

RADIATIVE COOLING BY SAND

A Thesis

by

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RADIATIVE COOLING BY SAND

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To my beloved father and mother

I extend my heartfelt gratitude to both of you. Your unwavering support and presence have been the cornerstones of my entire life. Your constant encouragement and guidance have shaped not only my academic pursuits but also the person I have become. From the earliest stages of my education, you instilled in me a deep appreciation for knowledge and a relentless pursuit of excellence. Your dedication has been a constant reminder that I am never alone on this journey.

To my family

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ABSTRACT

Passive radiative cooling is one of the upcoming technologies likely to help decrease the negative impacts of climate crises and reduce energy consumption in buildings. The principle of radiative cooling involves radiating heat from the surface of a building into space, thus reducing the reliance on air conditioning systems. Understanding the fundamentals of passive radiative cooling is important for thermal management related to buildings and industrial applications. The principle of radiative cooling in buildings involves selectively reflecting incident radiating heat from the surroundings and selectively emitting energy from a surface to the surroundings to achieve minimum heat load on the system. In the case of buildings, solar radiation incident on roofs can be reflected or absorbed. The absorbed heat is either transferred to the surrounding air by convection or it will be emitted back to the sky. If a surface is desired to be radiatively cooled it must have a large reflection of solar energy at the visible wavelength range and the highest radiation emission at the atmospheric window range (8-13 μm wavelength range) as discussed in detail in the thesis. This technique can significantly help reduce greenhouse gas emissions as it will help decrease air-conditioner use in warm climates during the summer months.

Recently, the use of nanoparticles and different structures has been considered for passive radiative cooling applications. In this study, we investigate the radiative cooling properties of sand particles experimentally and compare their performance with other materials including silicon dioxide, titanium dioxide, and zinc oxide. Scanning electron microscopy (SEM) is used to analyze the morphology of the surface of sand the particles, and the other materials used in the study for comparison. Fourier transform infrared spectroscopy (FT-IR) experiments were conducted in order to measure

the absorbance and transmittance percentage of the samples. In addition, the absorption coefficient was calculated from the FT-IR measurements. The absorption coefficient of the sand samples showed a high ratio (in the units of $1/\text{m}$ which was approximately between 0.4×10^4 and 0.8×10^4 for all the pellet samples at the atmospheric window range. Also, the behavior of the absorption profiles of the sand sample and ZnO were very similar in their ratios, especially in the atmospheric window range. Also, we observed that the emission shows the highest peaks at $6.78 \mu\text{m}$, $9.24 \mu\text{m}$, $11.69 \mu\text{m}$, and $14.05 \mu\text{m}$ for all sand pellet samples. Thus, the properties of sand samples experimentally studied in this work show that they can be used for radiative cooling due to their abundance and thermal properties with better performance.

ÖZETÇE

Pasif radyatif soğutma, iklim krizlerinin olumsuz etkilerini azaltmaya ve binalardaki enerji tüketimini düşürmeye yardımcı olması muhtemel yeni teknolojilerden biridir. Radyatif soğutma prensibi, ısıнын bir binanın yüzeyinden uzaya yayılmasını ve böylece klima sistemlerine olan bağımlılığın azaltılmasını içerir. Pasif radyatif soğutmanın temellerini anlamak, binalar ve endüstriyel uygulamalarla ilgili termal yönetim için önemlidir. Binalarda radyatif soğutma prensibi, sistem üzerinde minimum ısı yükü elde etmek için çevreden gelen ısıнын seçici olarak yansıtılmasını ve bir yüzeyden çevreye seçici olarak enerji yayılmasını içerir. Binalar söz konusu olduğunda, çatılara gelen güneş radyasyonu yansıtılabilir veya absorbe edilebilir. Absorbe edilen ısı ya konveksiyon yoluyla çevredeki havaya aktarılır ya da gökyüzüne geri yayılır. Bir yüzeyin radyatif olarak soğutulması isteniyorsa, tezde ayrıntılı olarak tartışıldığı gibi, görünür dalga boyu aralığında güneş enerjisini büyük ölçüde yansıtması ve atmosferik pencere aralığında (8-13 μm dalga boyu aralığı) en yüksek radyasyon emisyonuna sahip olması gerekir. Bu teknik, yaz aylarında sıcak iklimlerde klima kullanımını azaltmaya yardımcı olacağından sera gazı emisyonlarının azaltılmasına önemli ölçüde yardımcı olabilir.

Son zamanlarda, pasif radyatif soğutma uygulamaları için nanopartiküllerin ve farklı yapıların kullanımı düşünülmektedir. Bu çalışmada, kum partiküllerinin radyatif soğutma özelliklerini deneysel olarak araştırıyor ve performanslarını silikon dioksit, titanyum dioksit ve çinko oksit gibi diğer malzemelerle karşılaştırıyoruz. Taramalı elektron mikroskobu (SEM), kum parçacıklarının ve çalışmada karşılaştırma için kullanılan diğer malzemelerin yüzey morfolojisini analiz etmek için kullanılmıştır. Örneklerin absorbans ve geçirgenlik yüzdesini ölçmek için Fourier dönüşümlü kızılötesi

spektroskopisi (FT-IR) deneyleri yapılmıştır. Ayrıca, FT-IR ölçümlerinden soğurma katsayısı hesaplanmıştır. Kum numunelerinin soğurma katsayısı, atmosferik pencere aralığında tüm pelet numuneleri için yaklaşık 0.4×10^4 ve 0.8×10^4 arasında olan yüksek bir oran (1/m birimlerinde) göstermiştir. Ayrıca, kum numunesi ve ZnO'nun soğurma profillerinin davranışı, özellikle atmosferik pencere aralığında, oranlarında çok benzerdi. Ayrıca, emisyonun tüm kum pelet örnekleri için $6.78 \mu\text{m}$, $9.24 \mu\text{m}$, $11.69 \mu\text{m}$ ve $14.05 \mu\text{m}$ 'de en yüksek pikleri gösterdiği gözlemlenmiştir. Dolayısıyla, bu çalışmada deneysel olarak incelenen kum numunelerinin özellikleri, bollukları ve daha iyi performansa sahip termal özellikleri nedeniyle radyatif soğutma için kullanılacaklarını göstermektedir.

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CHAPTER I

INTRODUCTION

1.1 Energy efficiency in buildings

The operation of buildings accounts for 30% of global energy consumption and nearly 30% of total greenhouse gas emissions from the energy sector [1]. Cooling in conventional air-based active cooling systems requires a significant amount of energy consumption, which has a huge negative impact on climate change due to the carbon and other greenhouse gas emissions it produces [2]. In addition, it is reported that by reducing the cooling demand by only 1% across the United States, the annual carbon dioxide emissions from electricity production will decrease by 118 thousand metric tons [3]. Various aspects and effective practical steps should be applied to minimize the negative impact of the recent crises and to minimize the demand for air conditioning and energy consumption in the future. Technology and infrastructure should take the responsibility to create sustainable solutions for the increasing cooling demand and to reduce energy consumption in buildings through energy management and by achieving thermal efficiency in building design. Passive cooling can play a significant role in reducing or even eliminating space cooling, especially in buildings, and reducing the negative impact on the environment.

1.2 Passive radiative cooling for buildings

Radiative cooling is one of the most innovative approaches to reducing human dependence on energy-inefficient cooling methods in buildings and industry [4, 5]. Radiative cooling is an approach to cooling objects even below ambient air temperature without external energy consumption and greenhouse gas emissions [6, 7]. In general, cooling

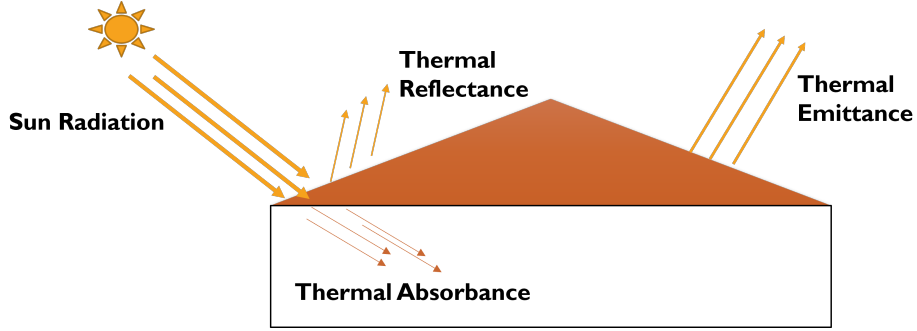


Figure 1: Behavior of incident rays on roof surfaces

problems are more challenging than heating problems due to the second law of thermodynamics [8]. Thus, during the last few decades, there have been many studies investigating different structures and methods to improve radiative cooling systems [8, 9, 10]. For example, by using appropriate solar reflectors or changing the surface properties of the materials used in buildings, material properties such as absorptivity, emissivity, and reflectivity can be improved to achieve passive radiative cooling. Passive radiative cooling is a sustainable approach that can help minimize the economic and environmental burden of cooling on our planet. Figure 1 shows a schematic of the behavior of incident radiation on roof surfaces.

A passive cooling system is an essential factor in reducing the need for space cooling and thus the negative impact of the building on the environment. Passive cooling systems are non-mechanical systems that can create a comfortable indoor temperature. The main process of passive cooling systems is the creation of an energy exchange between the system and the deep space. It is based on the fact that the atmosphere has a transparent region that is very similar to an extremely cold sink and is called the atmospheric window. The atmospheric window is a transparency window of the atmosphere for electromagnetic waves between 8-13 μm , which means that almost all the radiant energy in this region can be lost to the sky [11]

Nighttime passive radiative cooling has also been studied for many decades [12]. However, research on daytime radiative cooling has gained significant momentum in

the last two decades in cooling technology and has become increasingly relevant due to the development of nanotechnology and metamaterials with their wide potential applications and promising fundamental implications [13]. For surfaces that have high absorptivity within the UV and visible spectrum, their ability to absorb most solar radiation is very high, and they can have absorption similar to blackbody absorption. The spectral energy distribution of radiation emitted by a black body, which absorbs almost all of the radiant energy, is described by Planck’s law [14, 15]. Since the black body is a perfect emitter, its equilibrium temperature can be determined by applying the radiative energy balance to it. In the case where the surface is considered to be in a vacuum, its equilibrium temperature can be determined by pure radiative transfer analysis. However, if the surface is not in a vacuum, both conduction and convection heat transfer must be considered and added to the calculations [15]. For daytime radiative cooling, it is important to consider solar absorption and reflect more than 85% of incoming solar radiation to achieve a daytime radiative cooling equilibrium temperature below ambient [2, 16].

Passive radiative cooling can be achieved through various structures, such as the use of highly reflective coatings. In addition, making a surface that has a large absorptivity in the short-wavelength spectral region around the peak of solar energy, but a small absorptivity in the longer-wavelength spectral region can make it absorb nearly as much energy as a blackbody, but emit very little [2, 17]. Figure 2 shows a schematic of heat flow in white and black surfaces on buildings.

Spectrally selective surfaces are commonly used to cool objects exposed to incident radiation or the sun, such as building roofs and storage tanks in space. There are two basic functions to achieve passive radiative cooling. The first is to design a spectrally selective surface that can emit in the transparent atmospheric region (8-13 μm), known as the atmospheric window, and efficiently reject thermal energy to space [16]. The other primary function is that the surface must reflect most of the incoming

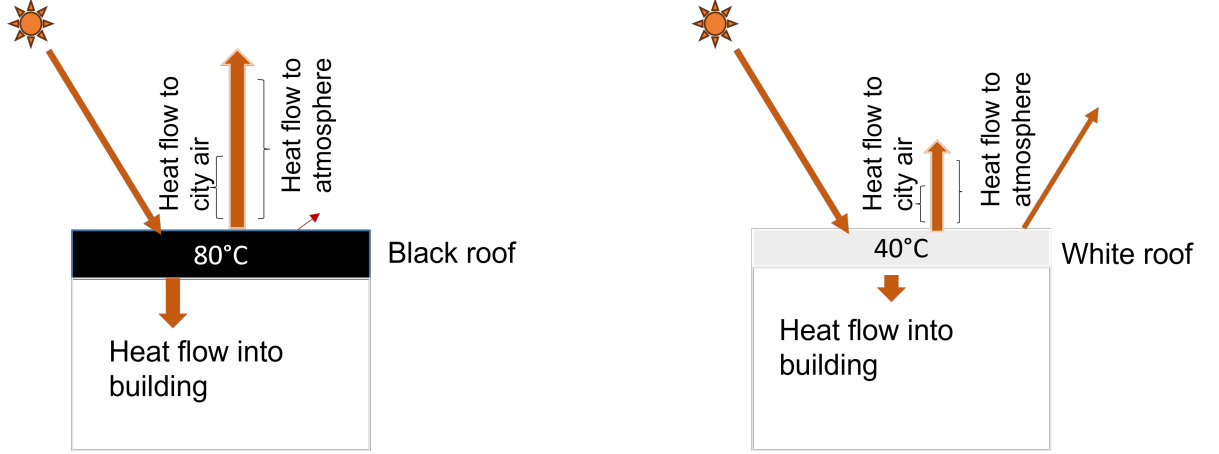


Figure 2: Schematic for heat flow in white and black surfaces on buildings

solar radiation. An example of spectrally selective surfaces is the use of white paint on roofs. White paint can reflect incident solar radiation at short wavelengths and also has a good radiative capability at long wavelengths [17].

Spectrally selective reflection can be achieved in several ways, such as using special coatings and paints or modifying material surface properties using nanoscale structures and nanoscale metal particles on surfaces. With its significant potential as one of the most promising next-generation renewable energy technologies, improving spectrally selective techniques for nighttime and daytime radiative cooling of buildings is essential for minimizing energy consumption [18].

In the case of buildings, solar radiation striking the roof can be reflected or absorbed. The absorbed heat is either transferred to the surrounding air by convection or emitted back to the sky. If a surface is to be radiatively cooled, it must have the highest reflection of solar energy in the visible wavelength range and the highest radiation emission in the atmospheric window (8-13 μm wavelength range). This technique can make a significant contribution to reducing greenhouse gas emissions by reducing the use of air conditioning in warm climates during the summer months.

1.3 Statement of the problem

Improving the energy efficiency of buildings can make a significant contribution to mitigating growing concerns about climate change. One of the main contributors to the high cooling load in buildings is heat gain during the summer months. Recently, many studies have worked on finding alternatives to space cooling and reducing the air conditioning load in buildings [19, 20, 21, 22]. Our goal in this study is to evaluate radiative cooling using some special sand particles and compare the radiative properties with other materials such as silicon dioxide, titanium dioxide, and zinc oxide. Scanning Electron Microscopy (SEM) is used to analyze the morphology of the surface of the sand particles and the other materials used in the study for comparison. Fourier Transform Infrared Spectroscopy (FT-IR) is used to measure the absorbance and the transmittance percentage of the samples and calculate the properties of the experimental work.

CHAPTER II

LITERATURE REVIEW ON RADIATIVE COOLING

2.1 History of radiative cooling

Although the concept of radiative cooling was reported in various studies in the early 19th century [23, 24], systematic studies did not begin until the 20th century [25]. In 1981, a study was carried out using aluminum coated with SiO of different thicknesses and the evaporation of SiO on smooth aluminum [26]. In the same year, another study was carried out by the same group to evaluate the radiative cooling of green leaves [27]. Several papers on radiative cooling paints and materials were published between the 1970s and the 1990s [28, 29, 30]. Each study has been a contribution to the field and an important foundation for radiative cooling. However, none of these studies has demonstrated a single-layer, substrate-independent radiative cooling paint that operates below ambient temperature for 24 hours [25].

Recent developments in nanotechnology and metamaterials over the last two decades, with their broad application potential and promising fundamental implications, have allowed studies of many types of material systems to demonstrate 24-hour sub-ambient cooling with direct solar irradiation.

One of the most notable studies in 2014 by Raman et al. [6] demonstrated a radiative cooling system that reaches 5°C below the ambient air temperature under direct sunlight. They introduced an integrated photonic solar reflector and thermal emitter consisting of seven layers of HfO₂ and SiO₂ of different thicknesses and an Ag bottom layer as a reflective metal, all deposited on a silicon wafer [6]. In 2016, a study by Ruan and his group proposed a low-cost double-layer coating embedded with particles for both nighttime and daytime radiative cooling [31]. A review of the

recent studies will be discussed in the next sections.

2.2 Sustainable materials for radiative cooling

A major benefit of using sustainable materials for radiative cooling is to protect the environment and provide alternative materials that have the lowest energy consumption. Sustainable materials can play an important role in the radiative cooling of buildings because materials such as straw, adobe, clay, or coconut palm are durable, strong, and can be replaced after a few years of use without harming the environment [2]. The different properties and cooling behavior of the materials can lead to an extension of the radiative cooling potential, allowing more applications in the field. The selected materials can also be used as a coating on the surface of the buildings.

In order for materials to be used both as solar collectors during the day and as solar radiators or radiative coolers at night, the materials must have high spectral emissivity in both the atmospheric window and the solar band. However, materials that have high spectral emissivity in the solar band but low emissivity in other bands could be considered good candidates for daytime radiative cooling, such as perlite pumice composite (PPC) and painted perlite pumice composite. Perlite has several properties that can make it an important raw material in geopolymerization technology. It has 2-5% of combined water and can expand from 10 to 30 times by increasing the temperature between 760°C and 1150°C. The pumice with a sponge-like structure has a low bulk density, low thermal conductivity, low sound transmission characteristics, and high strength-to-weight ratio, making it considered a good sustainable building material. [2, 11, 32]. The experimental results of Family and Menguc [2] emphasize that PPC and painted PPC have high spectral emissivity in the atmospheric window range but low in other bands [32].

In addition, materials that have high spectral emissivity in the solar band and low emissivity in all other bands can be considered for solar energy absorption. Black

membrane (EPDM) is an example of a sustainable material that can be used as a solar absorber. EPDM is a durable material made from propylene and ethylene, which can be derived from natural gases and oil [32, 33]. The results of the same study by Family show that the EPDM has low reflectivity and high emissivity in the solar radiation band and has shown no reflection peak in the UV light range, so it has a high absorption peak in the UV range and can be used for a solar heating model or solar collector [32].

2.3 Sustainable materials developed for passive radiative cooling systems

Passive radiative cooling has remained a challenge over the past decades due to the precise required properties of defined materials with the ability to reflect solar radiation and emit heat into deep space at about 3 K temperature. Various materials have been developed to achieve the desired optical properties for radiative cooling, thanks to recent advances and technological improvements in nanoscience and metamaterials. Simultaneously, these sustainably developed materials can be differentiated from conventional materials as they do not involve complex multilayer structures and non-sustainable polymers, and have the advantage of being renewable and environmentally friendly.

Environmentally friendly, configurable, and renewable nano-micro-structured plastics prepared by a locally confined polymerization method [34] for radiative cooling applications have been developed in a recent study. Compared with conventionally fabricated plastics used in electronic devices, the nano-micro-structured plastic achieved higher cooling efficiency. The developed structure showed a temperature drop of 8.6°C on electronic circuits and 7.5°C cooling improvements under sunlight [34]. In another study, electrospun and cast cellulose derivatives were used to develop a reflective radiative cooler and a transparent radiative cooler that reduced temperatures by up to 15°C and 5°C, respectively [35].

A cooling paper has also been reported as a low-cost, scalable, and environmentally friendly radiative cooling material that can be easily recycled [36]. Through the mature pulping and papermaking process, the cooling paper consists of dignified fibrillated cellulose fibers and nano-sized hydroxyapatite. This study [36] reported a high solar reflectance of about 94% and an infrared emissivity of about 0.95 in the atmospheric transparency window.

Another study reported that a combination of highly diffuse reflective polystyrene foam particles and fiber-based printer paper results in a reflectivity of 96% in the solar spectrum and a maximum radiative cooling power of 90 W m^{-2} . The reported foam-paper composite is produced by a simple crushing and mixing process using recycled polystyrene (PS) foam and printer paper [37]. Thus, it can indeed be stated that the recent technological improvement has opened the door for the development of materials that meet the desired optical characteristics for radiative cooling, and at the same time, these developed materials are sustainable and environmentally friendly.

2.4 Paints and coatings for radiative cooling

The purpose of using paints and coatings is to control the emission in the structure and to modify the absorption coefficient, scattering coefficient, and refractive index of paint or coating materials to achieve changes in spectral reflectance and emissivity. Paints consist of resin and solid pigments, which can be metal oxides and semiconductors. The semiconductors and doped semiconductors have selective absorption and can allow frequencies lower than the band gap for absorption. Various pigments have been used for radiative cooling, such as silicon (SiC) [38], carbon (C) [39, 40], Zinc sulfide (ZnS) [29, 41], titanium dioxide (TiO_2) [31], and barium sulfate (BaSO_4) [42].

In addition to paints, another fundamental approach to emission control is the use of selective coatings. Reducing the reflectivity of a surface by changing its properties

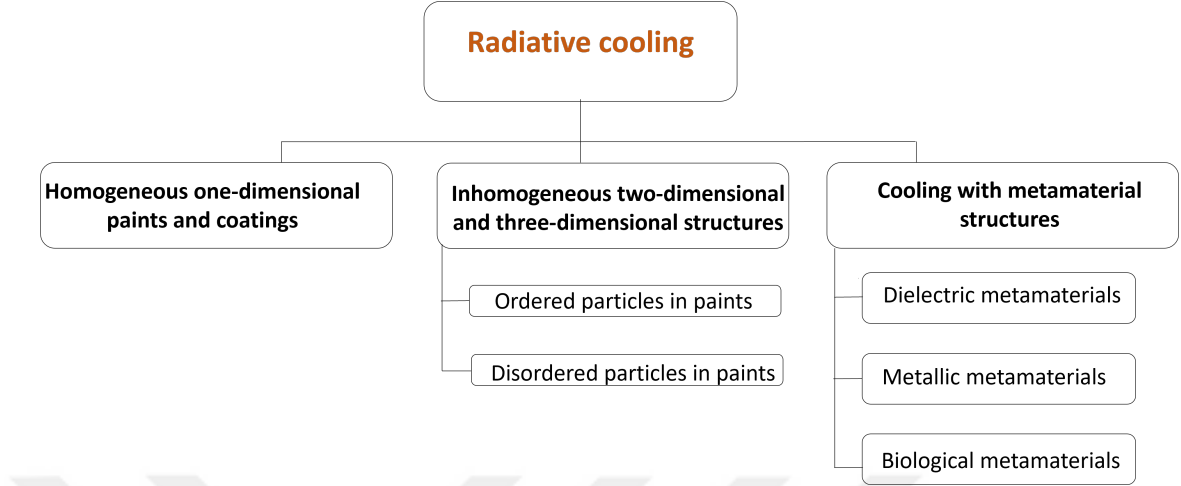


Figure 3: Diagram for passive cooling of buildings characterization

can improve its spectral absorption and emissivity. The use of metal films for coatings in radiative cooling can provide superior results due to their high reflectivity in the infrared and their semi-transparency in the visible. The coating can also show effective results with thin films such as nitrides, oxides, oxynitrides, and sulfides of silisium and silicon [2, 17]. In Figure 3, a diagram for passive cooling of buildings characterization. As shown in the diagram, radiative cooling can be achieved through different methods that can be divided into three categories: homogeneous one-dimensional paint and coating, two-dimensional and three-dimensional structures, and cooling with metamaterial structure.

As an early study showing high emissivity at the atmosphere window, TiO_2 particles were studied in the form of pigmented paints by coating an optically thick layer of white paint with 35% TiO_2 on an aluminum plate [43]. The cooling performance was improved by a further reduction of 3.2° Celsius by adding BaSO_4 to the paint [43]. Recently, two-layer coatings with TiO_2 nanoparticles as the top layer and SiC/SiO_2 nanoparticles as the bottom layer have shown significant temperature reduction [44]. However, due to the strong intrinsic absorption of TiO_2 in the UV (because of the 3.2 eV electron band gap of TiO_2), they have been replaced by SiO_2 particles, which

have shown better performance for nighttime cooling [44].

2.5 Radiative cooling with ordered particles in paints and coatings

Spectrally selective surface properties can also be achieved with recent advances in nanotechnology by creating selective emissivity, which is difficult to find in natural materials. Nanoscale structures and nanoscale metal particles embedded in a semiconductor matrix, for example, can be applied to surfaces. The reflection index of the material must have maxima or minima with respect to frequency to achieve the required selectivity in emission and absorption spectra [2, 17]. Reducing the size of the particles can lead to a decrease in reflectance. This allows the method of nanoparticle application to be effective for radiative cooling [44]. Thus, the large absorption cross-section of the nanoparticles allows them to absorb more than the incident light [44]. Various materials have been used for both day and night radiative cooling, such as silicon nanowires, copper oxide, titanium dioxide, and barium sulfate, to achieve spectrally selective properties.

A recent study by Ruan (2021) has demonstrated all-day cooling with a new paint that can reflect 98% of the sunlight as well as radiate infrared heat into space. Due to the high electron band gap for low solar absorption and the phonon resonance at 9 μm for high sky window emissivity, the sub ambient cooling performance was investigated with BaSO_4 nanoparticle films and BaSO_4 nano-composite paints. They achieve a high solar reflectance of 97.6% and a high sky window emissivity of 0.96, indicating surface temperatures more than 4.5°C below ambient temperatures [45]. By developing the BaSO_4 acrylic paint with a 60% volume concentration and a wide particle size distribution, reliability was improved to achieve a solar reflectance of 98.1% and a sky window emissivity of 0.95 [45].

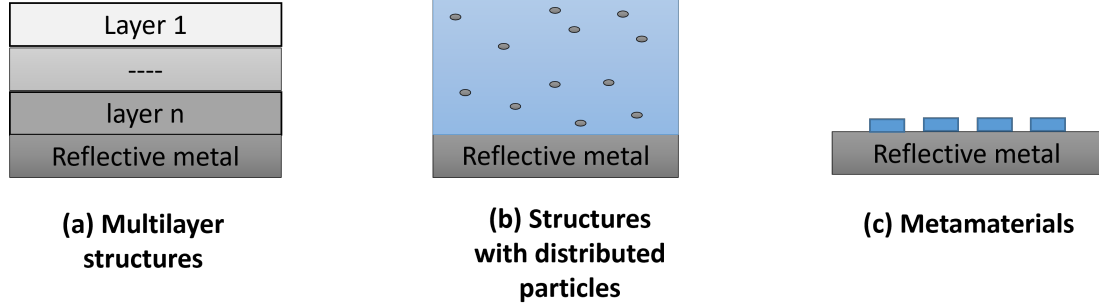


Figure 4: Three groups of radiative cooling materials with different structures: (a) multilayer structures, (b) distributed particle structures, and (c) metamaterials. Adapted from: [12]

2.6 Photonic doubled and multilayer structures for radiative cooling

Devices based on photonic structures have spurred a renewed interest in highly efficient cooling systems and have recently led to the achievement of full daytime sub-ambient cooling capability below ambient temperature. This can be achieved through various structures such as multilayer thin film structures, micro- and nanoparticles, photonic crystals, metamaterials, and bio-inspired photonic structures. The multilayer thin film structure can be ideal for proof-of-principle demonstration because of the scalable fabrication and experimental realization of the structure [44]. Figure 4 shows the three groups of radiative cooling materials with different structures: 4(a) multilayer structures, 4(b) distributed particle structures, and 4(c) metamaterials.

In a previous work by Ruan (2016), a cost-effective two-layer coating embedded with particles was proposed for both nighttime and daytime radiative cooling [31]. These two layers were acrylic resin with titanium dioxide embedded in one layer and carbon black particles embedded in the other layer. The top layer of titanium dioxide, which has high reflectivity in the solar spectrum and low reflectivity in the sky window, is used to reflect the solar radiation. The bottom layer of carbon black particles is responsible for heat dissipation in the sky window, which is designed to have high emissivity in the sky window to remove heat from the surface. The results

have shown that the coating embedded with particles of $0.2\ \mu\text{m}$ radius has a solar radiation reflection of 90%, and the average emissivity is greater than 0.9 for most directions in the atmospheric transparency window[31]. It was also concluded that the coating used can achieve a net cooling power of $100\ \text{W m}^{-2}$ and more than $180\ \text{W m}^{-2}$ at ambient temperature under direct solar irradiation and at night, respectively. They reported that the coating can achieve an equilibrium temperature of 278 K in the daytime and 255 K at nighttime [31].

The integrated photonic structure is a type of multilayer radiative cooling material. As shown in Figure 4(a), it consists of multiple layers with the reflective metal as the bottom layer. A study by Raman (2014) has demonstrated radiative cooling to 5°C below the ambient air temperature under direct sunlight. They presented an integrated photonic solar reflector and thermal emitter consisting of seven layers of HfO_2 and SiO_2 of different thicknesses and an Ag bottom layer as reflective metal. All layers are deposited on a silicon wafer as shown in Figure 5(a). The bottom four layers are designed to optimize solar reflection, while the top three layers are responsible for thermal radiation through the atmospheric window. HfO_2 serves as a high index material and has low ultraviolet absorption, which is an important property for optimizing solar reflectance. An alternative to HfO_2 is titanium dioxide, which is less expensive. SiO_2 is the layer with a low refractive index and it is optically transparent. All of these layers combined with the material properties and interference effects result in a device that emits strongly in the atmospheric window as shown in Figure 5(b) and reflects 97% of the sunlight as shown in Figure 5(c). [6].

Another study by Gentle and Smith (2015) achieved a high reflectivity of 0.97 in the solar band and 0.96 emissivity over the atmospheric window [7]. A multilayer composite of polymer pairs with silver underlining and evaluated at $1060\ \text{W m}^{-2}$ achieved a surface temperature 2°C below ambient. Another study has investigated the relationship between the performance of passive daytime radiative cooling and the

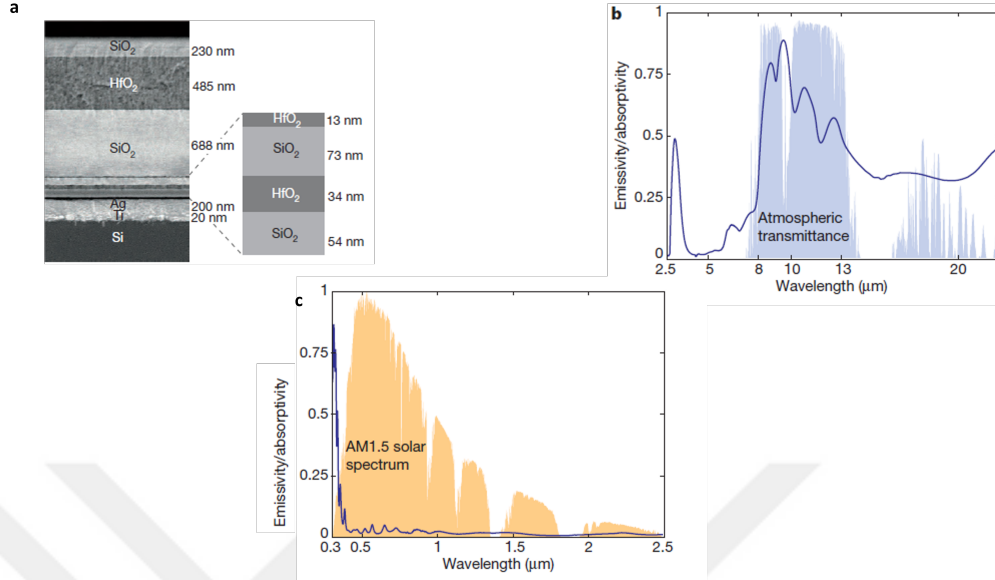


Figure 5: (a) Scanning electron microscope image of the photonic radiative cooler that consists of seven layers of HfO_2 and SiO_2 . (b) Measured emissivity/absorptivity of the cooler at a 5° angle of incidence over mid-infrared wavelengths using an unpolarized light source. (c) Measured emissivity/absorptivity at 5° angle of incidence of the photonic radiative cooler over optical and near-infrared wavelengths using an unpolarized light source. Adapted from [6]

dielectric micro-sphere coatings parameters such as radius, thickness, and fill rate [46]. They have presented that solar reflectance and thermal emittance can be improved by increasing the thickness of the micro-sphere coatings, and the dielectric sphere in the air environment can scatter more light in the solar spectrum.

Although the designed spectral emissivity can be achieved by the nanophotonic approach, the strict fabrication requirements and the prohibitive cost are the reasons for limiting the applications based on this structure. As shown in Figure 4 (b), a distributed particle structure is still a promising approach because of its comparatively simple and economical fabrication requirements [12]. The distributed particle layer is used to tailor the infrared emissivity to the atmospheric window, and the reflective metal at the bottom is responsible for the high reflectivity.

A recent study has shown a good reflection of solar radiation of 96% and an emissivity of 0.93 across the atmospheric window, which results in a cooling power of

93 W m^{-2} [47]. SiO₂ microspheres were distributed in a polymethylpentene matrix material backed by a thin film of silver, and the interaction of the SiO₂ microspheres with the infrared light caused the material to have a high emissivity across the entire atmospheric window. Different substrates have been used in the literature such as SiO₂ [47]; TiO₂ [48, 49]; and BaSO₄ [45]. The photonic structures are mainly limited to paint applications, and they require a reflective metal at the bottom to reflect sunlight, which could increase the difficulty and cost of the fabrication process in some cases. Thus, some recent studies have shown that using light-scattering air voids in a polymer instead of reflective metal can reduce the cost and increase the optical performance [12].

Another recent study has shown promising results by using a polymer-based porous structure to fabricate hierarchical porous poly coatings with exceptionally good passive daytime radiative cooling capability [49]. In this study, the pore sizes were 5.5 μm for micropores and 0.2 μm for nanopores, where the pores were formed after evaporation of water. While this technique is as simple as paint, the results showed a high solar reflectance of 0.96 and long wavelength infrared emittance of 0.97, allowing for sub-ambient temperature drops of 6°C and cooling powers of 96 W m^{-2} .

In a recent study, the use of a periodic pyramid-textured polydimethylsiloxane film for radiative cooling of silicon solar cells remarkably improved performance. The spectral test the researchers have conducted shows that the film emits as a black body, while the outdoor cooling test showed that the film can be cooled by 2°C, which can be effective for photovoltaic applications [43].

CHAPTER III

RADIATIVE PROPERTIES AND THEORETICAL ANALYSIS

3.1 Fundamental properties of particles

To understand the impact of different particle systems on radiative cooling and energy efficiency in buildings, we need to understand the fundamental properties of particles. To do this, we need to know how particles and substrates interact with instantaneous electromagnetic radiation at a given wavelength. The interaction of electromagnetic waves with various structures and surfaces has been the subject of many texts and research monographs since Maxwell summarized the concept of electromagnetic wave scattering in mathematical terms.

The complex refractive index and size are the two non-dimensional independent parameters that govern the radiative properties of a single spherical particle of radius r interacting with an electromagnetic wave of wavelength λ . The complex refractive index and the size parameter are calculated using formulas 1 and 2, respectively [15]:

$$\dot{n} = n - ik \quad (1)$$

$$x = \frac{2\pi r}{\lambda} \quad (2)$$

Absorption and scattering occur when light interacts with a droplet or particle, and the incident electromagnetic wave energy emitted during the interaction is either converted to internal thermal energy of the particle by absorption or scattered in all directions [15]. Absorption and scattering are affected by the shape and size of the particle and its optical properties. There are several examples in the Earth's



Figure 6: Diagram shows (a): a multilayer structure, (b): a multilayer with particles.

atmospheric system of light scattering and absorption by spherical particles in an absorbing medium, such as light scattering by air bubbles in sea ice and by cloud particles surrounded by water vapor in the atmosphere [50]. Light scattering by a spherical particle in a non-absorbing medium is studied based on the Lorenz-Mie theory. However, the Mie equations are modified to describe an absorbing medium.

In the same study discussed in the previous chapter by Ruan (2016), the effect of titanium particle size was investigated with radii $r=0.1 \mu\text{m}$, $0.2 \mu\text{m}$, $0.3 \mu\text{m}$, and $0.4 \mu\text{m}$, and a coating thickness of $500 \mu\text{m}$. By increasing the particle size, the scattering coefficient showed less dependence on the wavelength. In addition, the increase in particle size resulted in an increase in long wavelength reflectivity, but a decrease in the visible band [31]. Different complex refractive indices were observed for particles with parallel and perpendicular polarization.

In addition, due to the small size of the particles, they are not visible at long wavelengths, so the reflectivity showed exceptionally low values in the sky window and high emissivity in the spectral range. The coating has reflected about 91% of the solar energy and has a high emissivity of more than 0.95 in the sky window [31]. Figure 6 shows a diagram for two structures, Figure 6 (a): a multilayer structure, and Figure 6 (b): a multilayer structure with particles. In the following section, the Lorenz-Mie theory is discussed. In order to study the interaction of light with the particles, the discussion of Lorenz-Mie theory is necessary.

3.2 Lorenz-Mie Theory

The Lorenz-Mie theory is a powerful mathematical model based on a series solution of Maxwell's equations for describing the interaction of an electromagnetic wave λ and a spherical particle of diameter d , with a complex index of refraction $m = n + iK$ [25]. The theory was first developed by Gustav Mie in 1908 [25, 15]. The theory has been used in various fields, including optics, remote sensing, and atmospheric science. The mathematical explanation of the Lorenz-Mie theory is given in the following sections. There are two unidimensional parameters needed to calculate the Lorenz-Mie theory, which are the normalized complex refractive index, $m = \frac{n+iK}{N_0}$, and the size parameter, $x = \frac{\pi d}{\lambda}$. Thus, it allows us to calculate the absorption and scattering cross sections of particles based on their size and refractive index. First, the Mie coefficient can be calculated using the following formulas [25, 33]:

$$a_n = \frac{m j_n(mx) j'_n(x) - j_n(x) j'_n(mx)}{m j_n(mx) h'_n(x) - h_n(x) j'_n(mx)} \quad (3)$$

$$b_n = \frac{j_n(mx) j'_n(x) - m j_n(x) j'_n(mx)}{j_n(mx) h'_n(x) - m h_n(x) j'_n(mx)} \quad (4)$$

where:

$x = kr$ (size parameter)

$m = \frac{n_p}{n_m}$ (relative refractive index)

j_n and j'_n are spherical Bessel functions of the first kind and their derivatives

h_n and h'_n are spherical Hankel functions of the first kind and their derivatives

n = mode index (order of the spherical functions)

After calculating the Lorenz-Mie coefficient, the scattering efficiency and extinction efficiency can be calculated using the following formulas[33]:

$$Q_{\text{scat}} = \frac{2}{x^2} \sum_{n=1}^{\infty} (2n+1) (|a_n|^2 + |b_n|^2) \quad (5)$$

$$Q_{\text{ext}} = \frac{2}{x^2} \sum_{n=1}^{\infty} (2n+1) \text{Re}(a_n + b_n) \quad (6)$$

By taking the difference between the extinction efficiency and scattering efficiency the absorption efficiency can be calculated as in the next formula[25]:

$$Q_{\text{abs}} = Q_{\text{ext}} - Q_{\text{scat}} \quad (7)$$

After calculating the scattering efficiency, extinction efficiency, and absorption efficiency, the scattering coefficient and absorption coefficient can be calculated using the following formulas:

$$\sigma = N_t^T \times Q_{\text{scat}} \times \pi \times r^2 \quad (8)$$

$$\kappa = N_T \times Q_{\text{abs}} \times \pi \times r^2 \quad (9)$$

where:

σ : the scattering coefficient

κ : the absorption coefficient

N_T : the total number of particles per unit volume

Q_{scat} : the scattering efficiency

Q_{abs} : the absorption efficiency

r : the radius of the particle

To calculate the scattering amplitude[25]:

$$S_1(\cos \theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} (a_n \pi_n + b_n \tau_n) \quad (10)$$

$$S_2(\cos \theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} (a_n \tau_n - b_n \pi_n) \quad (11)$$

where:

a_n are the Mie scattering coefficients,

b_n are the Mie extinction coefficients,

π_n is the direction-dependent function associated with scattering,

τ_n is the direction-dependent function associated with absorption,

$P_n(\cos \theta)$ are the Legendre polynomials of order n ,

θ is the scattering angle.

TO calculate the scattering phase function for a single particle[25]:

$$P(\theta) = \frac{2|S_1|^2 + |S_2|^2}{x^2 Q_{\text{scat}}} \quad (12)$$

For a single particle, the asymmetric parameter can be calculated then, which is the average of the phase function and it shows the amount of the forward and backward scattering in the system.

3.3 Calculations for different types of particles

In the previous section, the mathematical explanation of the Lorenz-Mie theory was explained. From a physics and electromagnetics standpoint of view, Lorenz-Mie theory, a fundamental concept in scattering theory, explains the behavior of electromagnetic waves interacting with particles. Within this framework, the absorption cross-section represents the portion of the incident radiation that is absorbed by the particle and contributes to its energy conversion. The extinction cross-section combines absorption and scattering and includes the total attenuation of light due to particle interactions. The scattering cross section specifically quantifies the radiation scattered by the particle, describing its role in redirecting incident light. In addition, the relative permittivity governs the particle's response to an external electric field,

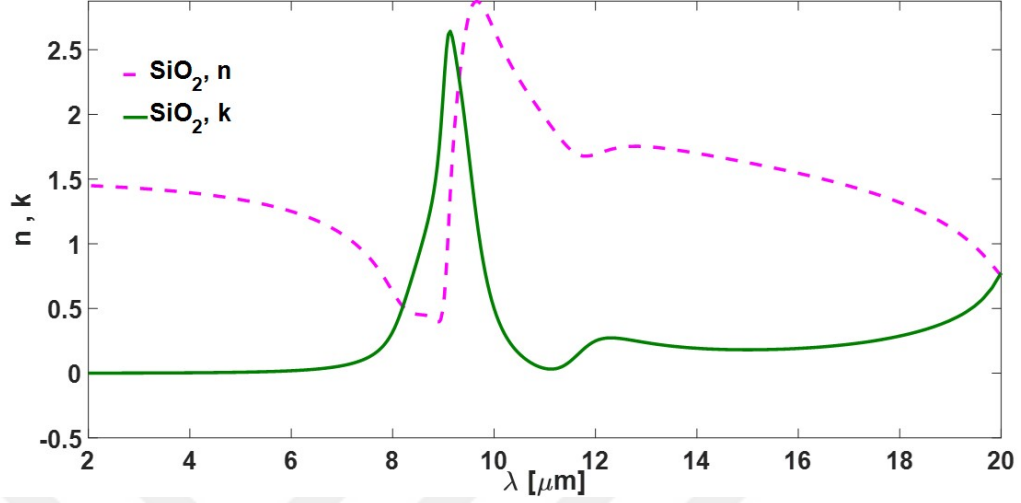


Figure 7: Index of refraction profile of SiO₂

affecting the scattering and absorption characteristics predicted by Lorenz-Mie theory. These parameters, critical to understanding particle behavior, provide insight into the intricate interplay between particles and electromagnetic radiation. As it was explained in the previous section, Lorenz-Mie theory is based on a series of solutions to the Maxwell equations for describing the interaction of electromagnetic wave λ , and spherical particle of diameter d , with a complex index of refraction. For this, the index of refraction profiles are presented in the next figures. Figures 7, Figure 8, and Figure 9 show the index of refraction profiles of SiO₂, TiO₂, and ZnO, respectively.

In addition, Figure 10 shows a comparison of k profiles for SiO₂, TiO₂, and ZnO. Figure 11 present a comparison profile of n for SiO₂, TiO₂, and ZnO as well.

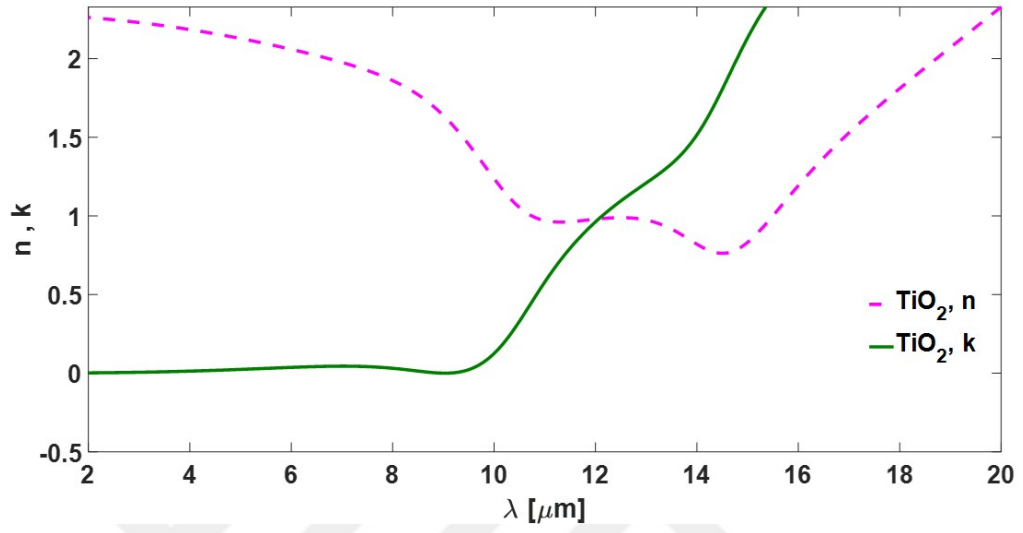


Figure 8: Index of refraction profile of TiO_2

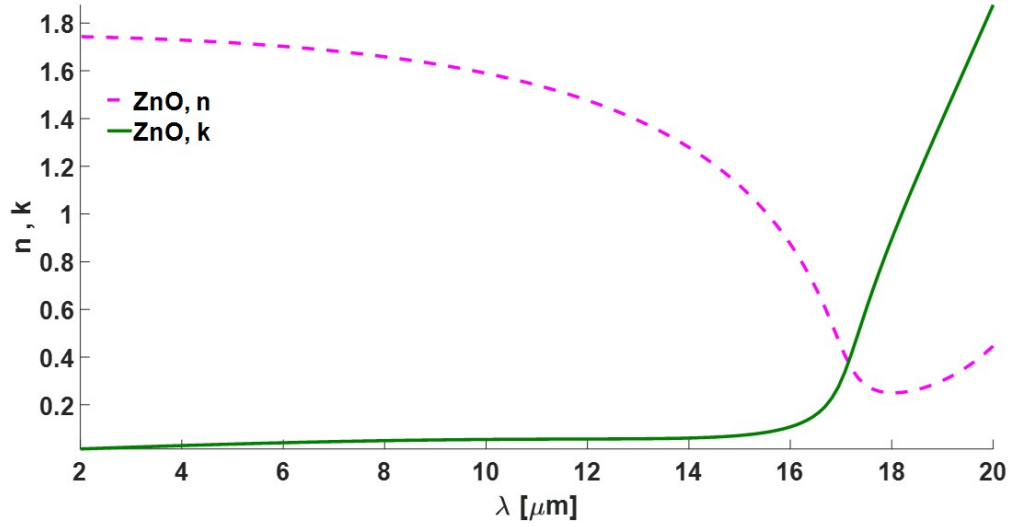


Figure 9: Index of refraction profile of ZnO

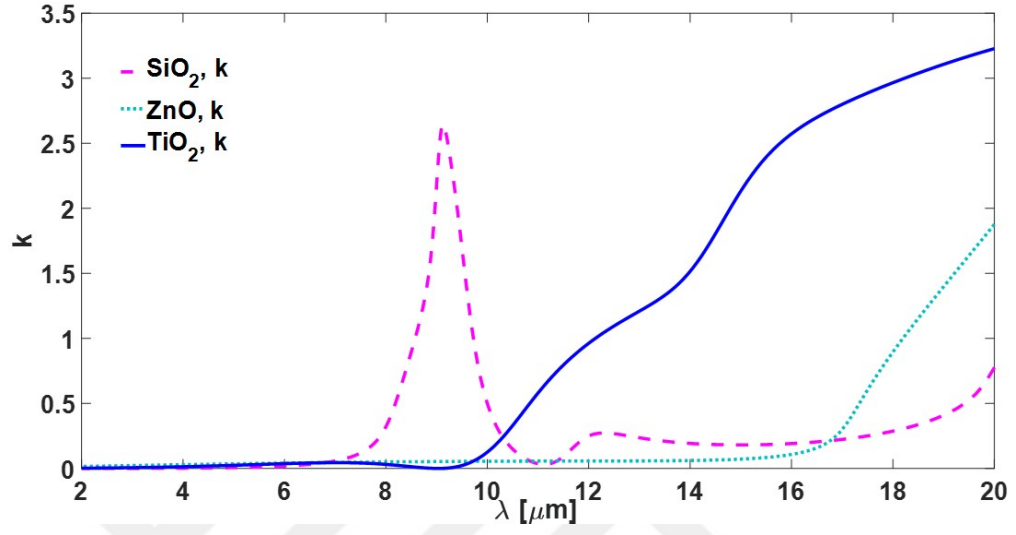


Figure 10: A comparison of k profiles for SiO_2 , TiO_2 , and ZnO

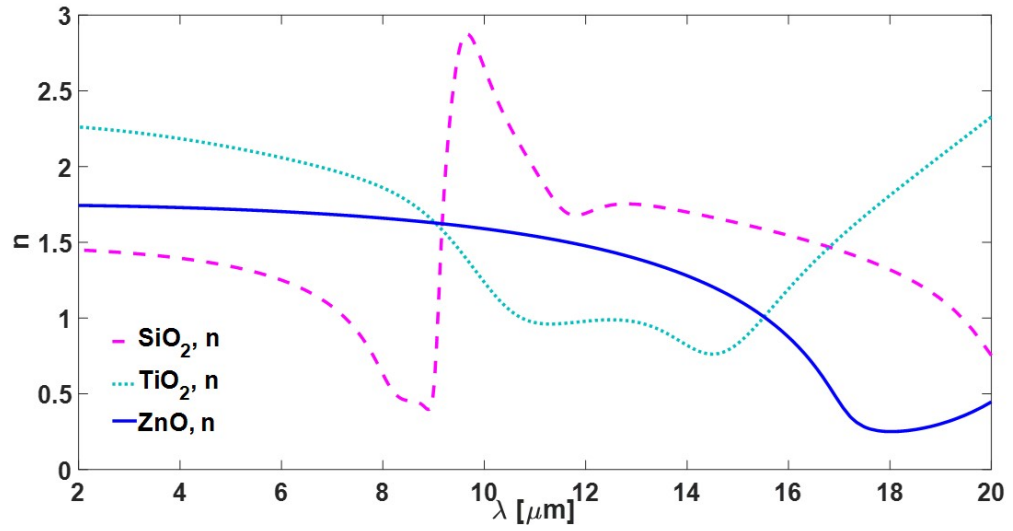


Figure 11: A comparison of n profiles for SiO_2 , TiO_2 , and ZnO

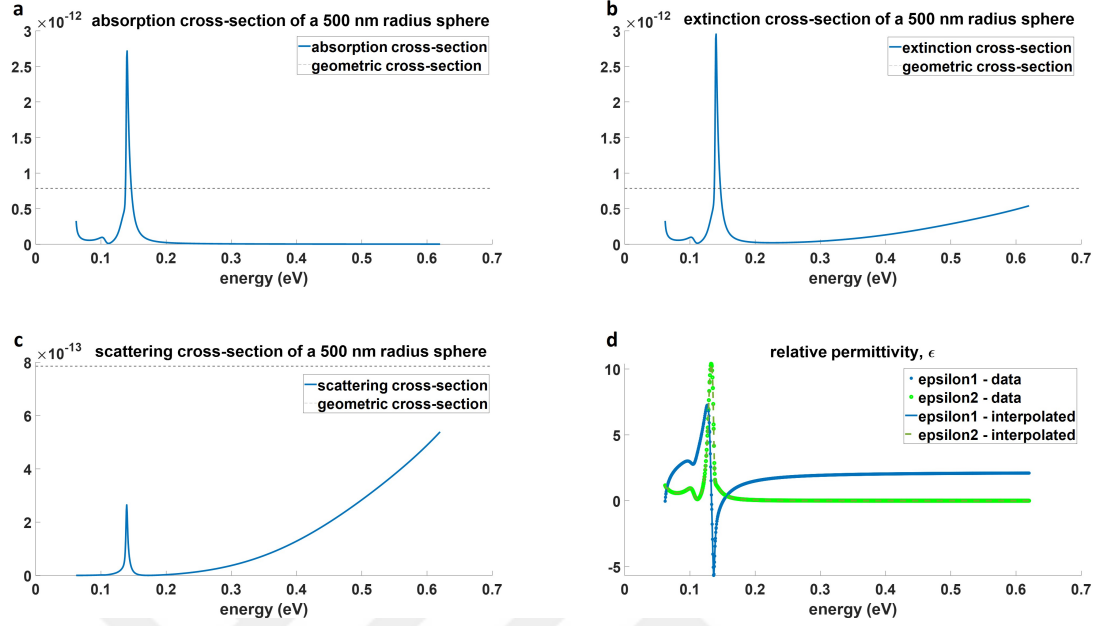


Figure 12: (a): absorption cross-section, (b): extinction cross-section, (c): scattering cross-section, and (d): relative permittivity of spherical SiO_2 particle of radius of 500 nm

In this section, the results of implementing Lorenz-Mie theory to calculate absorption cross-section, scattering cross-section, extinction cross-section, and showing the relative permittivity of SiO_2 , TiO_2 , and ZnO with a spherical radius of 500 nm are shown. Figure 12 depicts these results for a SiO_2 particle of radius 500 nm. Similarly, Figures 13 and 14 present these results for TiO_2 and ZnO , respectively.

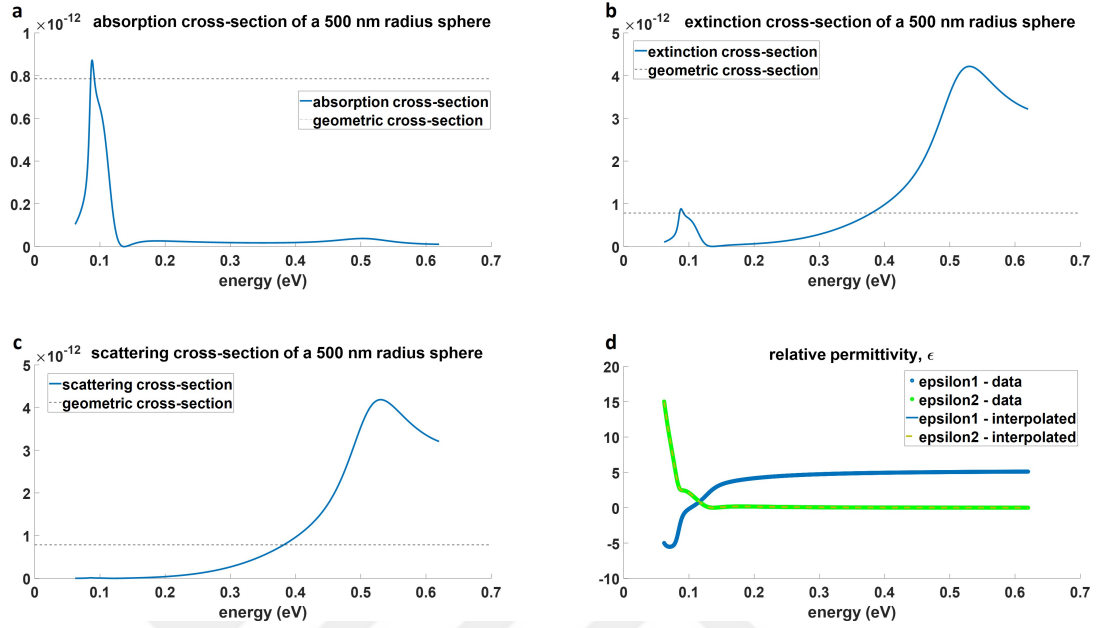


Figure 13: (a): absorption cross-section, (b): extinction cross-section, (c): scattering cross-section, and (d): relative permittivity of spherical TiO_2 particle radius of 500 nm

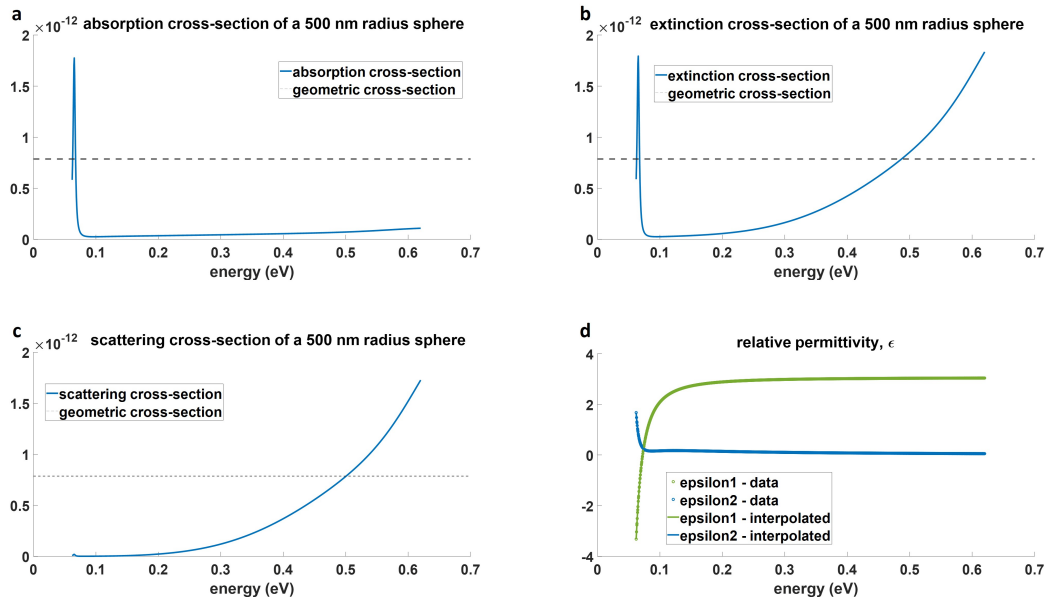


Figure 14: (a): absorption cross-section, (b): extinction cross-section, (c): scattering cross-section, and (d): relative permittivity of spherical ZnO particle of radius of 500 nm

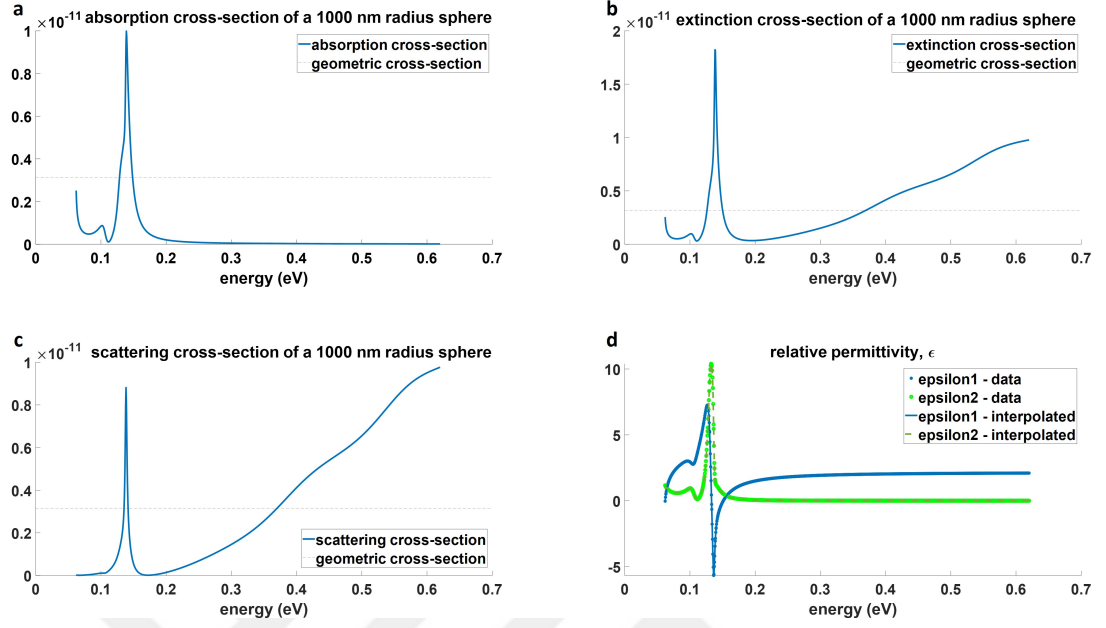


Figure 15: (a): absorption cross-section, (b): extinction cross-section, (c): scattering cross-section, and (d): relative permittivity of spherical SiO_2 particle of radius of 1000 nm

In addition, the results of implementing Lorenz-Mie theory to calculate absorption cross-section, scattering cross-section, extinction cross-section, and showing the relative permittivity of SiO_2 , TiO_2 , and ZnO with a spherical radius of 1000 nm are shown. Figure 15, 16, and 17 depicts these results for SiO_2 , TiO_2 , and ZnO particle, respectively.

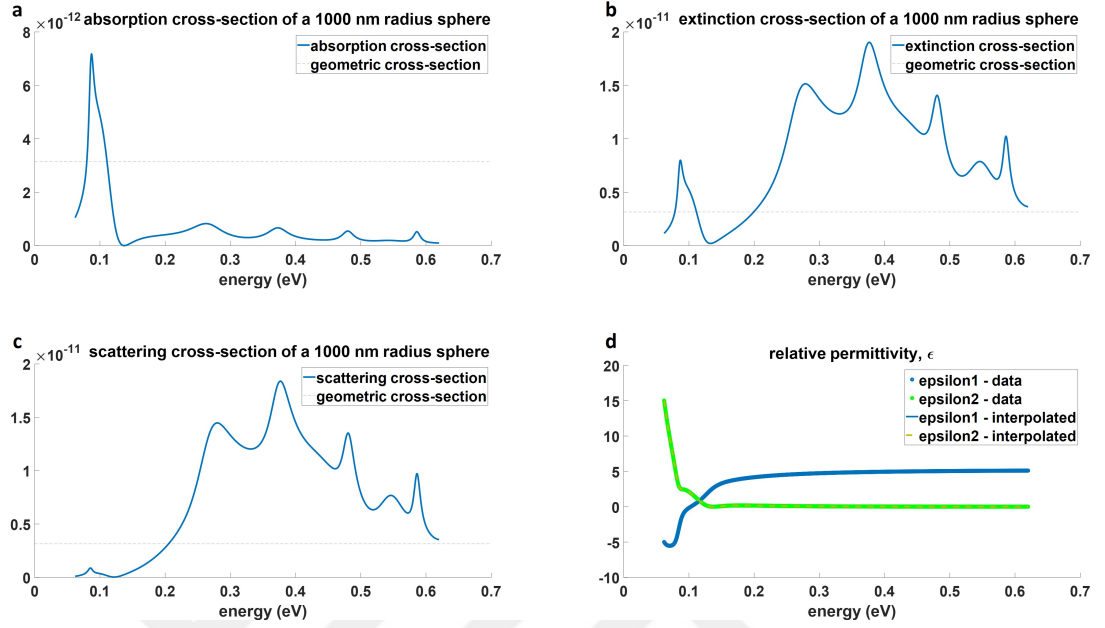


Figure 16: (a): absorption cross-section, (b): extinction cross-section, (c): scattering cross-section, and (d): relative permittivity of spherical TiO_2 particle radius of 1000 nm

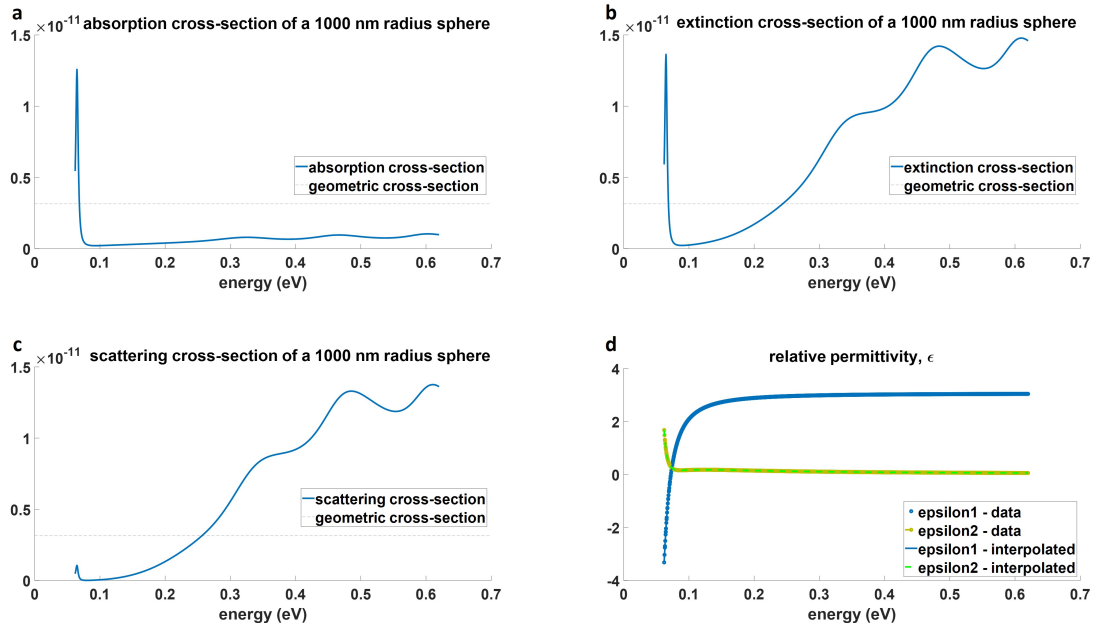


Figure 17: (a): absorption cross-section, (b): extinction cross-section, (c): scattering cross-section, and (d): relative permittivity of spherical ZnO particle of radius of 1000 nm

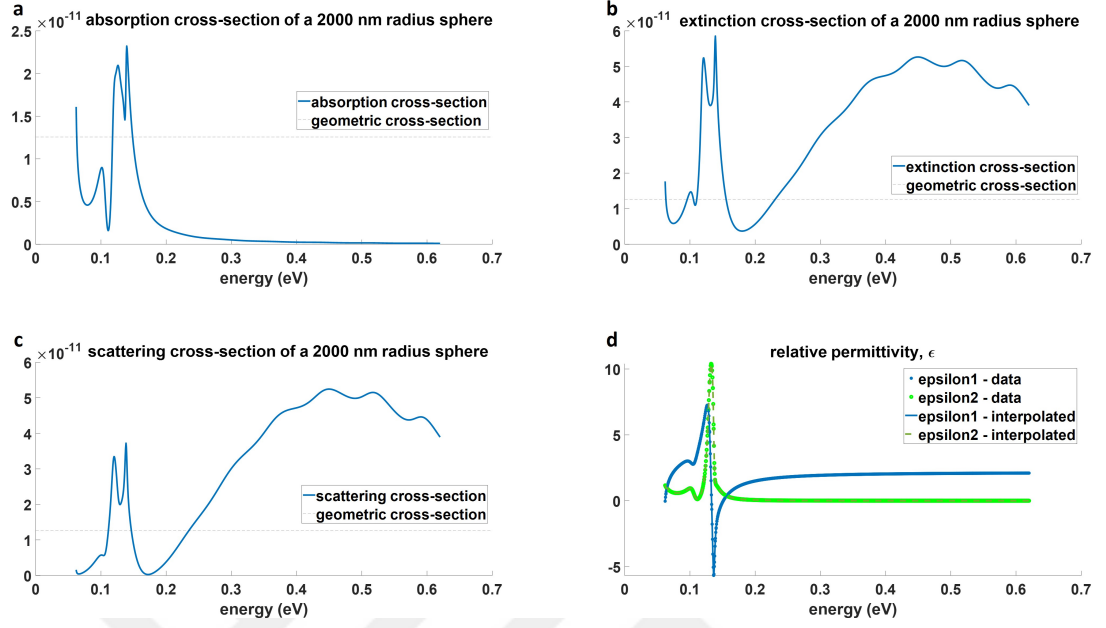


Figure 18: (a): absorption cross-section, (b): extinction cross-section, (c): scattering cross-section, and (d): relative permittivity of spherical SiO_2 particle of radius of 2000 nm

Similarly, the results of implementing Lorenz-Mie theory to calculate absorption cross-section, scattering cross-section, extinction cross-section, and showing the relative permittivity of SiO_2 , TiO_2 , and ZnO with a spherical radius of 2000 nm are shown. Figure 18, 19, and 20 depict these results for SiO_2 , TiO_2 , and ZnO particles, respectively.

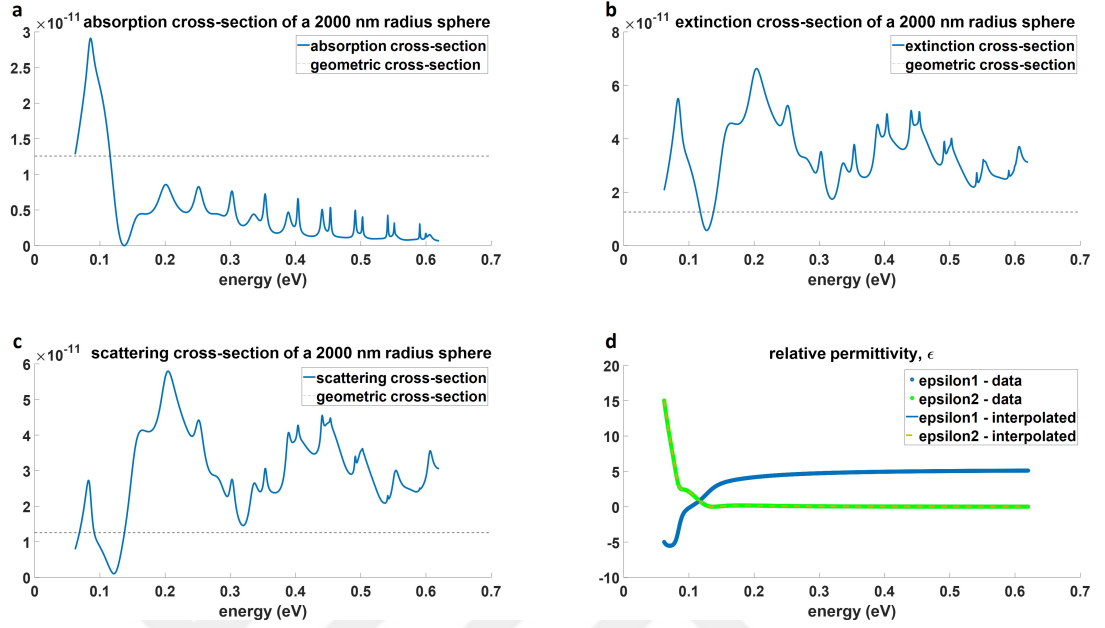


Figure 19: (a): absorption cross-section, (b): extinction cross-section, (c): scattering cross-section, and (d): relative permittivity of spherical TiO_2 particle radius of 2000 nm

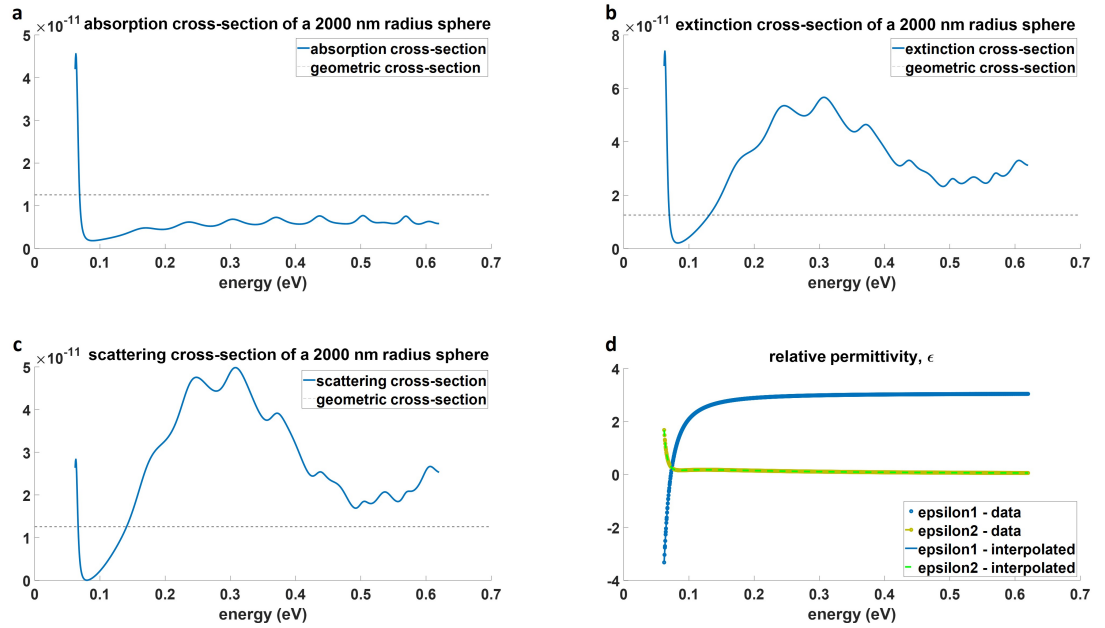


Figure 20: (a): absorption cross-section, (b): extinction cross-section, (c): scattering cross-section, and (d): relative permittivity of spherical ZnO particle of radius of 2000 nm

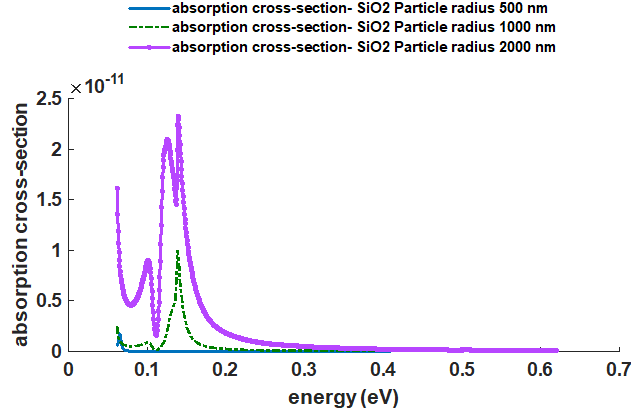


Figure 21: Comparison of particle size-SiO₂ absorption cross-section

In Lorenz-Mie theory, the absorption cross section for particles is affected by their size due to a couple of key factors. First, as particle size increases, the total surface area available for interaction with incident radiation increases. This increased surface area increases the probability of photon-particle interactions, resulting in increased absorption. Second, larger particles exhibit more pronounced resonances in their interaction with electromagnetic waves. These resonances, often associated with specific wavelengths of light, become more pronounced with increasing particle size, increasing the absorption cross-section. Consequently, larger particles tend to have higher absorption cross sections according to Lorenz-Mie theory due to their increased surface area and resonance effects.

We here present the results for comparison of the difference in absorption cross-section profiles of SiO₂ particles of 500 nm, 1000 nm, and 2000 nm radius. It can be seen that as the particle's radius size increases, the absorption cross-section increases, which matches the theoretical predictions based on Lorenz-Mie theory. Figure 21 shows these results.

Increasing particle size in Loren-Mie theory results in a larger scattering cross section for several fundamental reasons. First, larger particles present a larger physical area for incident electromagnetic waves to interact with, increasing the probability

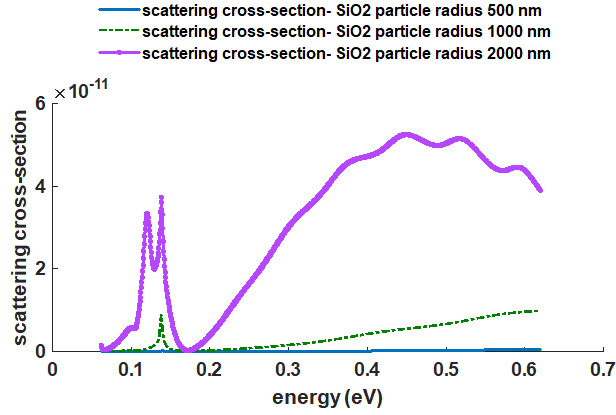


Figure 22: Comparison of particle size-SiO₂ scattering cross-section

of scattering events. Second, larger particles exhibit more pronounced and defined resonances in their interaction with light, resulting in stronger scattering effects at certain wavelengths. These resonances, such as Mie resonances, become more pronounced with increasing particle size, resulting in larger scattering cross sections. In addition, larger particles tend to scatter light more in the forward direction (i.e., less in the backward direction), further contributing to the overall increase in scattering cross section as predicted by Lorenz-Mie theory. Overall, the larger size of particles increases their scattering capability by providing a larger interaction surface and more pronounced resonances with incident electromagnetic waves.

In Figure 22, we present the scattering cross-section results for different SiO₂ particle sizes of 500 nm, 1000 nm, and 2000 nm.

In addition, in Figure 23 and in Figure 24, We present the results for comparison of the difference in absorption cross-section profiles of TiO₂ particles of 500 nm, 1000 nm, and 2000 nm radius, and scattering scattering cross-section profiles, respectively. It can be noted that the increase of the particle's radius size led to an increase in the absorption cross-section as well.

Similarly, Figure 25, and Figure 26 show the results for comparison of the difference in absorption cross-section profiles of ZnO particles of 500 nm, 1000 nm, and

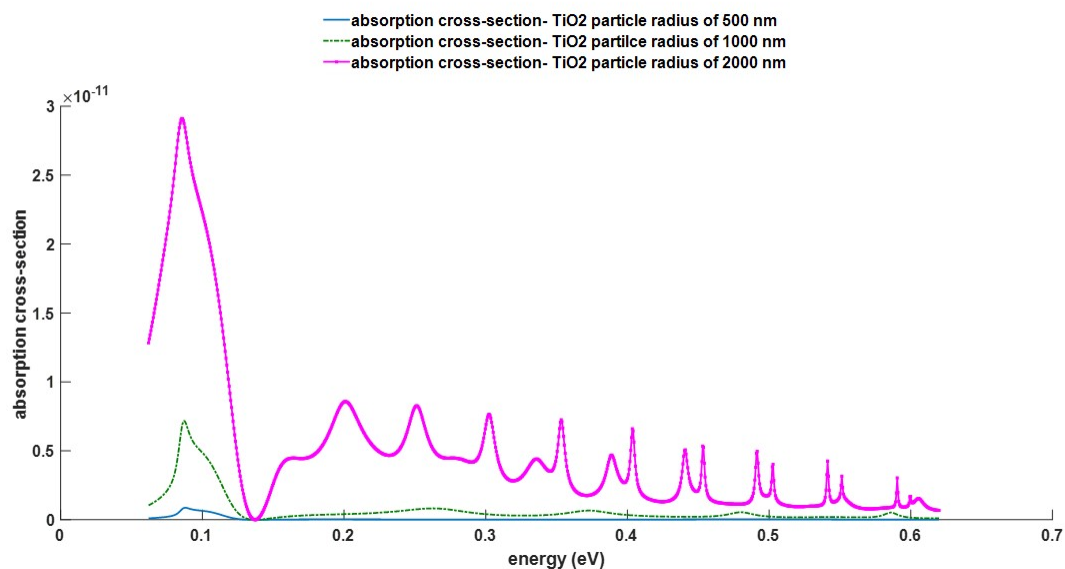


Figure 23: Comparison of particle size-TiO₂ absorption cross-section

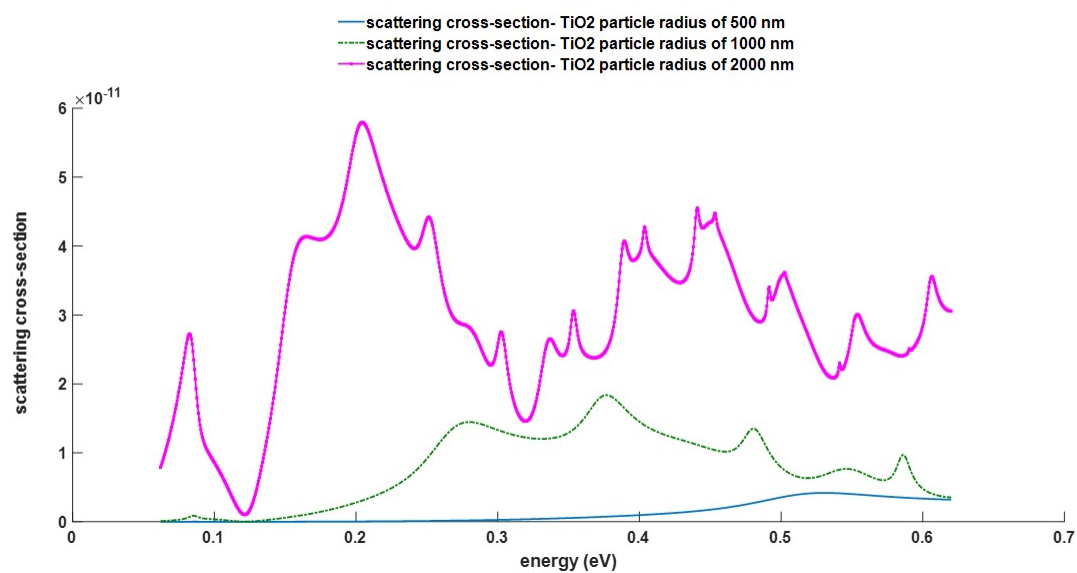


Figure 24: Comparison of particle size-TiO₂ scattering cross-section

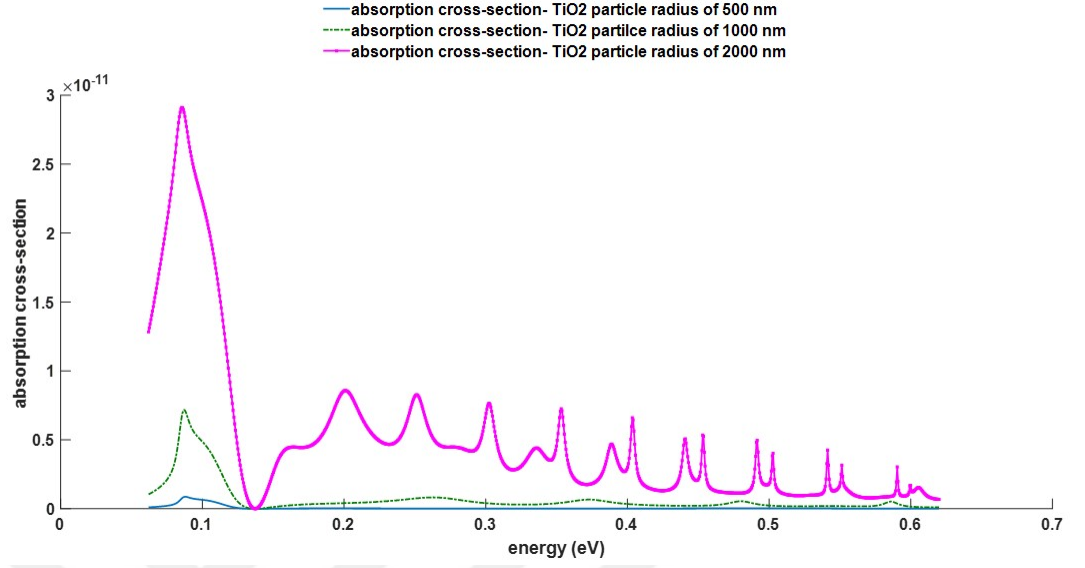


Figure 25: Comparison of particle size-ZnO absorption cross-section

2000 nm radius, and scattering scattering cross-section profiles, respectively.

Thus, the comparison results of the difference in particle sizes of the three materials SiO_2 , TiO_2 , and ZnO obviously show that as the particle's radius size increases, the absorption cross-section and the scattering cross-section increase, which matches the theoretical predictions based on Lorenz-Mie theory indeed.

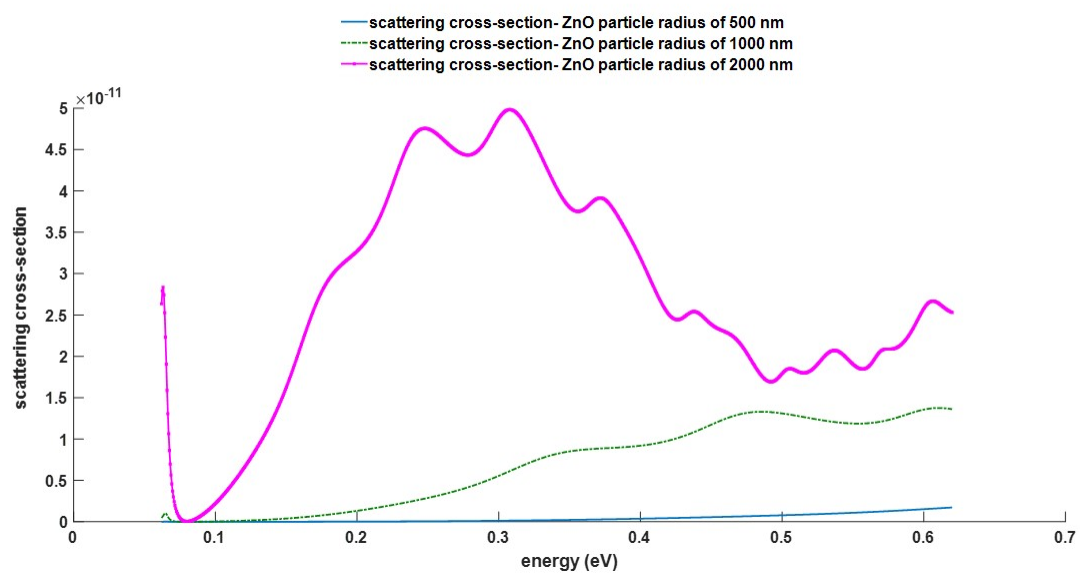


Figure 26: Comparison of particle size-ZnO scattering cross-section

CHAPTER IV

EXPERIMENTAL RESULTS

4.1 Objective of the experimental study

Our goal is to investigate the properties of different materials by experimentally analyzing sustainable or composite materials for radiative cooling. First, scanning electron microscopy (SEM) is used to analyze the surface morphology of a group of sustainable materials that can be used on the surfaces of buildings with accurate surface morphology. In addition, Fourier Transform Infrared Spectroscopy (FT-IR) is used to measure the infrared spectrum of transmittance and absorption of the samples in the atmospheric window region.

Radiative cooling is based on the optical properties of the materials, which can significantly improve the ability of the cooling system. At longer wavelengths, the materials should emit as much of the radiation as possible to reject the energy from the material to space; and at the solar window, the reflection of the energy should be maximized when the emissivity is as low as possible. The experimental analysis aims to analyze the properties of sand particles for radiative cooling applications and compare them with a group of materials such as silicon dioxide, titanium dioxide, and zinc oxide.

4.2 Repeatability of the study

In the pursuit of scientific progress, it is of paramount importance to ensure the reproducibility and replicability of research results. The importance of reproducibility lies in its ability to allow researchers to check the quality of their own work or that of others and to allow colleagues to reproduce and validate the results obtained in a

particular study. By carefully documenting methods, experimental setups, and analytical approaches, this thesis aims to create a framework that facilitates seamless experimental replication. Through transparent reporting and comprehensive documentation, future researchers can build on the foundation laid by the experiments in this study without having to repeat the same procedures redundantly. All experimental procedure steps are provided and discussed in detail in the next sections. In addition, five different pellet samples are prepared and used to repeat the Fourier Transform Infrared Spectroscopy (FT-IR) absorbance and transmittance percentage profile measurements several times, as will be discussed in the thesis. By repeating the sand FT-IR measurements five times and reporting consistent behavior and similar results from each measurement, we provided more details to ensure the repeatability of our study. A commitment to reproducibility not only enhances the reliability and credibility of scientific contributions but also fosters a collaborative environment in which researchers can collectively contribute to the cumulative knowledge of a given field. Thus, with the details of the experimental procedure and the presentation of the results of the repetition of the measurements, we report in this thesis a repeatable and replicable study indeed.

4.3 Experimental procedure

4.3.1 Scanning electron microscopy and measurements

4.3.1.1 Scanning electron microscopy and its operational principles

Scanning electron microscopy (SEM) is a microscope that uses electrons instead of light to produce a high-quality image. The importance of SEM comes from its ability to obtain detailed visual images of a particle with high-quality and spatial resolution [17]. Using electromagnets rather than lenses, SEM allows more control in the degree of magnification, so it can be used as a multipurpose instrument to observe the surface phenomena of materials and their characterization, providing information on the

topography, morphology, composition, chemistry, grain orientation, crystallographic information and so on of a material [51]. First, a beam of electrons is produced at the top of the microscope by an electron gun. The electron beam follows a vertical path through the microscope, which is held in a vacuum. As the beam passes through electromagnetic fields and lenses, it is focused down to the sample. The electrons and X-rays strike the sample and bounce back, allowing the detector to collect the X-rays, back-scattered electrons, and secondary electrons. They are then converted into a signal that is sent to the screen that provides the image.

4.3.1.2 Sample preparation for SEM

Special preparations must be applied to the samples, which are important due to the vacuum conditions used and the electrons used in scanning electron microscopy. It should be noted that all water should be removed from the samples to prevent the water from evaporating in the vacuum [51]. Also, if metallic samples can be used directly, non-metallic samples must be made conductive by covering the sample with a thin layer of conductive material, and this process is done using a sputter coater device. The sputter coater uses an electric field and argon gas [51, 52]. The sample is placed in a small vacuum chamber. By removing an electron from the argon, the argon gas and an electric field cause the atoms to become positively charged. These argon ions are attracted to a negatively charged gold foil and knock gold atoms off the surface of the gold foil. Finally, a thin layer of gold is formed on the sample surface [52].

4.3.1.3 SEM results from our experiment

Several SEM images were taken to examine the samples at different scales after the sand particles were prepared. Figure 27. shows two SEM images representing sand particles with two different scale bars (a): 100 μm and (b): 2 μm , respectively.

In addition, to provide more details and analyze the morphology of the surfaces of

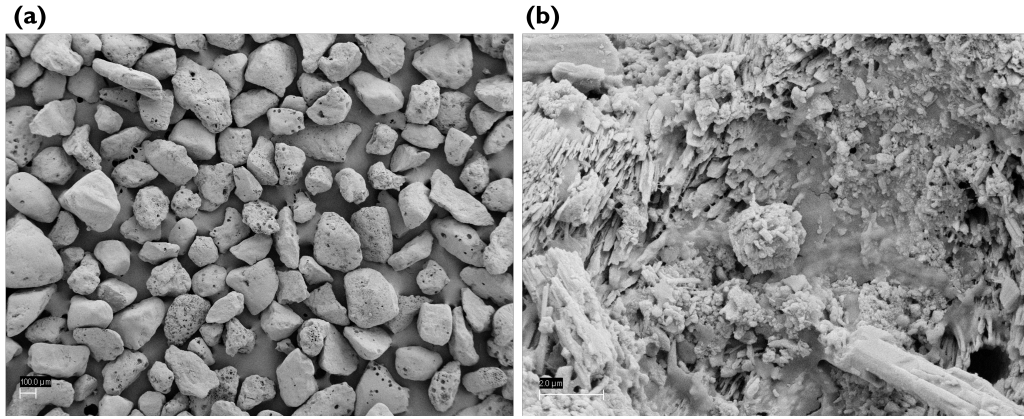


Figure 27: SEM images presenting sand particles with two different scale bars: (a): 100 μm and (b): 2 μm .

the other materials used in the study, different SEM images were taken for the other samples as well. Figure 28. shows SEM images (at 20 KX magnification and scale bar is 2 μm) presenting (a): sand particles, (b): SiO_2 particles, (c): TiO_2 particles, and (d): ZnO particles.

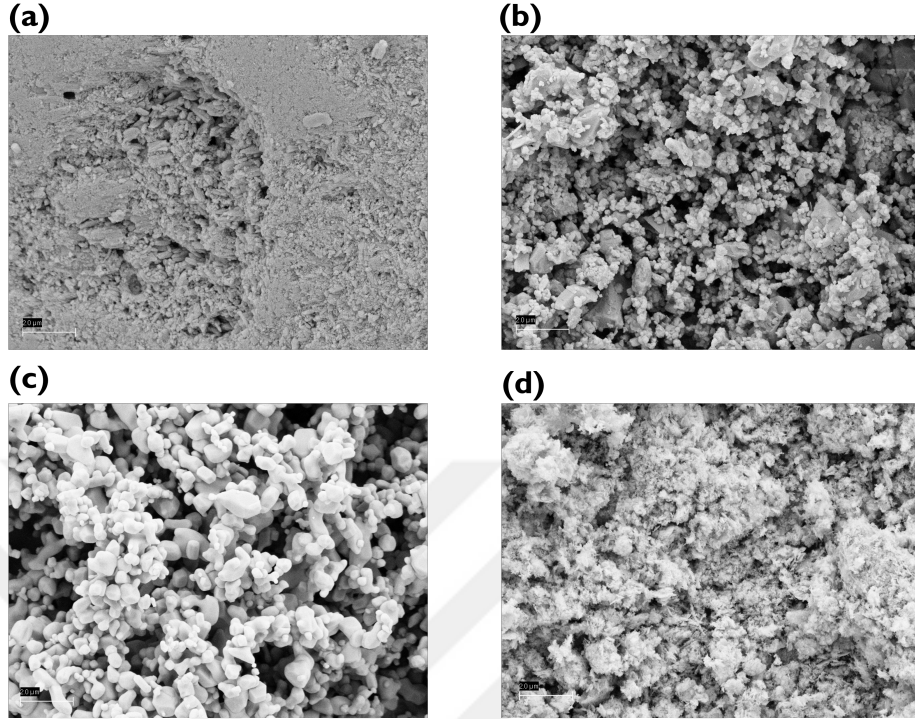


Figure 28: SEM images presenting (a): sand particles, (b): SiO₂ particles, (c): TiO₂ particles, and (d): ZnO particles (at 20 KX and scale bar is 2 μm)

4.3.2 Fourier Transform Infrared Spectroscopy (FT-IR) measurements and system

4.3.2.1 How does FT-IR spectroscopy work:

FT-IR is a well-known technique that can be used to find the spectrum of absorption and emission by measuring the amount of light energy absorbed at the wavelength by samples. Fourier transform infrared spectroscopy (FT-IR) experiment is conducted to find the absorbance and transmittance percentage of the samples.

In FT-IR spectroscopy, key components allow precise measurements of the reflectivity of materials on a sub-micron scale. The instrument's optical path consists of a beam splitter that separates the incoming light into two paths: a reference beam, which is used as the reference for the measurement, and a sample beam, which passes through a probe tip, interacts with the sample, and is then reflected back by a moving mirror, allowing the reflectivity of the material to be measured as a function of

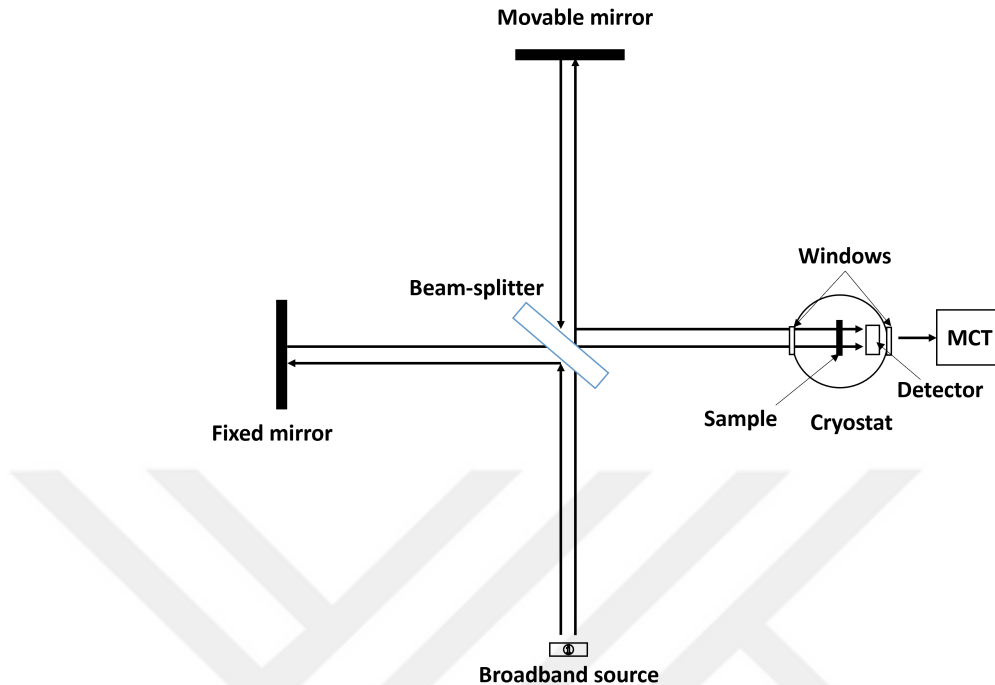


Figure 29: Schematic diagram of FT-IR spectrometer. Adapted from [30]

wavelength. After measuring the intensity of the reflected light, the information is processed by a computer to generate the spectrum. Figure 29 shows a schematic diagram of the FT-IR spectrometer and its components.

4.3.2.2 Sample preparation and FT-IR measurements:

The preparation of the sample pellets can be as important as the obtainment and analysis of the spectrum. The steps of pellet preparation were applied based on the experiment explained in the paper of S. Manickavasagam and M. P. Menguc [53]. To perform FT-IR on the sand particles and other materials used in this study, sample pellets are prepared by suspending the particles in a KBr matrix, which is transparent in the infrared spectrum. Approximately 100 mg of KBr is weighed in a chemical balance and mixed with approximately 10 mg of sand particles. Using a mortar, the KBr and sand particles are well mixed to form a homogeneous mixture. The mixture is then slowly fed into a dye to produce the pellet. To avoid any unevenness in the

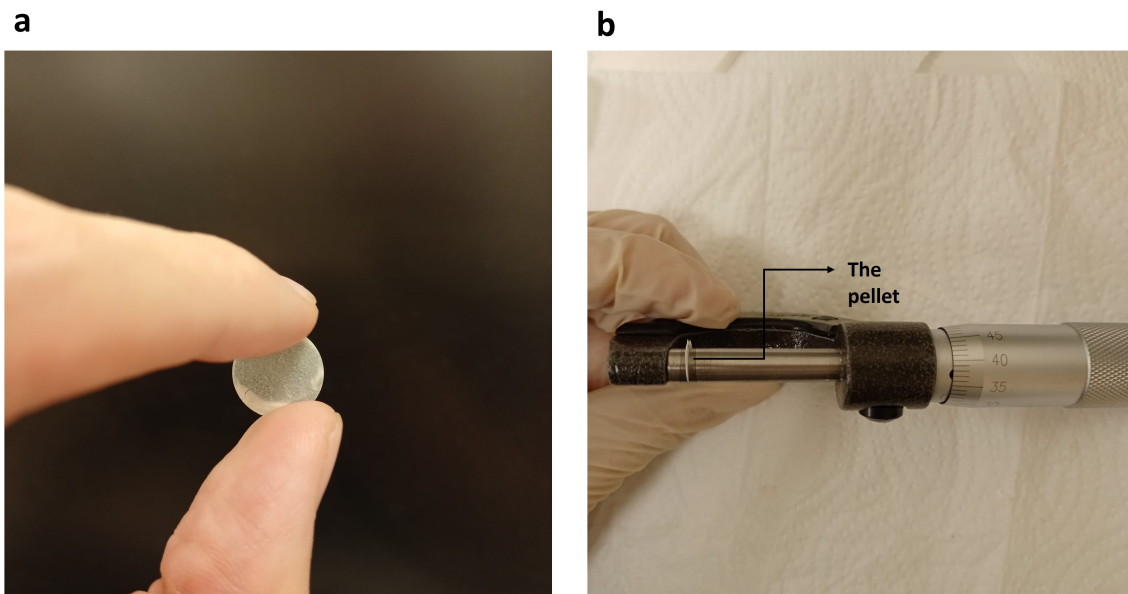


Figure 30: (a): a photo of Sand particles sample pellet, mixed and compressed with KBr 1/10 ratio by 10 Ton pressure. (b): A photo of a Sand particles sample pellet with a micrometer to measure the thickness

thickness of the pellet, sufficient care is taken during the process to ensure that the mixture is uniformly distributed within the dye. To form the pellet, the dye is held under a compression pressure of 10 tons for approximately 10 seconds. The pellet is visually inspected for small cracks or uneven distribution of the sample material within the matrix. After the pellet is removed from the dye, it is placed in a sample holder of the FT-IR spectrometer and the transmittance and absorbance spectra are measured. The pellet thickness is then measured. Figure 30. shows (a) a photo of a sand particle sample pellet, mixed with KBr 1/10 ratio and compressed by 10 tons pressure. (b): A photo of a sand particle sample pellet with a thickness of 0.25 mm. The thicknesses of the sample pellets were measured using a micrometer with an accuracy of 0.01 mm. The background is taken in FT-IR before measuring the transmittance and absorbance spectra.

All Fourier transform infrared spectroscopy FT-IR spectra were recorded as direct absorbance or transmittance, and carried out on a Thermo-Nicolet iS10 FT-IR



Figure 31: Thermo-Nicolet iS10 FT-IR (Sabanci University, Istanbul, Turkey)

Spectrometer (at Sabanci University, Istanbul, Turkey), shown in Figure 31.

Figure 32 shows the transmittance percentage of three different particle samples, which are SiO_2 , TiO_2 , and ZnO measured by Thermo-Nicolet iS10 FT-IR spectrometer. (Performed at Sabanci University, Istanbul, Turkey). Figure 33 shows the absorbance of three different particle sample materials which are SiO_2 , TiO_2 , and ZnO measured by Thermo-Nicolet iS10 FT-IR spectrometer. (Performed at Sabanci University, Istanbul, Turkey).

From Figure 33, it can be noted that SiO_2 shows a very high absorbance profile between 400- 1400 $1/\text{cm}$ (7.14- 25 μm). Also, ZnO shows a high absorbance between 400-600 $1/\text{cm}$ (16.6- 25 μm). It can be noticed that TiO_2 depicts a relatively constant and noisy behavior throughout the spectrum. By comparing the behavior of the TiO_2 particles in our study with the literature (examples are shown in chapter two), it can be seen that the TiO_2 particles in our study showed an unexpected behavior. One possible reason for that is moisture content. TiO_2 is sensitive to moisture, and the presence of water can interfere with FT-IR measurements causing the recorded behavior. One other possible reason for the unexpected behavior of TiO_2 particles could be non-homogeneity. The grinding time applied for mixing the TiO_2 with KBr

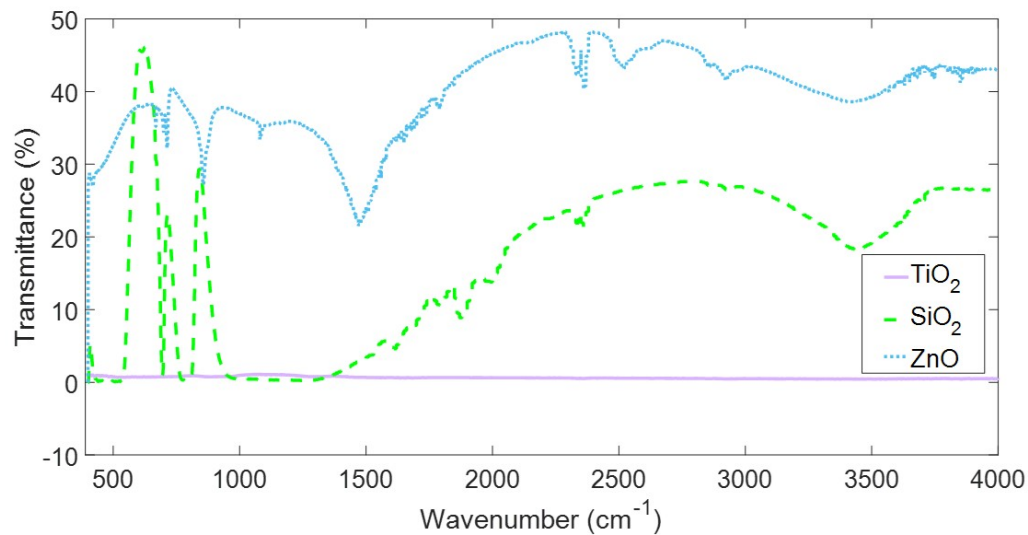


Figure 32: Transmittance of three different particle pellets samples SiO₂, TiO₂, and ZnO measured by Thermo-Nicolet iS10 FT-IR spectrometer. (Performed at Sabanci University, Istanbul, Turkey)

to prepare the pellet was about 3 minutes as explained before, which could be less than the needed time to make the sample homogenous and have consistent measurements.

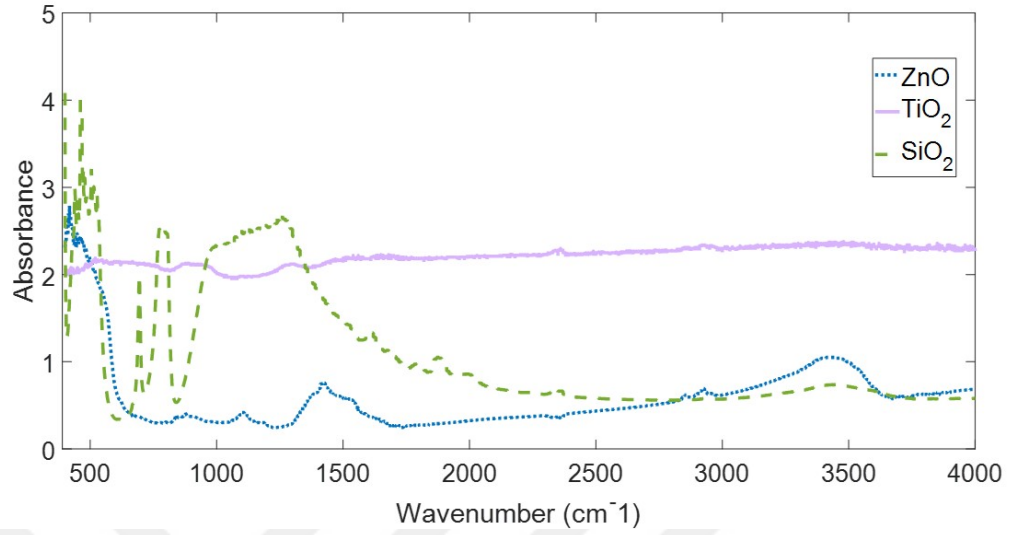


Figure 33: Absorbance of three different particle pellets samples SiO_2 , TiO_2 , and ZnO measured by Thermo-Nicolet iS10 FT-IR spectrometer. (Performed at Sabanci University, Istanbul, Turkey)

Figure 34. shows the transmittance of five Sand particle samples measured by Thermo-Nicolet iS10 FT-IR spectrometer. (Performed at Sabanci University, Istanbul, Turkey). The thicknesses of the five sand particle pellets are 0.26 mm, 0.35 mm, 0.34 mm, 0.31 mm, and 0.34 mm respectively. It can be noted that the five sand particle pellets had very small differences in their thickness after they had been mixed with the KBr and pressed, which might be the possible reason for the differences that appeared in the transmittance profile.

In addition, Figure 35 shows the absorbance of the sand samples measured by Thermo-Nicolet iS10 FT-IR spectrometer. The high spectral emissivity recorded for sand particles in the atmospheric window emphasizes that the sand particles can be good candidates for radiative cooling applications.

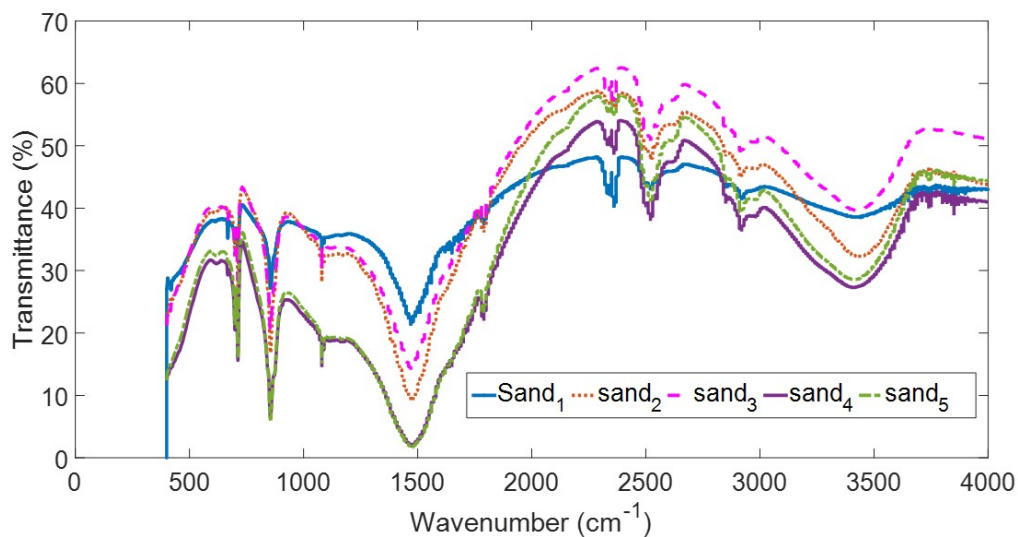


Figure 34: Transmittance of five Sand pellets samples measured by Thermo-Nicolet iS10 FT-IR spectrometer. (Performed at Sabanci University, Istanbul, Turkey)

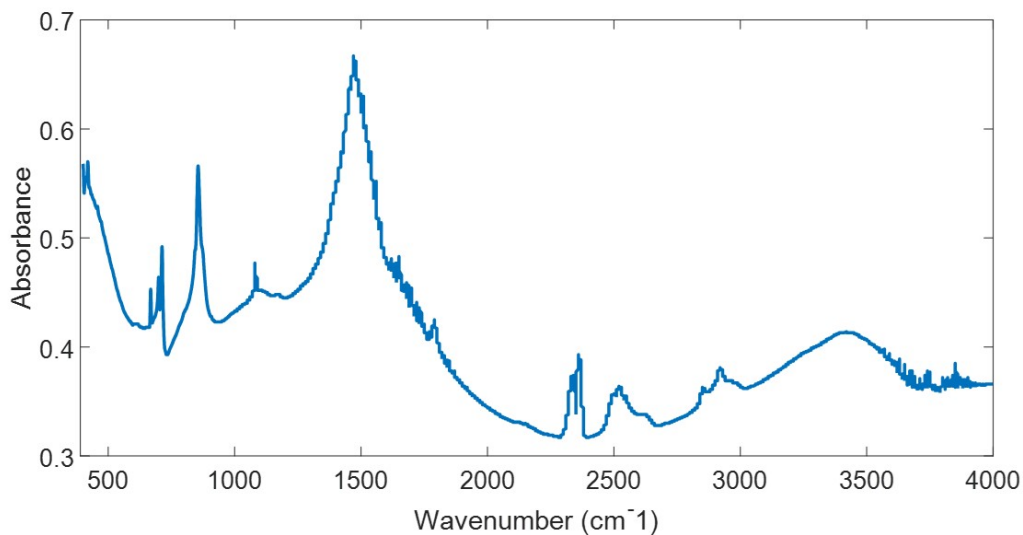


Figure 35: Absorbance of a Sand particles pellet sample measured by Thermo-Nicolet iS10 FT-IR spectrometer. (Performed at Sabanci University, Istanbul, Turkey)

4.4 *Extracting absorption coefficient from FT-IR transmittance*

When the pressed pellets are placed in the FT-IR spectroscopy for the transmittance measurements, a beam with an intensity of I_L irradiates the pellet as shown in Figure 36. Part of the intensity is reflected back by the pellet and the rest of it enters the pellet.

From an FT-IR measurement, the spectral extinction coefficient can be calculated from the measured spectral transmittance using the following formula [54]:

$$\tau_\lambda = \frac{I_T}{I_O} \approx \frac{I_T}{I_L} = \exp(-\kappa_\lambda \cdot L) \quad (13)$$

Where τ_λ represents the transmittance of the pellet, L denotes the sample thickness, and κ_λ signifies the spectral extinction coefficient. For KBr diluted pellet, KBr is assumed transparent to the infrared beam. The effective thickness of the diluted material is determined by [54]:

$$L_c = \frac{W \cdot P}{\rho \cdot A} \quad (14)$$

where W is the weight of the pellet, P is the mass ratio of the test material in the pellet, ρ is the density of the test material, and A is the cross-sectional area of the pellet. By combining equations (19) and (20), the spectral extinction coefficient can be calculated using the next formula [54]:

$$k_\lambda = -\ln \frac{\tau_\lambda}{L_c} \quad (15)$$

It should be noted that since KBr has very high transmittance, the reflectance of the KBr pellet is neglected when determining its extinction coefficient.

Thus, the absorption coefficient in this study is calculated using the following formula[11]:

$$\kappa_\lambda = \frac{2.3 \cdot A_\lambda}{L} \quad (16)$$

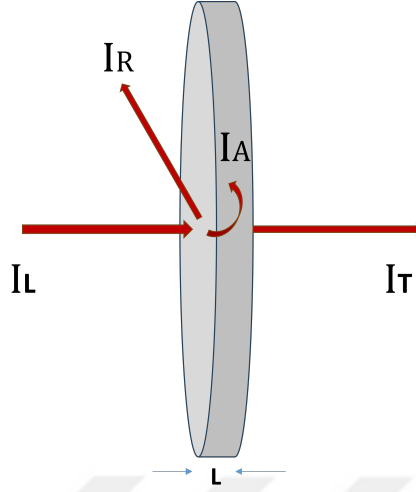


Figure 36: A schematic of a pellet used in FT-IR spectroscopy showing the reflected, absorbed, and transmitted intensity of a beam irradiates the pellet

Where κ_λ : is the absorption coefficient, A_λ : is the spectral absorbance measured from the experiments, L : is the thickness of the sample. The spectral absorption coefficient of a material determines how far the spectral radiation passes into the material, and the absorption coefficient depends on the nature of the material. The absorption coefficient of the sand particles sample is calculated and plotted as shown in Figure 37. The absorption coefficients of five different sand particle pellets are plotted as shown in Figure 38. The thicknesses of the five sand particle pellets are 0.26 mm, 0.35 mm, 0.34 mm, 0.31 mm, and 0.34 mm respectively.

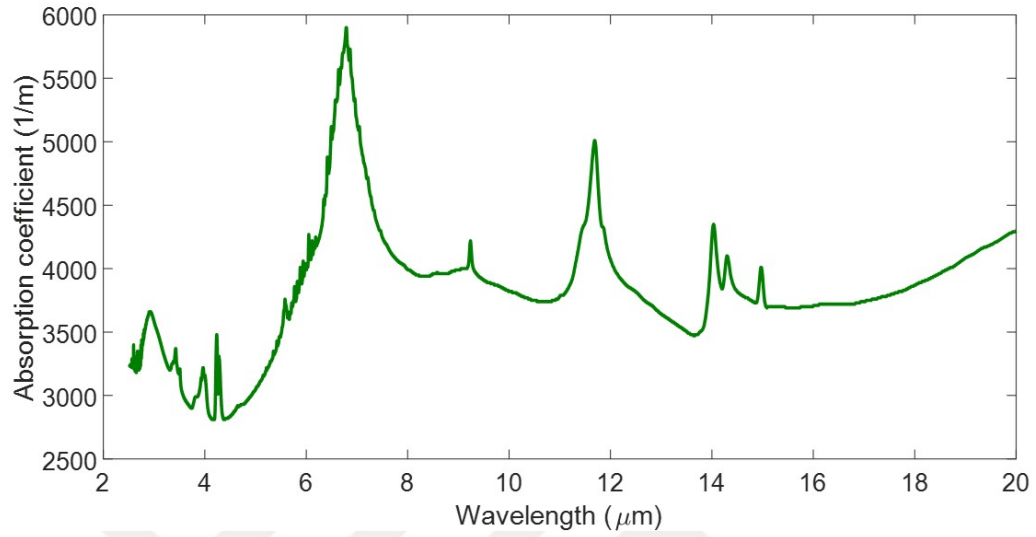


Figure 37: The absorption coefficient was calculated and plotted from a Sand particles pellet

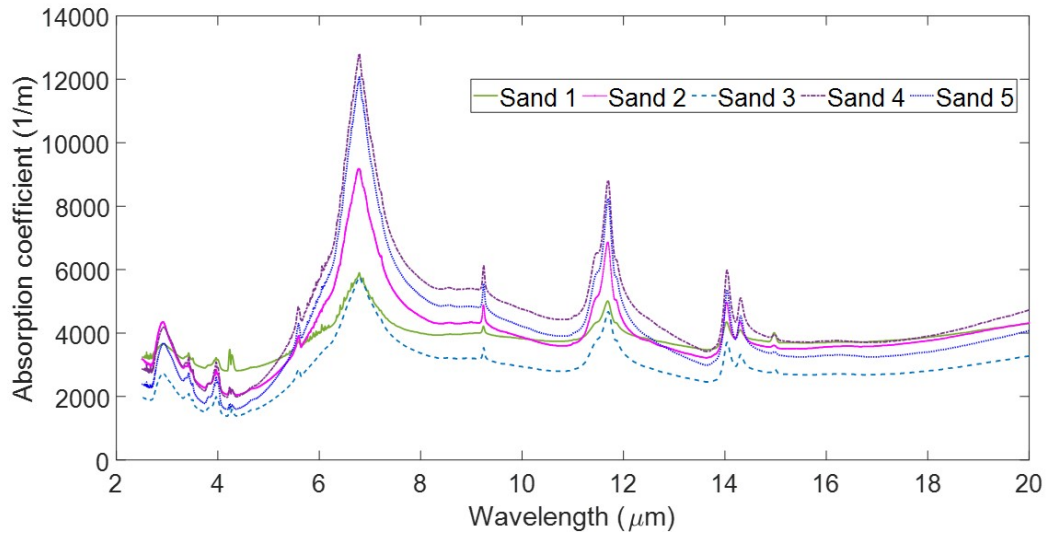


Figure 38: The absorption coefficient was calculated and plotted from five different sand particles pellets, and their thickness are 0.26 mm, 0.35 mm, 0.34 mm, 0.31 mm, and 0.34 mm respectively

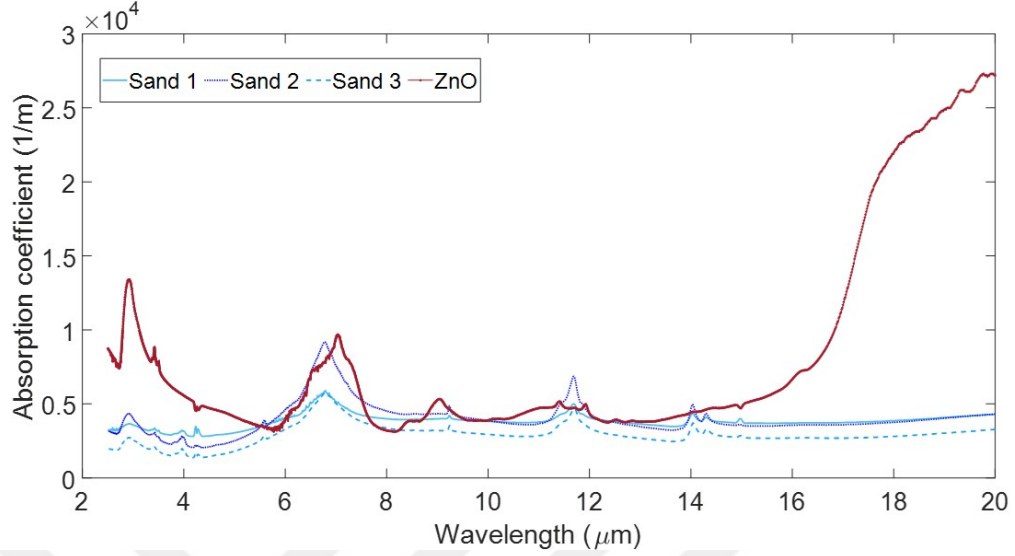


Figure 39: The absorption coefficient of three different sand particles samples compared with ZnO particles absorption coefficient

To compare the results of the sand particles, the absorption coefficient of ZnO particles is calculated and plotted with three different sand particle samples as shown in Figure 39. From the results presented in the figure, it can be noted that the sand particles showed promising results as the absorption coefficient of ZnO is almost the same as the absorption coefficient of the sand particles.

In Figure 40, the absorption coefficient of the SiO_2 is compared with the sand sample. The absorption coefficient of the SiO_2 particles showed higher results compared to the sand and ZnO samples. Considering the sustainability advantages of the sand particles and the cost, it can be reported that the sand sample is very promising for radiative cooling applications.

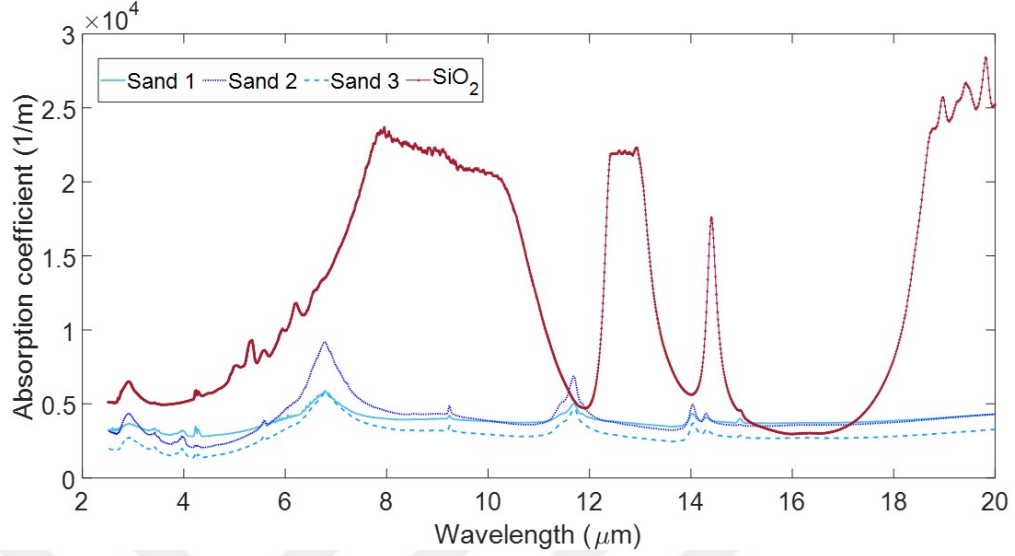


Figure 40: The absorption coefficient of three different sand particles samples, compared with SiO_2 particles absorption coefficient

4.5 Discussion of the results for sand particles

As discussed earlier, the atmospheric window region (8-13 μm) has a major impact on radiative cooling principles and systems. In this study, we focus on studying the radiative cooling properties of sand particles and some other materials, namely silicon dioxide (SiO_2), titanium dioxide (TiO_2), and zinc oxide (ZnO). As shown in the previous section in Figure 37, the high absorption recorded for sand particles in the atmospheric window emphasizes that the sand particles can be good candidates for radiative cooling applications. Similarly, Figure 38 has shown high recorded absorption coefficient values in the atmospheric window range with an approximate ratio between 0.5×10^4 and 0.7×10^4 1/m. It should be noted that the higher the absorption coefficient of a material in the atmospheric range, the better the performance of that material in radiative cooling systems. This is due to the second law of thermal radiation. This is based on Kirchhoff's second law of thermal radiation, which states that at thermal equilibrium, absorption should be equal to emission. Thus, it is a fact that if a material has a high absorptivity in the atmospheric window range (8-13 μm),

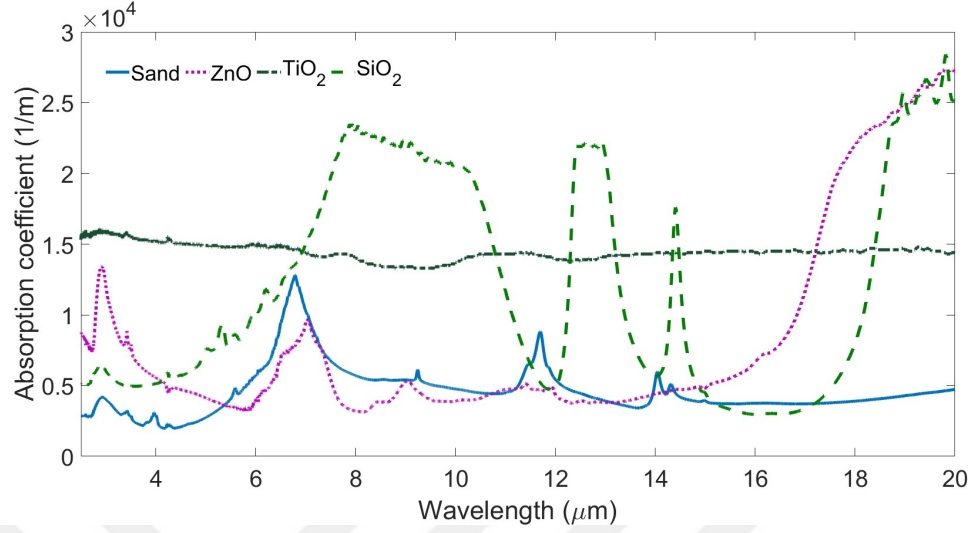


Figure 41: the absorption coefficient of SiO_2 , TiO_2 , and ZnO particles and the sand sample

it must also have a high emissivity and be able to emit thermal radiation to space through the atmospheric window. The spectral absorption coefficient of a material determines how far the spectral radiation passes into the material, and the absorption coefficient depends on the nature of the material.

In addition, the comparison of the absorption capability between the sand sample and the other materials shows that the sand sample can be used in radiative cooling applications. The absorption coefficient of ZnO particles compared with three different sand particle samples is shown in Figure 39. The absorption coefficient of the sand and ZnO samples in the atmospheric window region showed the same behavior and very similar ratios. When the absorption coefficient of SiO_2 is compared with the sand sample, the absorption of SiO_2 particles showed higher results compared to the sand and ZnO samples with similar behavior in the atmospheric window region. Considering the sustainability of the sand particles and perhaps the cost, the sand sample may still have promising advantages over other materials and may open the door for further applications and studies in this area. Figure 41 the absorption coefficient of SiO_2 , TiO_2 , and ZnO particles and the sand sample compared in one figure.

In the longwave infrared, the absorption coefficient of the sand sample was particularly high as was shown in the previous sections. This is also an indication of high emissivity at these wavelengths based on the second law of thermodynamics under thermal equilibrium. Such inherent optical properties are indeed expected for sand particles and can be explained by Wien's displacement law and Planck's radiation law. The wavelength at which an object emits the most radiation is inversely proportional to its temperature, according to Wien's displacement law. In other words, the hotter the object, the shorter the wavelength of maximum emission. Sand particles emit thermal radiation when they heat up. As their temperature increases, they emit radiation over a range of wavelengths. However, the peak of this emission shifts to longer wavelengths as the temperature increases. This explains why we observe peaks at $6.78\ \mu\text{m}$, $9.24\ \mu\text{m}$, $11.69\ \mu\text{m}$, and $14.05\ \mu\text{m}$ for all sand pellet samples.

Furthermore, we note that for a material to be a good candidate for passive radiative cooling applications, it must be highly reflective in the solar spectrum ($0.3\text{-}2.5\ \mu\text{m}$) and highly absorptive/ emissive in the atmospheric transparency window ($8\text{-}13\ \mu\text{m}$) [55], which matches the characteristics of the sand particles studied in this work. In addition, the thermal emittance increases when the particles are smaller or comparable to the wavelength of light [56, 57]. Therefore, the radiative cooling properties of the particles are enhanced, as is the case for the sand particles in this work, which had sizes of $1\ \mu\text{m}$, $2\ \mu\text{m}$, and $4\ \mu\text{m}$. Therefore, we conclude that the sand samples experimentally studied in this work have good potential for passive radiative cooling applications of buildings and electronic devices.

CHAPTER V

CONCLUSIONS

5.1 Conclusions

The purpose of this study was to assess the radiative cooling using some special sand particles and compare the radiative properties with other materials such as silicon dioxide, titanium dioxide, and zinc oxide. The morphology of the surface of the sand particles and the other materials used in the study for comparison are analyzed by scanning electron microscopy (SEM). In addition, Fourier Transform Infrared Spectroscopy (FT-IR) experiments were performed to measure the absorbance and the transmittance percentage of the samples. From the FT-IR measurements, the absorption coefficient was calculated as well. The absorption coefficient of the sand samples showed a high ratio (in the units of $1/\text{m}$ which was between 0.5×10^4 and 0.7×10^4 for five pellets. Thus, the results show that the special sand particles can be used for radiative cooling applications and systems due to their abundance and their thermal properties with better performance.

In the longwave infrared, the absorption coefficient of the sand sample was particularly high. This is also an indication of high emissivity at these wavelengths based on the second law of thermodynamics under thermal equilibrium. These inherent optical properties are expected for sand particles and are explained using the Wien and Planck radiation laws. Wien's law of displacement states that the wavelength for which an object radiates the most is inversely proportional to its temperature. This means that the hotter the target, the shorter the wavelength of the maximum radiation. Sand particles emit thermal radiation when they heat up. As their temperature increases, they emit radiation over a range of wavelengths. However, as the

temperature increases, the peak of this emission shifts to longer wavelengths. This explains why we observe peaks at $6.78\ \mu\text{m}$, $9.24\ \mu\text{m}$, $11.69\ \mu\text{m}$, and $14.05\ \mu\text{m}$ for all sand pellet samples. Taking everything into consideration, we can conclude that the sand samples experimentally studied in this work have a good potential for passive radiative cooling applications.

5.2 *Future Work*

- This study was conducted mainly for radiative cooling applications for the roofs of buildings using sustainable materials, such as sand. It can be developed for other applications as well.
- Fourier Transform Infrared Spectroscopy (FT-IR) measurements were performed to measure the absorption and transmission of the materials in the range between ($2.5\text{-}25\ \mu\text{m}$), focusing on the behavior of the materials in the atmospheric window range ($8\text{-}13\ \mu\text{m}$). The measurements of the solar spectrum need to be studied to understand the reflectivity of the materials in the range between ($0.3\text{-}2.5\ \mu\text{m}$). Due to technical issues beyond the author's control, the spectral behavior of the materials in the solar spectrum was not included in this thesis. In future studies, these additional studies need to be carried out to provide more information on the materials validation for radiative cooling applications.
- In this study, Scanning Electron Microscopy (SEM) imaging was performed to determine the morphology of the materials. To analyze the effect of the presence of other materials in the sand samples, other materials such as SiO_2 , TiO_2 , and ZnO were also studied. However, a more comprehensive study would need in parallel a chemical analysis of the samples.
- The absorption coefficient of samples is determined from the FT-IR absorbance and transmittance measurements. However, a more detailed study needs to

be conducted to account for the packing factor of sand particles used in the experiments. Also, the shape, size, and size distribution of particles need to be analyzed more in-depth. Temperature measurement tests of the samples need to be carried out to determine radiative cooling potential of samples in real-world conditions. For that, a coupled conduction radiation analysis needs to be carried out. Then, the materials can be characterized to demonstrate if they are suitable for different radiative cooling systems and applications.



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