of powder coatings to cover metal products has become common in recent years, and in some cases, the manufactured products are exposed to direct sunlight, it is necessary to protect the transfer of radiant thermal energy. In this regard, this study focused on the idea of using nanoparticles in powder coatings that are able to reflect sunlight, as one of the solutions to provide the coating with a proper thermal resistance in addition to a suitable low thickness.

Materials and method

Materials. Vinyltrimethoxysilane (VTMS) and isopropyl alcohol were provided from Merck Co. (Germany) and used for the chemical modification of nanoparticles. Formulated polyurethane powder coating resin 9016 WU18AX was prepared in Peka Chimie Co. (Iran). The nano ZrO₂ and Al₂O₃ (40 nm) were purchased from US Research Nanomaterials Inc. (USA).

Preparation of polyurethane nanocomposite. Nano ZrO₂ and Al₂O₃ particles were chemically modified by a silane coupling agent. Then, polyurethane resin and different percentages (1, 3, and 5% w/w) of modified nano ZrO₂ and Al₂O₃ particles were extruded in the twin screw extruder (Yantai Donghui Powder Processing Equipment Company, China). The prepared chips were powdered and sieved to the average particle size of 55 μm. The nanocomposite was electrostatically coated on 10 × 15 cm² galvanized plate and cured at 180 °C for 15 min. The composition of samples is presented in Table 1.

Instruments and test methods. The hemispherical total reflectance and emissivity were measured by spectrophotometer/reflectometer AZ-Technology’s TEMP 2000A model (USA) in the range of long wavelengths from less than 3 μm to greater than 35 μm (infrared region) and measurement accuracy (for specular and diffuse samples) 1% of full scale for gray samples and 3% of full scale for non-gray samples. Due to the changes in the reflectance and emissivity coefficient, the coating performance is related to the wavelength in all wavelengths and then assumed as the non-gray body. This instrumentation is not limited in wavelength range due to filters, windows, or coatings, placed in the optical path. The measurements were repeated five times at different parts of each sample, according to ASTM E408 test method. In long wavelengths, the absorption and emissivity coefficient for the black body was the same. Also, due to the constant temperature in the experimental test, it is assumed that thermal equilibrium is established and according to Kirchhoff’s law, if there is thermal equilibrium in the long wavelengths, absorption and emissivity coefficient can be considered equal for all samples. A large amount of solar heat gain is in the region wavelength between 0.2 and 2.5 μm (UV/Vis/NIR). Similarly, the Shimadzu spectrophotometer UV-3600 model (Japan) was used to obtain the amount of reflection coefficient of the surfaces in the same wavelengths at this region.
Increasing the nanoparticles to 5% w/w results in the better dispersion with the suitable percentage of nanoparticles, the aggregates of particles are higher than in zirconium oxide and larger particles are seen, which is related to the intrinsic properties of aluminum oxide and its intermolecular interactions. These gatherings are evenly dispersed. This proper distribution is also evident in the results. However, in modified aluminum oxide nanoparticles, the aggregates of particles are higher than in zirconium oxide and larger particles are seen, which is related to the intrinsic properties of aluminum oxide and its intermolecular interactions. These gatherings are evenly dispersed.

The dispersion in the coating is approximately the same for both particles, and the modified nanoparticles are evenly dispersed. This proper distribution is also evident in the results. However, in modified aluminum oxide nanoparticles, the aggregates of particles are higher than in zirconium oxide and larger particles are seen, which is related to the intrinsic properties of aluminum oxide and its intermolecular interactions. These gatherings are also well-distributed. As can be seen in Fig. 1, in the 5% sample of both nanoparticles, these aggregations have also well-distributed. As can be seen in Fig. 1, in the 5% sample of both nanoparticles, these aggregations have

Results and discussion

Morphology. The scanning electron microscope (SEM) images of polyurethane nanocomposite with different amounts of modified nano ZrO2 and Al2O3 particles are shown in Fig. 1. In general, a good dispersion of nanoparticles in the urethane matrix could be viewed in samples with modified nano ZrO2, which is the effect of modification on the dispersion of nanoparticles. The best dispersion with the suitable percentage of nanoparticles was observed in a sample with 3% w/w nano ZrO2. Increasing the nanoparticles to 5% w/w results in the poor dispersion of nanoparticles. The best dispersion with the suitable percentage of nanoparticles is within ± 0.08 nm and for the near-infrared region is within ± 0.32 nm.

A 50 W halogen lamp and deuterium lamp (socket type) have a built-in mechanism for automatically adjusting the light-source position. The measurements were performed in the range of 282 to 393 nm at least five times at different parts of each sample according to ASTM E903-96 test method.

Infrared thermography (IRT) was measured in 8–15 µm (infrared region) with infrared camera Testo, 875-2 model (Germany). According to the obtained results and given that the transmission coefficient is zero (τ = 0), the absorption coefficient can be calculated using Eq. (1):

\[ \alpha + \tau + \rho = 1 \]  

where \( \alpha \) is absorption coefficient, \( \tau \) is transmission coefficient and \( \rho \) is reflection coefficient.

The amount of solar irradiance on the exterior surface of the exterior is \( Q_{\text{in} \text{out}(i)} \), which can be calculated using the equation:

\[ Q_{\text{in} \text{out}(i)} = \varepsilon \sigma(T_s^4 - T_{\text{sky}}^4) \]  

where \( \varepsilon \) is the emissivity coefficient, \( T_s \) is the surface temperature, \( q_s \) is solar incident radiation and \( T_{\text{sky}} \) is the sky temperature.

According to Eq. (2), if the emissivity coefficient is high, the amount of incident solar radiation heat flux to the exterior surfaces is reduced. This decreases the temperature of the interior surface, and therefore reduces the amount of cooling load necessary to bring the temperature of the interior space to the comfort temperature zone.

The amount of light passing through the atmosphere is not the same for all wavelengths and only in certain areas light passing is high and unlimited. Therefore, the amount of irradiance in the regions of 0.4–1.5, 1.5–2.5, 3–5, and 8–12 µm could be extraordinary and could be used for electro-optical systems. Meanwhile, only the regions of 3–5 and 8–12 µm are appropriate to make and apply passive infrared imaging system (i.e. thermography). Since, objects have significant reflectance in ambient temperature conditions, only in these two regions (3–5 and 8–12 µm) and are not absorbed by atmospheric effects.

The thermography measurement was performed outdoors and exposed to direct sunlight at noon on a sunny summer day. The samples were on a plastic platform to eliminate the effects of heat transfer between platform and samples. Data of weather conditions including temperature, relative humidity, wind speed, and amount of solar radiation are obtained from the local weather center as presented in Table 2.

Table 2. Environment weather conditions in the time of experiments. Obtained by Carrier software.

| Time        | Air temperature (ºC) | Relative humidity (%) | Wind speed (m/s) | Maximum solar heat gain (w/m²) |
|-------------|----------------------|-----------------------|------------------|-------------------------------|
| Sunny summer day 1:00 PM | 32 | 36 | 21 | 19 | 3.13 | 768 |

The wavelength accuracy in ultraviolet and visible regions is ± 0.2 nm, in near-infrared region is ± 0.8 nm, for wavelength repeat accuracy ultraviolet and visible regions is within ± 0.08 nm and for the near-infrared region is within ± 0.32 nm.

The emissivity coefficients results of the samples in the wavelengths of 0.4–1.5, 1.5–2.5, 3–5, and 8–12 µm could be extraordinary and could be used for electro-optical systems. Meanwhile, only the regions of 3–5 and 8–12 µm are appropriate to make and apply passive infrared imaging system (i.e. thermography). Since, objects have significant reflectance in ambient temperature conditions, only in these two regions (3–5 and 8–12 µm) and are not absorbed by atmospheric effects.

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Emissivity of long wavelength. The emissivity coefficients results of the samples in the wavelengths range of 3 to 35 µm with standard deviation are presented in Fig. 2. The variance analysis of the samples was the exterior surfaces is reduced. This decreases the temperature of the interior surface, and therefore reduces the amount of cooling load necessary to bring the temperature of the interior space to the comfort temperature zone.

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Figure 1. SEM images of polyurethane nanocomposite coatings with different amounts of nano ZrO₂ and Al₂O₃.

Figure 2. Emissivity coefficient with standard deviation for polyurethane nanocomposite coatings.
done to obtain accurate results using the data measured (minimum five times) at different parts of each sample. Results indicated that, in the wavelength of solar radiation (infrared region), the average emissivity of coatings is about 0.87. Results showed that by adding modified nano ZrO₂ and Al₂O₃ to polyurethane coatings, the emissivity coefficient of coatings compare to coating without nanoparticles increased. Also, the emissivity coefficient of coatings for the sample with 3% w/w nano ZrO₂ compare to 1 and 5% w/w of nano ZrO₂ and 3% w/w nano Al₂O₃ compare to 1 and 5% w/w of nano Al₂O₃ are increased. It can be concluded that these nanoparticles with the optimum amount of 3% w/w have a positive role in changing the emissivity coefficient and the effect of nano ZrO₂ are more than nano Al₂O₃. The reason for the higher emissivity coefficient of samples with 3% w/w nanoparticles in comparison to the samples containing 5% w/w nanoparticles is due to a better dispersion of nanoparticles in the polymeric matrix. In samples containing 5% w/w aggregation of nanoparticles has happened.

**Absorption coefficient of short wavelength.** The reflection coefficients of the coated samples in region wavelengths of 0.2 to 2.5 μm were measured as shown in Figs. 3 and 4. In all cases, the reflection coefficient in the UV region (about 0.4 μm) is a bit more than expected, which is encountered due to the fluorescent effect of nano-
particles in coatings, which has been reported in the literature. Binder weight ratio effects and additives that adding to the polyurethane, change color and surface roughness that may help to increase reflection coefficient.

Since the samples are not transmittance, the transmission coefficient is equal to zero. By integrating the results in this region, hemispherical spectral reflection coefficients were obtained, and using Eq. (1), the absorption coefficient was calculated. It is observed that in a region wavelength of UV/VIS/NIR, the absorption coefficient of the coatings is satisfied at about 0.2. The variance analysis was done to obtain accurate results using a minimum of five data measured at different parts of each sample. Figure 5 presents a comparison of absorption coefficient results of different samples with the standard deviation.

Results indicated that by adding modified nano ZrO2 and Al2O3 to polyurethane coating, the absorption coefficient of the coating decreased in all samples compared to the coating without nanoparticles. It means that modified nano ZrO2 and Al2O3 have the ability to reduce absorption coefficient in coatings as a natural feature. It is worth mentioning that the same result is captured for the sample with 3% w/w nanoparticle with decreased absorption coefficient more than the other cases (the best sample for increasing the emissivity coefficient).

According to Eq. (2), it is clear that if the emissivity coefficient is high and the absorption coefficient is low, the amount of input radiation heat flux to the exterior surfaces is reduced, which, decreases the temperature of the interior surface and consequently reduces the amount of cooling load necessary to bring the space temperature to comfort temperature range.

The infrared thermography results. The temperature results of infrared thermography (IRT) of polyurethane coating with different amounts of nano ZrO2 and Al2O3 particles are shown in Fig. 6. As indicated in this Figure, the temperature of points M1 to M5, average surface temperature, and the temperature points of the P line, were determined for each sample. It should be noted that the measurements of surfaces temperature were done according to the impact emissivity coefficient of the specimens.

It is worth mentioning that, in addition to the variation of the emissivity coefficient of the surfaces dependent on different weight percentages of nanoparticles, the temperature of the surfaces is changed as the result of the effect of different weight percentages of ZrO2 and Al2O3 nanoparticles.

The temperature at different points of the samples with nanocomposite coating having 0, 1, 3, and 5% of ZrO2 and Al2O3 nanoparticles, is shown in Fig. 7. The result indicated that the temperature at 3% is lower than the other samples, which is due to the influence of ZrO2 and Al2O3 nanoparticles with polyurethane matrix, dispersion quality of nanoparticles in coatings. A slight abnormal increase in temperature in the 1% zirconium oxide sample compared to the blank sample is due to the lack of unintentional unequal conditions or error at the time of measurement. But considering the errors, it is clear that it is almost equal to the blank sample.

It is worth mentioning that, the amount of flux crossing of the surface is decreasing by reducing the temperature, which causes the reduction of heat transfer reduces and the amount of energy consumption.

Conclusion
In this study, polyurethane powder coatings nanocomposite with 1, 3, and 5% of aluminum oxide and zirconium oxide were prepared and then coated on metal plates and various tests were performed on them. Results showed that the existence of nanoparticles could have a significant effect in changing the thermal behavior of polymeric coatings. These effects depend on the nature, amount, and morphology of nanoparticles in the nanocomposite coating. Also, it is shown that by adding zirconium oxide and aluminum oxide nanoparticles to the polyurethane matrix, the emission coefficient of the coating in a region of IR was increased. The maximum emission
coefficients were observed in PU/3% w/w nano ZrO₂. In addition, the absorption coefficient of the PU/nano ZrO₂ and Al₂O₃ coatings in a region of UV/VIS/NIR (all cases) compared to the blank case was decreased. The minimum absorption coefficient was measured for PU/3% w/w nano ZrO₂ case. The lowest surface temperature was determined for PU/3% w/w nano ZrO₂ sample as proven by the thermography results.

Figure 6. Thermography of images of polyurethane nanocomposite coatings with different amounts of nano ZrO₂ and Al₂O₃.
Temperature of the samples of polyurethane coating with 0, 1, 3, and 5% of nano ZrO₂ and Al₂O₃.

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Author contributions
Conceptualization, methodology, validation, formal analysis, and investigations were done by A.A.A., S.S.R.K., H.K., M.P., G.A.S., M.S., and B.S.H.; Resources, funding acquisition, supervision and project administration: A.A.A., S.S.R.K., M.P., and B.S.H.; Data curation, writing—original draft preparation, writing—review and editing were done by A.A.A., S.S.R.K., and B.S.H. All authors have read and agreed to the published version of the manuscript.

Competing interests
The authors declare no competing interests.

Additional information
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