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A Study of Coating and Characterization of Sputter Deposited Solar Selective Absorbers with Metal Interlayers for Solar Thermal Applications

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ABSTRACT: Solar energy is a renewable form of energy available in abundance across the globe, which has given hope to be a potential alternate energy source for the present depleting non-renewable energy sources. Solar energy can be harnessed either by converting it into heat energy or electrical energy. Presently there is a great need for the efficient conversion of solar energy into heat energy. Trapping solar energy with the help of solar collectors, coated with materials of different refractive index is one of the most effective way to do it. The selective coating on solar collectors should help maximize the absorption at the same time minimize the emittance. The design of solar selective coating can be single, double or can have multiple layers. Multi-layer design has been proven to be more effective for absorption of solar radiation in the recent past. In case of multi-layer tandem stack, each layer will have its role to either increase the absorption or decrease the emission as these two are independent parameters. The present study has focused on some of the recent work carried out in the design of solar selective coatings with intermetallic layer developed by sputtering process and analyzing individual layer effect in the performance of coating. Specifically, coatings developed with Tungsten, Titanium, Molybdenum and Aluminium intermetallic layers as IR reflectors, deposited on different substrates are discussed.

KEY WORDS: Solar energy, Sputtering, Absorption, Emittance, Thermal stability

I. INTRODUCTION

For any country to grow economically, modernize and advance technically, energy is the basic requirement. Economic development of any country is greatly influenced by industrial growth [1]. About 80 percent of energy requirements for various needs of humans across the globe is being supplied by non-renewable energy sources. But with time these non-renewable energy sources are getting exhausted, as the consumption is accelerated. Also usage of most of these energy sources are harmful for environment. This has made to think about alternate non-conventional energy resources, which are more promising in developing solutions in a sustainable way for present energy requirements [2]. Many non-conventional energy resources like solar, tidal, wind, biomass, hydro and geothermal release no toxic gases like carbon di oxide and hence are sustainable alternatives [3].

Among the available non-conventional energy sources, solar energy has the potential to accomplish the heat demand to various processes like sterilization, drying, pasteurization, dyeing, washing and boiling. These types of processes are found industrial sectors like food processing, agriculture, bricks manufacturing, pharmaceutical and textile which play an important role in economic development mainly in developing nations [4,5].

Collection of Solar energy is being done either as solar thermal energy or being converted into electrical energy. Solar thermal energy is obtained by converting solar radiation energy into heat. Solar collectors receive the solar radiation and convert it into heat with the help of working fluids. This heat energy can be stored and utilized for various

processes requiring heat or can be used to heat water or air directly. The process of conversion of solar energy to heat and its utilization for mechanical work is represented in figure 1. Solar collectors used for this purpose, can be of two types mainly, non-concentrating type and concentrating type. Non- concentrating type collectors are fixed at an angle based on the location to increase collection of radiation. Concentrating type of collectors have been considered to be more effective and they can take any shape such as parabolic, cylindrical, flat, convex or concave. Positioning of these collectors has a significant impact as solar radiation tracking has to be done for the entire day [6].

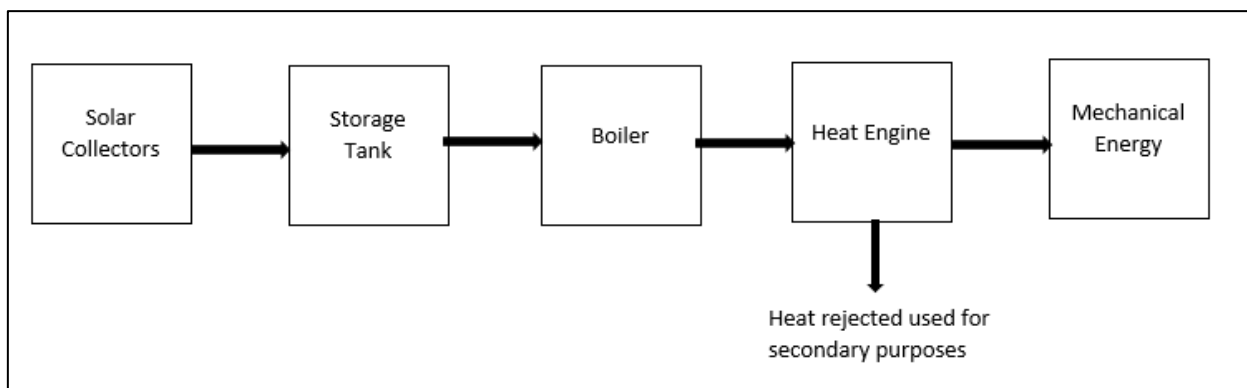


Figure 1: Solar thermal conversion Process [6]

This paper reviews some recent developments that the researchers have reported about solar selective absorber coatings using sputtering as the deposition process. The intermetallic layer or IR reflector layer coated on substrate plays a very important role in the performance of coatings. The study is mainly carried out on the basis of metal interlayers deposited on different substrates and their influence in improvement in the absorption and reduce emission, while considering the impact each layer in the tandem stack.

II. SPUTTERING – AN EFFECTIVE THIN FILM COATING TECHNIQUE

Solar collector surfaces can be coated with different materials for effective absorption of solar radiation selectively. These are called as selective solar absorbers for solar thermal applications. Coatings for these absorbers can be developed either by physical vapor deposition method or chemical vapor deposition method.

Physical vapor deposition is a coating method carried out on usually metallic substrates from a source material called as target, in vacuum or in presence of an electrolyte. Thickness of layers deposited on these substrates can vary from few atomic layers to some micro meter [7]. The important focus with respect PVD from past few decades has been on improving the performance of these coatings to make them eligible for industrial application [8-10].

Among many PVD methods, evaporation and sputtering are used for most applications. Evaporation yields comparatively thicker coatings than sputtering techniques, which is attributed to higher pressures utilized during deposition process, hence sputtering is the mainly used PVD method for thin film coatings [11-15]. Also sputtering yields coatings with superior characteristics in terms of morphology, mechanical properties as well as thermal stability required for mainly solar thermal applications. There are many variants in Sputtering process as evident in figure 2. Among all types, the most commonly used is Magnetron sputtering which itself has many variants.

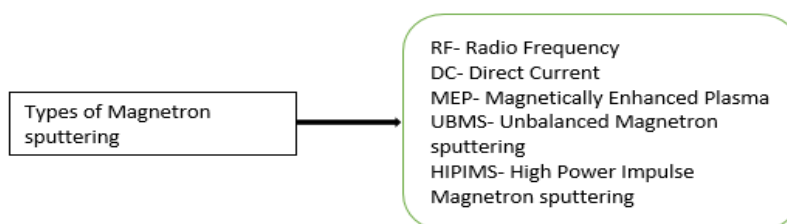


Figure 2: Variants of Sputtering Process [18]

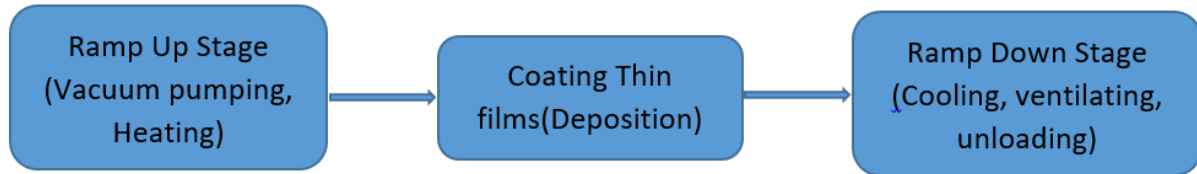


Figure 3: Steps involved in the process of sputtering [18]

Steps involved in sputtering are shown in figure 3. First the ramp up process involves setting up the required pressure inside the vacuum chamber. Next, substrate cleaning will be carried out by etching process using ions present in plasma. The process of cleaning is very important to ensure good adhesion of coating on substrate [16,17]. Coating is the third step during which the material called target is etched and projected on substrate to get deposited. Most commonly coated materials in sputtering are Ti, Cr, Zr oxides, nitrides and oxy-nitrides. Finally, the deposition chamber is brought back to normal temperature and pressure, moving out the coated substrate carefully [18].

III. SOLAR SELECTIVE ABSORBERS WITH TUNGSTEN IR REFLECTORS

K P Sibin et al., has investigated methods to reduce the emittance of stainless steel by depositing a layer of Tungsten which has high infrared reflectance, using DC magnetron sputtering device. During the study, tungsten layer greater than or equal to 830nm, with different roughness was coated on SS. A tandem absorber stack consisting of AlTiN/AlTiON/AlTiO was deposited over tungsten to understand the influence of tungsten on the emittance stainless steel. The emittance of SS with W coating was compared with SS without W coating. It was observed that the emittance of SS reduced to 0.09 with W coating and as the thickness of W layer increased above 830 nm, emittance reduced to 0.03. Further increase in W layer did not improve the emittance. Absorptance of SS improved with the increased W layer thickness. Influence of surface roughness on emittance was analyzed, and found that a rough layer of W will increase the emittance but there was increase in absorption value as well. This increase in absorption was attributed to the presence of grooves present on the rough surface. XRD study was carried out to understand the structure of W layer with 180nm, 375nm, 580nm, 830 nm and 880nm thickness. The chemical structure was analyzed by XPS. To investigate the resistivity and sheet resistance of W, it was coated on a non-conducting substrate like glass and was observed that with increase in W layer resistivity and sheet resistance both reduced to 1.5×10^{-7} Ohm and 0.1 Ohm/degree respectively. To study the effect annealing, W layer was heated separately in air as well as in vacuum, annealed to observe its thermal stability in air as well as in vacuum. Up to 750⁰ C the vacuum annealed W coating exhibited stability without undergoing any changes. For air annealed W coatings, stability was observed below 400⁰ C, and above this temperature emittance values increased. These layers started peeling off from Stainless Steel substrate for temperature above 600⁰ C. The absorber layers coated on W was investigated, which revealed an absorption of 0.955 and emittance of 0.08. W had an influence on the optical properties of tandem absorption layers by contributing to the inhibition of outward diffusion of SS. The stacks were annealed in vacuum and observed stability up to 600⁰ C for 100 h [19].

J Jyothi et al., designed a five layered tandem stack on substrates of stainless steel, the bottom most layer was tungsten coated on SS acting as an IR layer, with 3 absorbing layers, a semitransparent and a protective layer at the top. The coatings consisted transition metals, Titanium-aluminum carbides and nitrides, titanium- Aluminium- silicon carbide nitrides and oxides with different metallic content to facilitate absorption. A gradient in refractive index was introduced by varying the metallic content, bottom layers of absorption having the highest and top layer the least. The deposition was carried out in presence of reactive gases C₂H₂, Oxygen, Argon and Nitrogen using a reactive sputtering machine. Targets materials were Titanium, Silicon and Aluminium. Deposition parameters were varied and tandem stack was optimized, which exhibited an absorption of 0.961 and emittance of 0.07. It was observed that initially emittance value was 0.15, which got reduced to 0.07 by introducing W interlayer. The tandem stack was thermally stable in vacuum up to 900⁰ C for 2 h, also stable in air for 100 h till 650⁰C [20].

Atasi Dan et al., has developed a multi layered absorption surface based on Tungsten as an IR reflector layer, above W are deposited layers of WAlN/WAlON/Al₂O₃. Three targets were used namely W, Aluminium and Al₂O₃ for the developing IR reflector, absorber, transparent and antireflection layers along with N₂ and O₂ as reactive gases. A reactive sputtering direct current as well as RF magnetron sputtering devices were used for the deposition purpose.

Each of the above layer was optimized for all the process parameters to maximize the efficiency of absorption. The structural properties of coatings were investigated by X-ray diffraction techniques, optical properties of each layer was investigated separately, to know their influence in absorption phenomenon. WAIN coated on SS and W showed an absorption value of 0.753, while the addition of WAION increased the absorption to 0.902. Further the results showed the absorption increased to 0.958 with the inclusion of Al_2O_3 antireflection layer. The physical properties of the coatings were studied by AFM technique, which revealed the information of surface roughness values dependent on temperature. The coatings were thermally stable up to 500°C in air for 2 h[21].

Further Atasi Dan et al., continued the investigation to improve efficiency of the tandem stack W/WAIN/WAION/ Al_2O_3 through tailoring the values of refractive index and extinction coefficient. Through spectroscopic ellipsometry method of analysis, it was revealed that WAIN has a metallic behavior by showing an increase in RI with increase in wavelength, while WAION exhibits a nonmetallic nature by reduction in RI value with increase in wavelength. Al_2O_3 being a ceramic layer maintained constant RI for all values of wavelength. W being a pure metal has high value RI equal to 3.83. optical simulation was carried out with SCOUT software and the data collected through spectroscopic analysis. This computational study revealed the presence of WAIN and WAION layers and contributing in the shift of minima of reflectance towards the higher values of wavelength, thus increasing the absorption [22].

AL-Rjoub et al., investigated the effect of tungsten diffusion process by introducing a barrier layer consisting of chromium-Aluminum-silicon nitride. As diffusion of tungsten into the SS substrate can lead to the formation of compounds of iron and tungsten which in the long run deteriorate the performance of coatings. Using a direct current magnetron sputtering machine, a tandem stack was developed using Chromium-Aluminium composite material along with silicon pellets as target to deposit barrier layer as well as first absorption layer, tungsten metal as target to deposit back reflection layer. Si80Al20 composite target was used to generate the top antireflection coating. The coatings were analyzed before annealing heat treatment and after heat treatment for thermal stability. Four different designs were of tandem stack was prepared, first design without barrier layer between substrate and IR layer, second, third and fourth designs with barrier layer coated above SS, being S/W/CrAlSiN/CrAlSiON/SiAlO. In all three designs oxygen pressure was constant whereas nitrogen partial pressure was varied from 0.05 Pa to 0.11 Pa. as this work mainly focused on investigating diffusion phenomenon in coating with aging, RBS test was carried out, and found that by increasing the partial pressure of nitrogen improved the oxidation resistance of coating as well as reduced diffusion, provided with a right combination of chromium, Aluminium and silicon. As a whole this tandem stack has been reported to be thermally stable after annealing for 600°C , 200 h[23].

AL-Rjoub et al., has developed a four layered tandem stack in which bottom most is the tungsten, on this layer an absorption, semi-transparent and a protective antireflection layers have been developed consisting of Aluminium-silicon-titanium nitrides, oxy-nitrides as well as oxides. This was mainly designed for application at higher temperature. A composite target consisting of Ti50Al50 and silicon pellets were utilized to develop absorption (AlSiTiN_x) and semi-transparent ($\text{SiAlTiO}_y\text{N}_x$) layers whereas, Si80Al20 composite target was used to develop SiAlO_x anti-reflection layer. To develop the IR reflector layer tungsten target was used. Nitrogen, argon and oxygen were used as reactive gases during sputtering. This work was mainly focused on study of elemental diffusion of tungsten towards the stainless steel substrate, which was done by RBS. The analysis revealed a difference in values of RBS spectra before and after annealing in samples, confirming the diffusion of tungsten into substrates of SS. A good thermal stability was observed in coatings after annealing for 600°C in vacuum for 200 h[24].

Vijaya G et al., investigated the performance of W deposition made on SS and glass substrates using a direct current magnetron sputtering device for selective solar absorption. His study was mainly focused on analyzing the effect of power density on microstructure as well as absorption, emission and mechanical properties of W layer. The power density was varied between 0.6 kW and 2 kW. The experimental results revealed that coatings had good morphology and performance at 0.6 kW than the one coated at 2 kW. Also emittance value was as high as 0.16 with coatings developed at 2 kW whereas the emittance was very low with 0.07 for coatings deposited at 0.6 kW. This clearly indicated that W is a potential candidate for solar selective absorber coatings [25].

Later Vijaya G et al., has reported the performance of a multilayer stacking designed to improve absorption, consisting a ceramic adhesive layer, Ni metallic layer, Tungsten as an IR reflector, and a top anti reflection layer. The design consisted of repeated layers of W and Al_2O_3 on the Ni metallic layer. The experiment results have shown an improvement in absorption values from 0.73 to 0.94 as a result of this increased sequence of deposition. Emission value

observed was 0.07, thermal stability for the coating was tested at 400⁰ C in air for 8 h. This new combination, in which Ni is being used as intermetallic layer along with W and Al₂O₃, has optimized the absorption to be 0.94[26].

IV. SOLAR ABSORBERS WITH TITANIUM IR REFLECTORS

Barshilia et al., has developed a tandem stack consisting of two layers for absorption and a layer at the top for protection, it is schematically described in figure 4. All these three layers were deposited on a Ti interlayer which itself was deposited on SS and Copper substrates. But for the application in steam generation, temperature reaches above 400⁰ C and any substrate other than SS start diffusing outward to the absorption layers. The absorption coating consisted of Aluminium titanium nitrides and oxy-nitrides, while the antireflection layer consisted of Aluminium titanium oxide. This combination of multilayer stack was developed addressing the issues faced in the past related usage of composite target material, multiple sputtering machines and a top down approach of deposition process. This multi-layer was developed using Al and Ti targets in presence of Ar, O₂ and N₂ reactive gases. Ti interlayer plays the role of IR reflector as well as adhesion layer between the substrate and absorption layers. The absorption layers were designed such that AlTiN had metal volume fraction higher than that of AlTiON. This was done to develop a gradient in refractive index values in the layers decreasing from top to bottom, facilitating high absorption. The coatings were exposed to UV for about 10 h and no changes were observed in absorption and emittance post exposure. The coating also exhibited a good chemical inertness which was revealed through a corrosion test. The robustness of the coatings was revealed through a scratch test. This was because of the presence of Ti coating on the substrate. It also exhibited resistance to harsh condition of external environment when tested for 100 h. The coating exhibited thermal stability in air for 1000 h at 350 C. high absorption greater than 0.930 and emittance of 0.16 and 0.07 was observed on SS and copper substrates respectively[27].

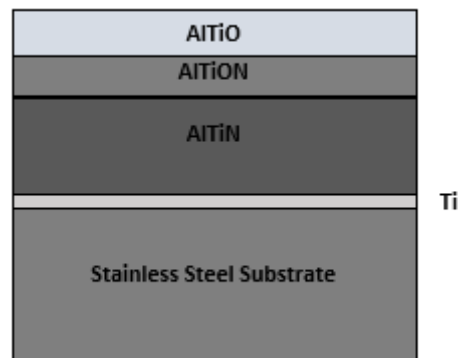
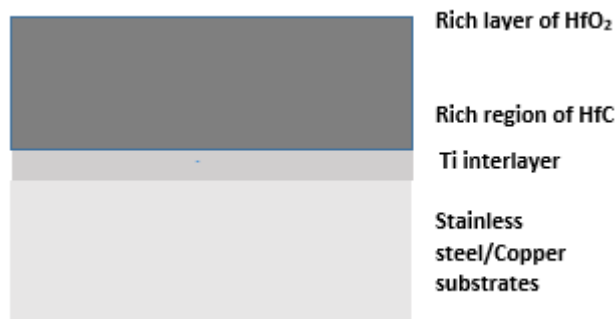


Figure 4: Tandem stack showing all individual layers [27]

Kushagra Hans et al., investigated the effect a HfC layer deposited on Ti metallic layer. The substrates used were SS, silicon as well as copper by means of sputtering technique. The coatings had high absorption of 0.925 and 0.948 on SS and copper substrates respectively. Emittance values obtained were moderate low with 0.13 for SS and very low with 0.03 on coated copper substrate. HfC gets oxidized forming HfO₂, which was revealed through a chemical analysis of coating. The upper portion of layer was rich in HfO₂, whereas lower portion of the layer was rich in HfC as evident in figure 5. Hafnium oxide layer thus formed acts like protective layer for HfC and aids in good absorption. Also HfC is a good candidate material for selective absorption with appropriate optical constant values. The structural analysis revealed that both HfC and HfO₂ had amorphous structures which in turn is more effective for high absorption[28].

Figure 5: Formation of HfO₂ as HfC gets oxidized [28]

Kondaiah et al., has tried to explore the optical properties of Tantalum carbide by developing a tandem stack consisting of a Titanium metallic layer, TaC and Al₂O₃. Titanium acts as a IR reflector layer as well as adhesion layer. In this tandem stack, TaC is the absorber layer and Al₂O₃ is the antireflection layer. The Ti layer is deposited using a Ti target and reactive direct current sputtering machine, TaC is deposited using Ta target in presence of C₂H₂ reactive gas by pulsed direct current sputtering device. The top layer is deposited using Al₂O₃ target, RF sputtering device. The coating was designed to achieve a high absorption by varying the process parameters of the main absorption layer TaC. Variation of reflectance value of Tantalum carbide for different thickness of titanium interlayer was evident in the results obtained. Chemical analysis revealed the formation of Tantalum oxide in the upper portion. A spectroscopic ellipsometry was used to calculate the refractive index of each layer in the stack separately. The metallic content decreases from top to bottom in the tandem stack as a result the refractive index also reduces which is 3.45, 2.34 and 1.56 for Ti, TaC and Al₂O₃ respectively. This gradient in the RI values aids in high absorption of solar energy. However, the extinction coefficient for Al₂O₃ is zero as it is a dielectric material. To analyze the thermal stability, annealing of the coating was done in vacuum at 500⁰ C for 2 h and noticed no change in coating behavior. However, there was reduction in absorption and reflectance by 3% at 600⁰ C. The coating exhibited a high absorption of 0.957 and moderately low value of emittance of 0.14[29].

V. SOLAR ABSORBERS WITH MOLYBDENUM IR REFLECTORS

Yuping Ning et al., has developed a tandem stack consisting of Molybdenum interlayer, Zirconium silicon nitride, zirconium silicon oxy nitrides and silicon di oxide. Here ZrSiN is the cermet absorption layer with higher metal volume fraction, whereas ZrSiON is a second cermet absorption layer with lower metal volume fraction. SiO₂ plays the role of anti-reflection as well as oxidation protective layer. For the optimized values of all layer thickness, an absorption of 0.94 and moderately low emittance of 0.12 was obtained for the tandem stack designed with a good thermal stability at a temperature of 500⁰ C in vacuum, after annealing. Optical constants of all layers were investigated and it was observed for Mo, refractive index(n) and extinction coefficient (k) increase with increase in wavelength which can be attributed to the metallic behavior. With optimized reactive gas flow rate first absorber layer ZrSiN and second absorber layer ZrSiON showed reduction of n and k values with increasing wavelength, which is an intermediate behavior between metals and dielectric materials. Whereas refractive index for SiO₂ was observed to be 1.45 and k was zero, indicating its antireflection property[30].

VI. SOLAR ABSORBERS WITH ALUMINIUM IR REFLECTORS

Muralidhar et al., has reported the performance of Aluminium layers deposited on ceramic substrates using direct current magnetron sputtering device, developed for the purpose of solar selective absorption. This study was carried out by varying process parameters of deposition such as power density of DC, reactive gas flow rate and rate of deposition. Characterization of Aluminium films was done to analyze different mechanical properties as well as optical properties. It was observed that the surface roughness of Al films had direct relation with flow rate of Ar gas and increased from 4 nm to as high as 21nm. Whereas the hardness of Al was observed to have increased with increased layer thickness. Also Al films exhibited high reflectance for wavelengths between 250 nm and 2500 nm[31].

Further Muralidha et al., has designed a multilayer stack consisting of three layers, first being Al₂O₃ as adhesion layer deposited on a ceramic substrate, second layer being Al as IR reflector and third layer being again Al₂O₃ playing the

role of antireflection as well as protection. The coating performance was analyzed for different thickness of Al_2O_3 with 100 nm, 200 nm and 300 nm. It was observed that, with increase in Al_2O_3 thickness the multilayer reflectance increased from 92 percent to 96 percent. This was attributed to the shift in reflectance with the influence of Al_2O_3 . No. The hardness and roughness of the coatings increased from 4.66 GPa to 6.83 GPa and 6.46 Ra to 22.99 Ra respectively, with increased thickness of Al_2O_3 . There was an increase in stiffness as well as modulus values of coating by 10 percent[32].

VII. CONCLUSION

Stainless steel is a ferrous alloy with intrinsic thermal emittance between 0.12 to 0.13, which is high and needs to be reduced when used as a substrate material in case of solar selective absorbers. When a transition metal like Tungsten was coated on stainless steel substrate the emission reduced to 0.03, the emittance of tandem stack $\text{AlTiN}/\text{AlTiON}/\text{AlTiO}$ was observed to be 0.08 with the inclusion of W interlayer. Emittance value was reduced to 0.07, while absorption being 0.961 in the multilayer $\text{TiAlC}/\text{TiAlCN}/\text{TiAlSiCN}/\text{TiAlSiCO}/\text{TiAlSiO}$ with the introduction of W coating, the lowest value of emittance of this coating can attributed the Al_2O_3 in addition to W interlayer. For $\text{W}/\text{AlN}/\text{WAlON}/\text{Al}_2\text{O}_3$ emittance was found to be 0.08, while absorption being 0.958. The multi-layer $\text{CrAlSiN}_x/\text{CrAlSiO}_x/\text{SiAlO}_x$ exhibited a very low emittance. $\text{W}/\text{AlSiTiN}_x/\text{SiAlTiO}_x/\text{SiAlO}_x$ absorption was 0.955 while emittance being 0.096. $\text{Ti}/\text{AlTiN}/\text{AlTiON}/\text{AlTiO}$ tandem stack had absorption of 0.933 and emittance 0.07 on copper substrates. When HfC was coated as a single layer on SS substrate with Ti interlayer the absorption was 0.925 and emittance 0.13, which can be attributed to high intrinsic emittance of SS, whereas for the same HfC single layer with Ti interlayer absorption was 0.948 and emittance was 0.03 for copper substrate. $\text{Ti}/\text{TaC}/\text{Al}_2\text{O}_3$ multi-layer had an absorption of 0.957 and emittance 0.15. there was an out diffuse of C from Tantalum Carbide which deteriorates the coating performance at temperatures above 500°C . $\text{Mo}/\text{ZrSiN}/\text{ZrSiON}/\text{SiO}_2$ at 500°C emittance was 0.12 and absorption was 0.94. It is evident from the above study that metal interlayers coated on substrates of SS, Cu, ceramic materials improved the absorption of the coatings and also prevent diffusion of substrate material into coatings.

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