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RADIATIVE PROPERTIES OF COPPER THIN FILM AS RADIATIVE COOLING MATERIALS

MOURAD BENLATTAR[1,2], EL MOSTAFA OUALIM [2]

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Abstract. *Optical and radiative properties of glazing materials are primary imputes for determination of the radiative cooling performance. In this paper, SiO_2 shield for radiative cooling was designed and fabricated using the copper as a metal layer. The thin layer (0.2) on SiO_2 substrate (2mm) was produced using a sputtering technique. Spectral specular reflectance and transmittance of the film were measured accurately for 0.3 to 20 using an automated spectroradiometric system of high resolution. This class of coating is characterized by high solar band reflectance and high IR band emittance; so cooling to low temperature is possible with Cu/SiO_2 system which is good reflectance in the visible range and are strongly emitting in 8- 13 band.*

Keywords: *Radiative cooling, reflectance, transmittance.*

1. INTRODUCTION

Over the past, the introduction and the developments of an important new area of technology in the architectural filed has occurred, namely, the use of thin film coating to enhance the radiative cooling in the buildings. Radiative cooling has many others potential applications [1-3], including keeping food, seeds and medicines and water desalinization. Now, in view of the growing energy crises, it's widespread interest world because that could contribute to energy savings [4, 5]. Passive radiative cooling is one among today's challenges in materials science research. Scientists know that the radiation from the clear sky with different wavelengths varies in its capacity to cross the atmosphere layer. If humidity is not too high, there is very little downward radiation within the 8-13 band, known as the ''atmospheric window''. It's then obvious that a surface which faces the clear sky can emit radiation that is not balanced by an equal amount of counterradiation within the "window range". Thus, people introduced the concept of radiative cooling to low temperature by radiative exchange. By exchange, the heat energy of objects on the ground passes through the atmosphere to interplanetary space in the form of electromagnetic waves with wavelength of 8- 13 , so the objects are cooled. The key to implementing the above process is to choose the materials with spectral selectivity. Little effort has been devoted to use radiative cooling during the day. At noon, with the sun in its zenith, the solar illumination is very intense and heats every exposed material existing in nature. A shield possibly changes this and promotes cooling or, at least, avoids heating. In general there are two principal designs of a shield which could be used for this purpose.

[1] Hassan II University, Faculty of Sciences Ben M'sik, Department of Physics, 7955 Sidi Othman Casablanca. Maroc. E-mail: m.benlattar@um5s.net.ma.
[2] Hassan 1er University, Faculty of Sciences and Technologies, Laboratory of Applied Optics and Energy Transfer , Maroc.

The first design of a shield is based on high IR transmittance and high solar reflectance. The radiator is exposed to the sky in order to allow radiative cooling and solar gains are decreased by the shield [6].

In the second design the cover is opaque to both solar light and the IR. If the upper side of the cover has a high solar reflectance and a high IR emittance most of the absorbed power from the sun will be emitted as thermal radiation towards the sky and the temperature of the cover will be close to that of the ambience [7]. This paper deals only with the second design covers employing Cu/SiO_2 system.

Introductory installation of coated glass began in the 1960s with the developments of large-scale coating facilities, and these were expended considerably over the last four decades as the merits of coatings began appreciated [8]. Vacuum evaporation is generally used to deposit such films. In this work, we use the sputtering technique for Cu film preparation. This method appears promising for the design of radiative cooling shield, where specific properties are required [9]. The main objective of the present work is to discuss Copper thin film onto SiO2 substrate and their radiative properties in order to assess the possibility of Cu thin film for radiative cooling uses. So, our aim is to investigate different approaches for design of the shield for radiative cooling; focusing on film deposition by the sputtering technique with emphasis on their radiative properties as a shield for passive cooling devices.

2. EXPERIMENTS

2.1. COPPER SPUTTERING

A vacuum chamber was used to perform the experiment. The in situ cleaning was carried out an argon plasma glow discharge, followed by copper sputtering. A pressure of 10^{-5} bar was maintained to produce the reactive plasma by applying 0.5 A with an advanced energy dc power supply. For a few minutes prior to deposition, the SiO_2 sheet was moved away from the copper magnetron as the plasma erodes the target surface. This eliminates impurities and oxides which come from atmospheric exposure while the chamber is kept open. The SiO_2 substrate was moved horizontally inside the chamber in order to cover the largest possible area with a uniform thickness throughout the deposition process [10]. The thickness of the film was measured by mechanical techniques (DEKTAK 3030). Table 1 shows a typical deposition condition for copper sputtering.

Table 1. System conditions for copper sputtering in argon plasma.

Reactor pressure (bar)	Flow rate (sccm*)	Electrical current (A)	Discharge voltage (V)
10^{-5}	25.5	0.5	365-372

**sccm : standard cubic centimetre per minute*

2.2. MEASUREMENT EQUIPMENT

Spectral specular reflectance and transmittance measurements are made over the full (0.3-20 μm) using an OL-750 Spectroradiometer equipped with a controller [11]. In the system, a dual source was used: a tungsten filament source was used from 300 nm to 1100 nm and a ceramic glower source from 1 μm to 20 μm. The lamps were driven by a stabilized dc power supply to avoid fluctuation. After an initial settling period of a few minutes, the light intensity was found to be constant (within 1%) over several hours. With the light source placed near the sample, the illuminated sample area extended well beyond that seen by the light collection system. The beam divergence in central region was less then 1°. The sample is placed in the center of a horizontal ring in a holder, which can rotate around its vertical axis. The detector can be swept around the ring then allowing for measuring the spectral specular reflectance at different angles of incidence. Normal transmittance was measured at an angle of 1° to decrease the influence of multiple reflections. Near normal reflectances; s- and p-polarized reflectance were measured at an angle of 20°. We have measured the spectral specular reflectance and transmittance at room temperature.

The monochromator has a rotating diffraction grating that sweeps the spectrally resolved light past a fixed detector. A silicon detector is used for the wavelength range from around 300 to 1100 nm, whereas a pyroelectric detector covers the range between 1 to 20 μm. Under these conditions used here, the spectral resolution was about 0.01 nm. The entire device was purged to eliminate water molecules [11].

2.3. MODEL FOR RADIATIVE COOLING

The model under study is an ideal absorber surface placed on the earth, surmounted by a horizontal shield [12, 13]. The metallic coating is facing the radiator. The shield is assumed to steadily receive energy from the sun. The solar energy is first absorbed by the shield, which reflects back a part in space and transmits the rest toward the absorber. The absorber emits thermal radiation toward the window which is partially reflected and absorbed by the absorber (blackbody). However, the window emits by its to faces, according to law Lambert, toward the space and the absorber (see Fig. 1). In the model, heat transfers by convection or conduction are neglected [6].

The window is characterized at each wavelength by three coefficients measuring the fraction of the incidental spectral power being able to be subjected to the reflection, transmission and absorption. For each wavelength, there are two spectral transmission coefficients ($T_1(\lambda)$, $T_2(\lambda)$) and two spectral reflection coefficients $R_1(\lambda)$ and $R_2(\lambda)$ corresponding, respectively, to the waves traveling from the side (1) to the side (2) and from the side (2) to the side (1).

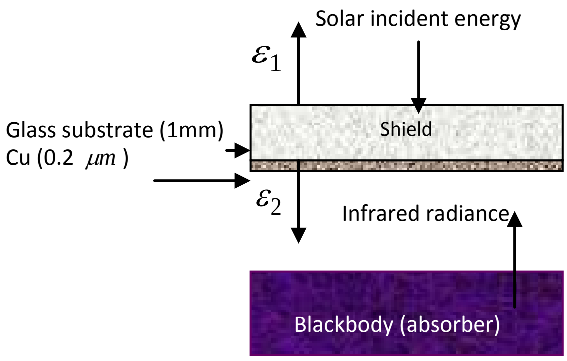

Fig. 1. The schematic description of the model study.

In order to be able to evaluate and compare the characteristics of optical properties of the shield, we will define some optical functions. We define integrated reflected intensity and integrated transmitted intensity for VIS/NIR and for "atmospheric window". We follow the definition of Nilsson et al. [14] for solar band reflectance (R_{sol}) of the film over the solar spectral (0.3-1 μm), as given by

$$R_{sol} = \int_{\lambda_1}^{\lambda_2} R(\lambda) W(\lambda) d\lambda \Big/ \int_{\lambda_1}^{\lambda_2} W(\lambda) d\lambda \tag{1}$$

where $W(\lambda)$ is the standard solar irradiance data [15] and $R(\lambda)$ is the spectral reflectance of the film.

In an equivalent way we calculated the solar band transmittance (T_{sol}) and solar band absorption (A_{sol}) as given by the following equations

$$T_{sol} = \int_{\lambda_1}^{\lambda_2} T(\lambda) W(\lambda) d\lambda \,/\, \int_{\lambda_1}^{\lambda_2} W(\lambda) d\lambda \tag{2}$$

and

$$A_{Sol} = 1 - (R_{sol} + T_{Sol}). \tag{3}$$

Applying the same calculation in the atmospheric window is a more complex issue, since the sky spectrum (atmospheric window range) is highly dependent on the humidity of the atmosphere. The structure of the earth's atmosphere is complex, and the methods used to model the transport of radiation through it can be equally complicated. The minimum parameters required to characterizing the atmosphere for optical modeling and spectral measurements research are turbidity, precipitable water vapor and carbon dioxide [16, 17]. In this paper, we will use the results of Passman and Larmore giving the atmospheric transmission from the absorption coefficient from H_2O and CO_2 [18], and in same way, $T_{8\text{-}13}$, $R_{8\text{-}13}$ and $A_{8\text{-}13}$ are calculated in the atmospheric window range. The IR band properties are evaluated for a spectral distribution of blackbody at radiance of 300 K.

3. RESULTS AND DISCUSSION

Measured spectral specular reflectance, aborptance and transmittance of copper thin (0.2 μm) on SiO_2 substrate (side 2) is shown in Fig 2. From spectral reflectance, we can see the position of the critical point structure at 0.4 eV (0.31 μm) [19]. The onset of this interband absorption is associated with transitions from Fermi surface to the next empty band or with transitions from a lower lying filled band to the Fermi surface. Interband absorption can be identified with the structure in the real and imaginary parts of the dielectric constant [20]. The Cu film on a glass substrate was used as spectrally selective filter that reflects most of the visible spectrum and absorbs infrared radiation (due to the properties of the metal layer). The highly reflective metal film, that otherwise transmits very little energy in the infrared region, was absorptive coatings so as to enhance the energy emitted in infrared window (8-13 μm).

It can be observed from Fig. 3 that the reflectance of the SiO_2 (side 1) is higher in solar region and lower in the (1-20 μm) except for the peak situated at 9 μm corresponding to the Si-O bonding [21]. Most of the measurements for radiative cooling materials were made using a SiO_2 substrate, but in some cases Si substrate was required [22]. SiO_2 substrate is one of practical substrates for use as radiation shield. It was selected as the substrate material due to its availability and constant optical properties, which were difficult to obtain for the homemade samples. Therefore, SiO_2 has high-emissivity behavior because due to low reflectance and to low transmittance results in a high absorptance in the IR window.

Optical measurements were carried out using two different configurations: the first one, the SiO_2 substrate (side 1) on the upper side (Fig. 2). The second one, the layer of copper (side 2) is on the side facing the absorber (Fig. 3). The above radiative properties for the shield reported in this paper are calculated and presented in Table 2.

In the first case, Table 2 shows that Cu/SiO_2 has the highest solar band reflectance over the solar region (R_{sol}= 0.41) and increase to (0.5) in the visible region. As can be seen from Fig. 2, the solar band transmittance is reasonably very low (T_{sol}= 0.03) which means that (A_{sol}= 0.56). In the window IR region the corresponding values were R_{8-13}= 0.05, T_{8-13}= 0.01 and A_{8-13}= 0.89.

In the second case, the solar band transmittance is very low (T_{sol}= 0.02), the same extent at the solar band reflectance is (R_{sol}= 0.3) (Fig.3) and furthermore the spectral specular reflectance depended strongly on the configuration of the film. In window IR region, the nature of the spectral specular transmittance in the two optical configurations is almost identical (T_{8-13}= 0.03).

Table 2. Radiative properties, R_{8-13}, A_{8-13}, T_{8-13}, T_{sol}, A_{sol}, and, R_{sol} of shield faces for application in radiative cooling devices.

Face	R_{sol}	T_{sol}	A_{sol}	$T_{8-13\ \mu m}$	$A_{8-13\ \mu m}$	$R_{8-13\ \mu m}$
Side 1	0.41	0.03	0.56	0.01	0.89	0.05
Side 2	0.30	0.02	0.68	0.03	0.59	0.46

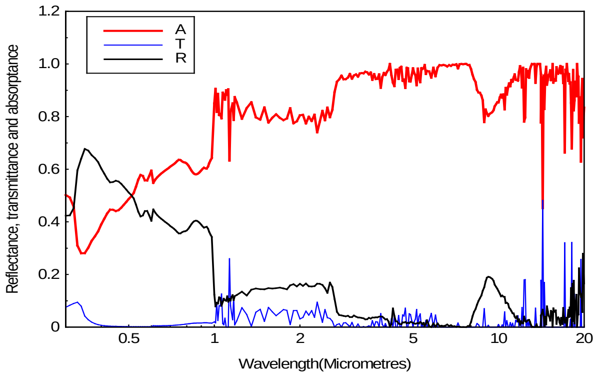

Fig. 2. Reflectance R, transmittance T and absorptance A of a thin metal film of copper on SiO_2 substrate, glass is facing the incident beam (side 1).

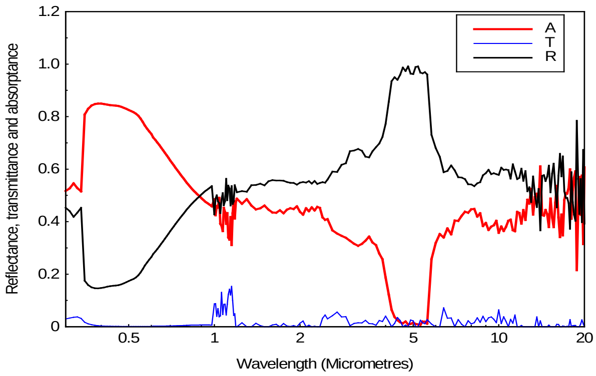

Fig. 3. Reflectance R, transmittance T and absorptance A of a thin metal film of copper on SiO_2 substrate, copper is facing the incident beam (side 2).

The treated window is opaque to most of the visible radiation received from the sun. It has also an opaque behavior of the thermal radiation emitted by the absorber. Even if its opacity for the IR window radiation, the amount of energy so emitted by the shield toward the upper side. It's checked that the emission of the treated side is very weak compared to that of the untreated side.

The coating emissivity ε_N is calculated by integrating spectral reflectance in the spectral region of 2.5–20 μm, expressed as [23]

$$\varepsilon_N = \frac{\int_{2,5}^{20} (1 - R(\lambda, \theta = 20°)) I_b(\lambda, T) d\lambda}{\int_{2,5}^{20} I_b(\lambda, T) d\lambda} \tag{4}$$

where T is the temperature and $I_b(\lambda,T)$ is the spectral intensity of a blackbody at temperature T.

The absorbed energy is preferentially reemitted toward the sky due to the high emissivity of SiO_2 (67% at room temperature) compared to that of copper (17%) [24]. If SiO_2

is facing the sky, these figures indicate that the upper face prevents the transmission of the greatest part of radiation coming from the sky, and allows the lower face to evacuate most of the thermal radiation emitted by an underlying material, a black radiator in the present case.

We can deduce that a cooling radiant can be realized by a metallic mirror effect deposition on a SiO_2 substrate which evacuates the maximum of the infrared radiation in the atmospheric window; this latter could be used for radiative cooling [25]. A consideration of the two geometries leads to an expected difference in the spectral specular absorbance. However, the side 2 facing blackbody appears to possess the best properties for use as a shield radiative cooling devices. In the conditions of a normal incidence and an ambient temperature of 316°K [6, 26], the calculated temperature of the black absorber in thermal equilibrium, when is covered by the shield, is 284.6°K less than the ambient temperature with a difference of 31°K.

4. CONCLUSIONS

In the present work we have described a method for preparation of large area thin film of copper on SiO_2. Optical properties were measured both in the solar region and in the atmospheric window both for the coated and for the uncoated faces.

We have applied also the information to radiative cooling of a black body radiator shielded with a Cu/ SiO_2 window. It has been determined that the window (Cu/ SiO_2) exhibits high emissivity in the mid-IR (8-13 μm) and highly blocking in the solar spectral region. These characteristics are suitable for use as solar radiation shields in a radiative cooling device.

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