

1 W/AlSiTiN_x/SiAlTiO_yN_x /SiAlO_x multilayered solar thermal selective absorber coating

2 A. AL-Rjoub^{a,b,1}, L. Rebouta^a, N. F. Cunha^a, F. Fernandes^c, N.P. Barradas^{d,e}, E. Alves^c

3 a Centre of Physics, University of Minho, Campus de Azurém, 4800-058 Guimarães, Portugal

4 b SEG-CEMMPRE, Department of Mechanical Engineering, University of Coimbra, Rua Luís Reis Santos, 3030-
5 788 Coimbra, Portugal

6 c Department of Control Engineering, Czech Technical University in Prague, Technicka 2, Prague 6 166 27, Czech
7 Republic

8 d Centre for Nuclear Sciences and Technologies, DECN, IST, University of Lisbon, Bobadela, Portugal

9 e Institute for Plasmas and Nuclear Fusion, IST, University of Lisbon, Lisbon, Portugal

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11

12 **Abstract**

13 Solar thermal energy has been used as a renewable green energy source. Here we present a
14 design of highly selective solar thermal absorber coating deposited by dc magnetron sputtering,
15 which is four layers based on W/AlSiTiN_x/SiAlTiO_yN_x/SiAlO_x. The coating revealed an excellent
16 average solar thermal absorbance $\alpha = 95.5$ % with very low emittance $\varepsilon = 9.6$ % (calculated for
17 500 °C) together with an excellent thermal stability at 500 °C, in air for 350 h, and 630 °C in
18 vacuum for 220 h.

19

20 **Keywords:** Selective coating, Sputtering, Thermal stability, AlSiTiN_x/SiAlTiO_yN_x

21

22 **Introduction**

23

24 Recently, there has been a significant increase in the levels of interest for the use of solar
25 thermal energy as a renewable green energy source. For example, the concentrated solar power
26 (CSP) is being used to produce the electricity by utilizing a parabolic-trough solar system, which
27 concentrates the sunlight onto evacuated tubes containing a selective absorber coating. Then, the
28 conducting heat can be used in steam turbines to produce electricity (Fernández-García et al., 2010)
29 (Odeh et al., 1998) (Kalogirou et al., 1997). On the other hand, the cost of such generated electricity
30 is relatively higher than the other resources. So, there is a need to improve the optical properties

¹ Corresponding author – e-mail address: abbasp185@yahoo.com (A. AL-Rjoub)

31 and the thermal stability of the coatings' materials to be competitive with the other resources
32 (Cagnoli et al., 2019). This can be done by improving the coatings' thermal stability such that
33 operating at higher temperatures for longer terms (Rebouta et al., 2015), which can be achieved by
34 smart selection of materials. Any design should have high solar absorbance (α) in the wavelength
35 region of the solar radiation and very low thermal emittance in IR region (to minimize the energy
36 losses by emission), good thermal stability and high oxidation resistance for long-term.

37 The current selective solar absorber coatings available in the market are two types: i) coatings
38 based on metal-dielectric nanocomposite coatings (Cermet), which are multilayered coatings
39 composed of a metal layer to improve the IR reflection as tungsten or molybdenum, interference
40 absorber layers (high absorber (HA) and low absorber (LA)), finished with an anti-reflection (AR).
41 As examples, the coatings based on Mo-Al₂O₃ (Zhang et al., 2006), Pt-Al₂O₃ (Nuru et al., 2012),
42 WTi-Al₂O₃ (Wang et al., 2017) and W-AlSiO_x (Dias et al., 2017). ii) multi-layered designs based
43 on transition metal nitrides and oxynitrides. This type showed a better thermal stability, oxidation
44 resistance and perfect selectivity, which made them the most common recommended materials for
45 the selective solar thermal absorbers used in CSP. The transition metal used in such coatings can
46 be Ti (Barshilia et al., 2007) (Du et al., 2013) (Rebouta et al., 2012a) (Soum-glaude et al., 2017),
47 Cr (Wang et al., 2019) (A AL-Rjoub et al., 2018), W (A. AL-Rjoub et al., 2018) (Dan et al., 2016),
48 Nb (Y. Wu et al., 2015), and Al (Rebouta et al., 2015) or a combination between them. All of these
49 optical stacks shared the designed structure as substrate\back-reflector metal\metal nitride\metal
50 oxy-nitride\anti-reflection oxide layer.

51 In our previous work, designs of solar absorber coatings based on nitride/oxynitride layers
52 (W/CrAlSiN_x/CrAlSiO_yN_x/SiAlO_x) (A. AL-Rjoub et al., 2018) and with structure (W/WSiAlN_x
53 /WSiAlO_yN_x/SiAlO_x) (A. AL-Rjoub et al., 2018) were studied. In this study we present a design
54 of highly selective solar thermal absorber coating based on W/AlSiTiN_x/SiAlTiO_yN_x/SiAlO_x for
55 high temperature applications.

56 1. Methods and experimental details

57 **1.1 Materials:** the materials used in this coating include: a) Several targets of 10 cm in
58 diameter with purity 99.99% as Ti50Al50 at% with 9 silicon pellets of 10 mm diameter added on
59 the target erosion zone (used to obtain the AlSiTiN_x and SiAlTiO_yN_x layers), Si80Al20 at% (used
60 to produce the oxide anti-reflection layer of the stacks) and W target which was used for the back-

61 reflector coating. b) Oxygen and nitrogen as reactive gases, and argon as sputtering gas. c) Glass,
62 p-doped Boron Si (100) mirror polished wafer and well-polished stainless-steel (AISI304)
63 substrates. d) Acetone and ethanol for substrate cleaning.

64
65 **1.2 Experimental description:** All coatings were produced at room temperature by dc-
66 magnetron sputtering with argon working pressure of 0.37 Pa, applied current density of 6.4
67 mA/cm², pulsed bias of -60 V and base pressure of 2×10^{-4} Pa), except the tungsten layer where
68 the used current density was 12.7 mA/cm². Glass substrates were cleaned by ethanol, whereas the
69 silicon and stainless-steel substrates were cleaned in acetone by ultra-sound 10 min and by ionic
70 etching. Several single layers of AlSiTiN_x and SiAlTiO_yN_x were deposited on substrates of glass
71 with changing the nitrogen and oxygen gasses flow, which used in simulation of the optical
72 constants and thickness. Thicker single samples similar to those in the final tandem were produced
73 on silicon and on stainless-steel substrates to study their morphology, chemical composition and
74 structural. The tandem was applied to annealing tests in air at 500 °C for 350 h and in vacuum at
75 630 °C for 220 h.

76 **1.3 Optical constants and simulated thicknesses:** the refractive index (n), extinction
77 coefficient (k) and thicknesses of thin AlSiTiN_x and SiAlTiO_yN_x layers were obtained from
78 SCOUT software (version 2.99, WTheiss Hardware and Software) simulation after measuring their
79 transmittance (T) and reflectance (R), using a Shimadzu PC3100 spectrophotometer, in the
80 wavelength range of 0.30 – 2.5 μm as completely described in our previous studies (A. AL-Rjoub
81 et al., 2018) (A. AL-Rjoub et al., 2018).

82 **1.4 Chemical composition, structural analysis and chemical-bonding of coatings:** EDS
83 with 10 keV accelerated voltage was used to evaluate the chemical composition of the coatings,
84 and cross-section SEM images of the coatings was used to study the morphology and thicknesses
85 calculations.

86 Structure and coatings' thermal stability in as deposited and after annealing was studied by X-
87 ray diffraction (XRD) and by the Rutherford Backscattering (RBS) (Barradas and Jeynes, 2008).

88 The chemical-bonding states of individual thick layers of AlSiTiN_x and SiAlTiO_yN_x was
89 investigated by using X-Ray Photoelectron Spectroscopy (XPS), taking in the account the C1s line
90 at 285.0 eV to calibrate the binding energies and the spectra were fitted by CasaXPS software
91 (version 2.3.19, Casa Softw. Ltd). The Ti 2p core level was fitted considering the peak doublets
92 spin-orbit separation (ΔE_p) 2p_{1/2} – 2p_{3/2} as 5.7 eV, 6.0 eV and 6.1 eV for oxides, nitrides and metal
93 states, respectively. with peaks' intensity ratio $\frac{I_{Ti\ 2p_{1/2}}}{I_{Ti\ 2p_{3/2}}} = 0.5$ (Biesinger et al., 2010) (Rahman et al.,
94 2016).

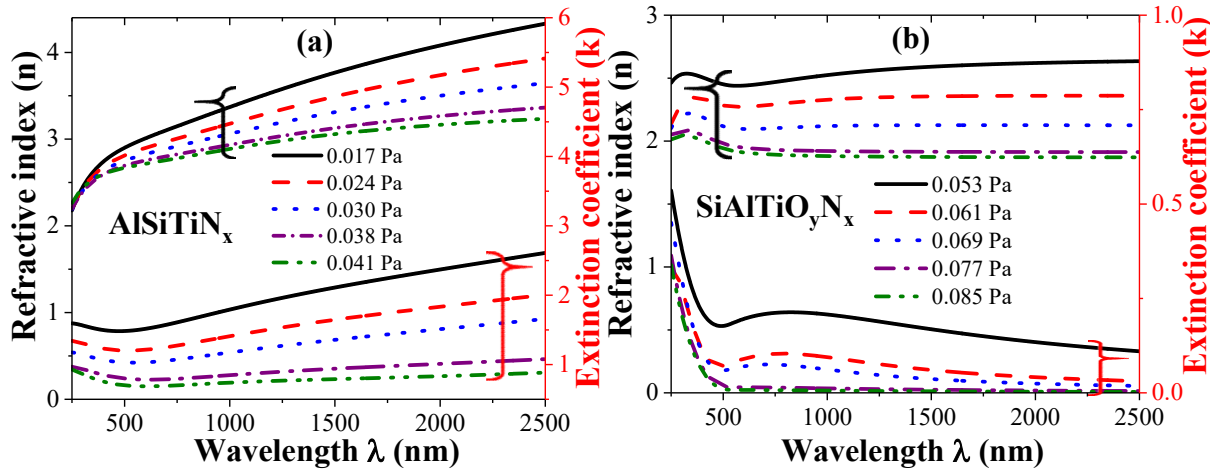
95 **2. Results and discussion**

96 **2.1.1 Single layers**

97 The back reflector tungsten and the antireflection SiAlO_x layers were studied in detail in our
98 previous studies (Dias et al., 2017) (A. AL-Rjoub et al., 2018) (A. AL-Rjoub et al., 2018). So,
99 here only AlSiTiN_x and SiAlTiO_yN_x are presented.

100 **2.1.2 Optical properties of single layers AlSiTiN_x and SiAlTiO_yN_x**

101 Several AlSiTiN_x and SiAlTiO_yN_x thin single layers were deposited on glass substrates with
102 different nitrogen partial pressures ranged from 0.017 to 0.041 Pa, in the case of AlSiTiN_x, and
103 with oxygen and nitrogen partial pressures ranged from 0.053 to 0.085 Pa in the case of
104 SiAlTiO_yN_x. The optical constants n and k of those layers were simulated from their experimental
105 reflectance and transmittance measurements with the help of SCOUT software. As shown in Fig.
106 1, n and k decreased as the partial pressures of the gases increased in both nitrides and oxynitrides
107 layers, due to the loss of their metallic behavior and due to the formation of transparent nitrides
108 and oxides of Al and Si. Also, the refractive indices of AlSiTiN_x increased in the wavelength range
109 of 300 – 1000 nm, which improves the solar absorption thus enhancing through the interference
110 effect (Yin and Collins, 1995). The refractive indices of SiAlTiO_yN_x layers behave similarly in
111 lower gas partial pressure and become constant for higher gas partial pressures.



112

113 **Fig. 1:** n and k as a function of wavelength of: (a) AlSiTiN_x and (b) $\text{SiAlTiO}_y\text{N}_x$, prepared with different
 114 reactive gases partial pressures.

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116 2.1.3 Chemical-composition of single layers

117 The chemical composition of $\text{AlSiTiN}_{P=0.038 \text{ Pa}}$ and $\text{SiAlTiO}_y\text{N}_{xP=0.069 \text{ Pa}}$ thick layers similar to
 118 the one used in the final absorber stack were evaluated by EDS and the results are presented in
 119 Table I. From nitride to oxynitride a strong increase of oxygen was found, as expected. A small
 120 decrease was found for aluminum and titanium at%, but in contrast a small increase in Si content
 121 was found. This is due to the effect of the addition of oxygen as a reactive gas, which induced a
 122 stronger target poisoning effect on Al and Ti than on Si, decreasing the relative sputtering yield of
 123 Al and Ti in comparison to Si, resulted in an increase of Si content. These differences explain why
 124 $\text{AlSiTiN}_{P=0.038 \text{ Pa}}$ has higher optical constants than $\text{SiAlTiO}_y\text{N}_{xP=0.069 \text{ Pa}}$ and as a result high and
 125 low solar thermal absorber layers, respectively.

126 **Table I:** The chemical composition of $\text{AlSiTiN}_{P=0.038 \text{ Pa}}$, $\text{SiAlTiO}_y\text{N}_{xP=0.069 \text{ Pa}}$, thick layers
 127 deposited on stainless-steel.

Layer	N at%	O at%	Al at%	Si at%	Ti at%	Ar at%
$\text{AlSiTiN}_{P=0.038 \text{ Pa}}$	33	-	26	22	17	2
$\text{SiAlTi(ON)}_{P=0.069 \text{ Pa}}$	16	22	22	25	12	3

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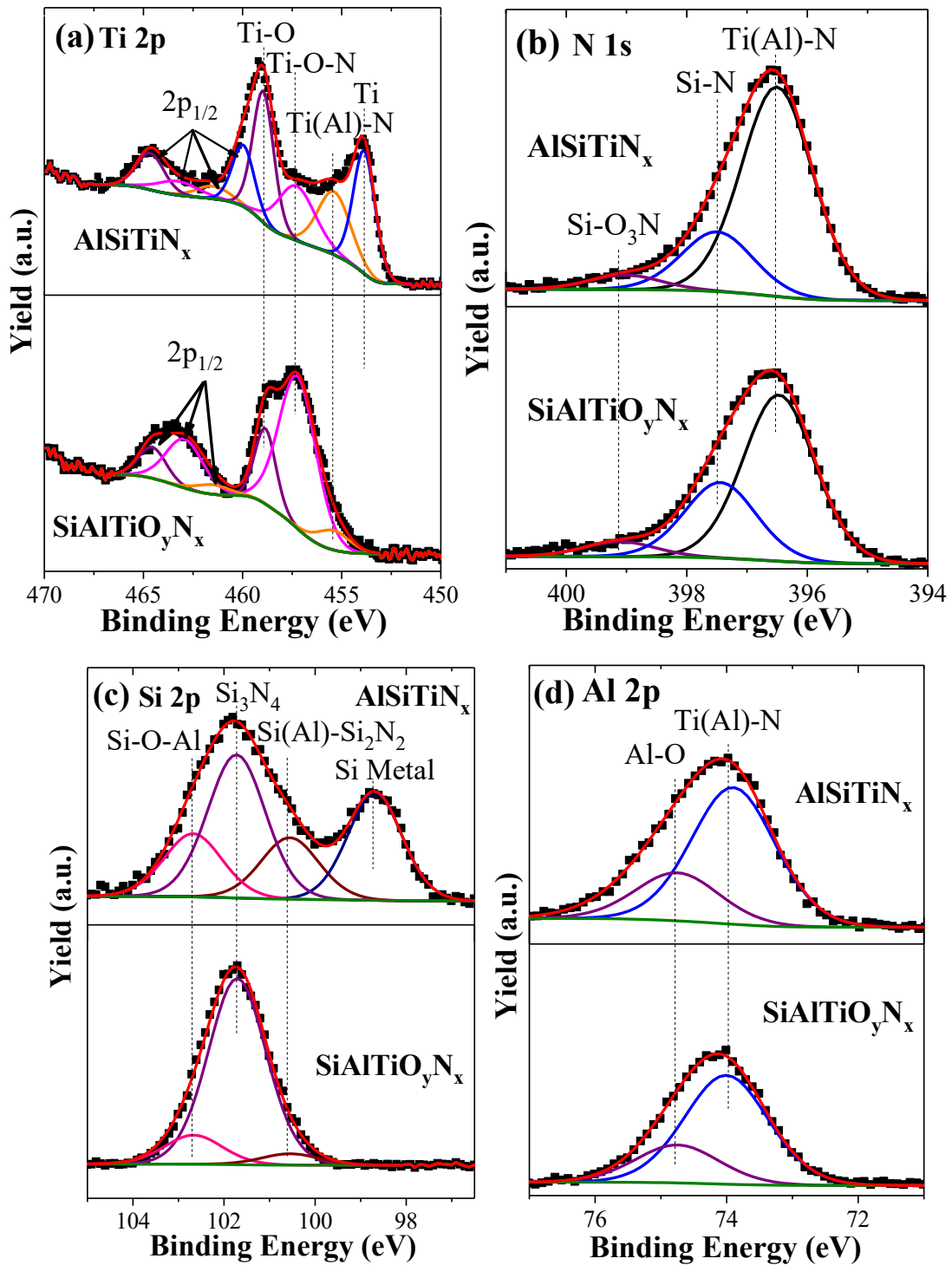
129 In order to evaluate the chemical bonding states, another individual samples of $\text{AlSiTiN}_{\text{P}=0.038}$
130 Pa and $\text{SiAlTiO}_y\text{N}_{\text{XP}=0.069 \text{ Pa}}$ similar to the used in the multilayered designs were surveyed by XPS.
131 Fig. 2 shows the spectral core level of Ti 2p, N 1s, Si 2p and Al 2p. Fig. 2a shows the Ti 2p core
132 level spectra, which show an evidence of the existence of different titanium oxidation states in
133 both samples. These peaks were divided into three doublet components considered as Ti $2p_{3/2}$ and
134 Ti $2p_{1/2}$. The Ti $2p_{3/2}$ peaks centered at 455.4, 457.3 and 459.0 eV can be corresponded to Ti(Al)-
135 N, Ti-O-N and Ti-O bonds (Zhang et al., 2017) (Zhao and Wackelgard, 2006)(Wainstein and
136 Kovalev, 2018) (Chen et al., 2019) (Rahman et al., 2016) (Chen et al., 2019) (Z. L. Wu et al.,
137 2015), respectively. In the nitride sample ($\text{AlSiTiN}_{\text{P}=0.038 \text{ Pa}}$), the Ti $2p_{3/2}$ peak at 453.9 eV
138 corresponds to the Ti atoms in metallic oxidation state (Biesinger et al., 2010) (Z. L. Wu et al.,
139 2015), which disappeared in the case of $\text{SiAlTiO}_y\text{N}_{\text{XP}=0.069 \text{ Pa}}$ sample.

140 The core level of N 1s can be deconvoluted into three peaks in the two samples as shown in
141 Fig. 2b. The peaks centered at energies 396.5, 397.5 and 399.0 eV associated to Ti(Al)-N, Si-N
142 and Si- O_3N compounds, respectively(Oliveira et al., 2015) (Zhang et al., 2017) (Rahman et al.,
143 2016) (A. AL-Rjoub et al., 2018).

144 Fig. 2c the Si 2p core-level, where the two specimens have the common three peaks located
145 at 100.6, 101.7 and 102.6 eV, which they recognized as Si(Al)- Si_2N_2 , Si_3N_4 and Si-O-Al,
146 respectively (Oliveira et al., 2015) (Costa et al., 2018)(Rebouta et al., 2015) (AL-Rjoub et al.,
147 2017). The fourth peak of nitride sample ($\text{AlSiTiN}_{\text{P}=0.038 \text{ Pa}}$) centered at 98.7 is considered as Si
148 elemental bond (Z. L. Wu et al., 2015) (Oliveira et al., 2015).

149 Finally, Al 2p core level is shown in Fig. 2d, the two samples share the components centred at
150 73.9 and 74.8 eV related to Ti(Al)-N and Al-O, respectively(Z. L. Wu et al., 2015) (Wainstein and
151 Kovalev, 2018) (Costa et al., 2018) (Rahman et al., 2016).

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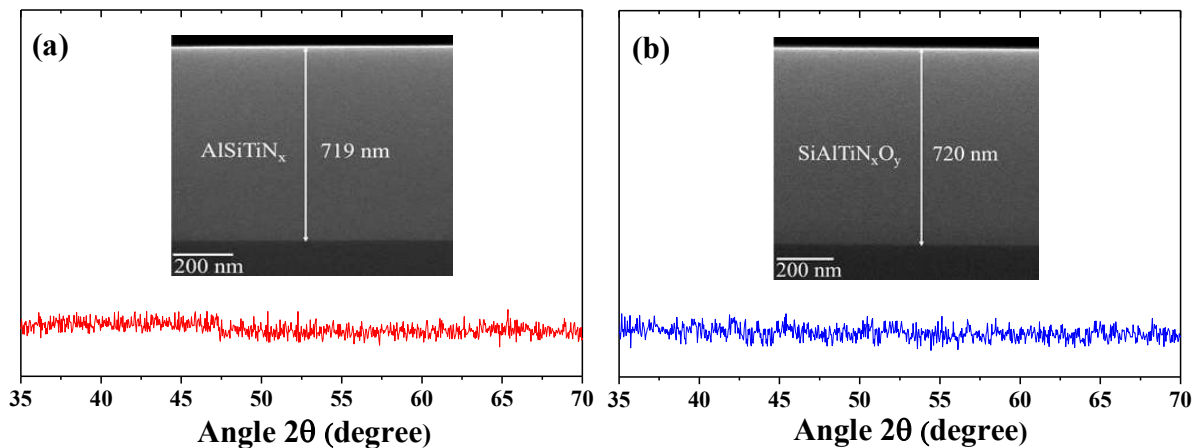
155 **Fig. 3.** XPS-spectra of: (a) Ti 2p, (b) N 1s, (c) Si 2p, (d) Al 2p core level of $\text{AlSiTiN}_{xP=0.038}$ Pa and

156 $\text{SiAlTiO}_y\text{N}_{xP=0.069}$ Pa single layers. The green line in figures is the background correction.

157

158 **2.1.4 Morphology and crystalline structure of single layers**

159 Thick individual layers of $\text{AlSiTiN}_{\text{P}=0.038}$ Pa and $\text{SiAlTiO}_y\text{N}_x\text{P}=0.069$ Pa deposited on stainless steel
160 substrates were used to study their crystalline structure by X-ray diffraction (XRD). Both samples
161 are amorphous as also no features appears in SEM as presented in Fig. 3.

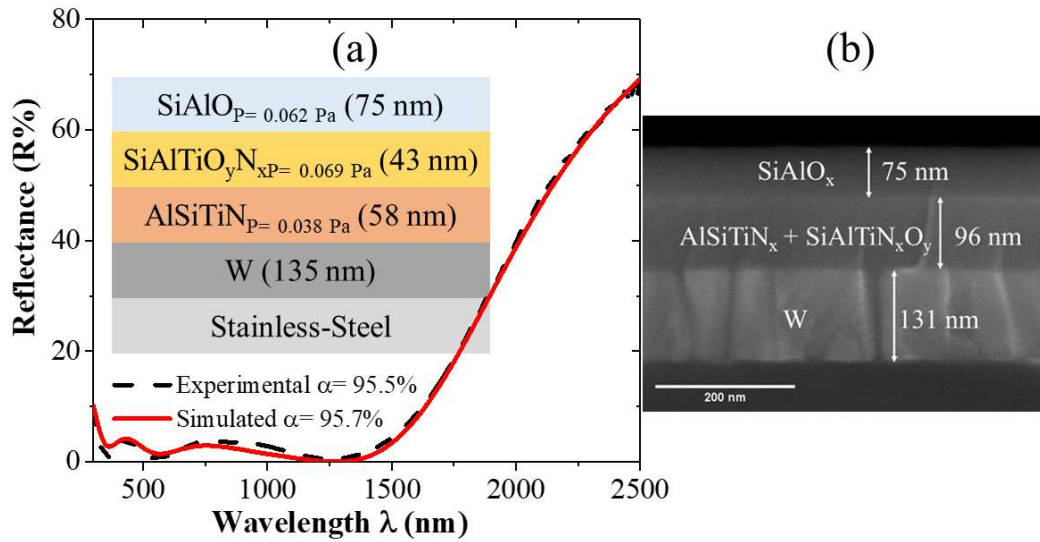


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163 **Fig. 3:** XRD-pattern and cross-section SEM images of: a) $\text{AlSiTiN}_{\text{P}=0.038}$ Pa and b) $\text{SiAlTiO}_y\text{N}_x\text{P}=0.069$ Pa thick
164 single layers.

165 **2.2 Multilayer Design**

166 The multilayer stack coatings were previously modelled by SCOUT-software depending on
167 their single layers' information, that allows the calculation of their optical constants n and
168 extinction k as a function of wavelength, together with their thicknesses. Then the structure and
169 the reflectance curve of the final stack was simulated, and deposited on 304 stainless-steel
170 substrates. The experimental total reflectance of the stack was also measured and as shown in Fig.
171 4a.



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173 **Fig. 4:** (a) Schematic-diagram of the simulated optical tandem with corresponding simulated and
 174 experimental reflectance curves. (b) SEM Cross-section SEM image of the multilayered coating optical
 175 tandem.

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As it is also displayed in Fig. 4a, the tandem is a multilayer composed of four layers: i) the tungsten layer, with thickness ~ 131 nm as back-reflector to improve the whole reflectance in IR wavelength region. ii) $\text{AlSiTiN}_x / \text{SiAlTiO}_y\text{N}_x$ two absorption structural layers, which they are undistinguishable in the SEM image. iii) SiAlO_x as antireflection protective layer, which is protect the inner layers and allow the light to pass through. The reflectance of as deposited tandem is in agreement of the simulated one and has a thickness of ~ 301 nm, and it shows a very high solar absorptance, $\alpha = 95.5\%$ with very low emissivity, $\varepsilon = 9.6\%$ (calculated for 500°C using the IR reflectance measurements) and $\varepsilon = 7.8\%$, when calculated for 400°C , simultaneously.

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2.3 Thermal stability

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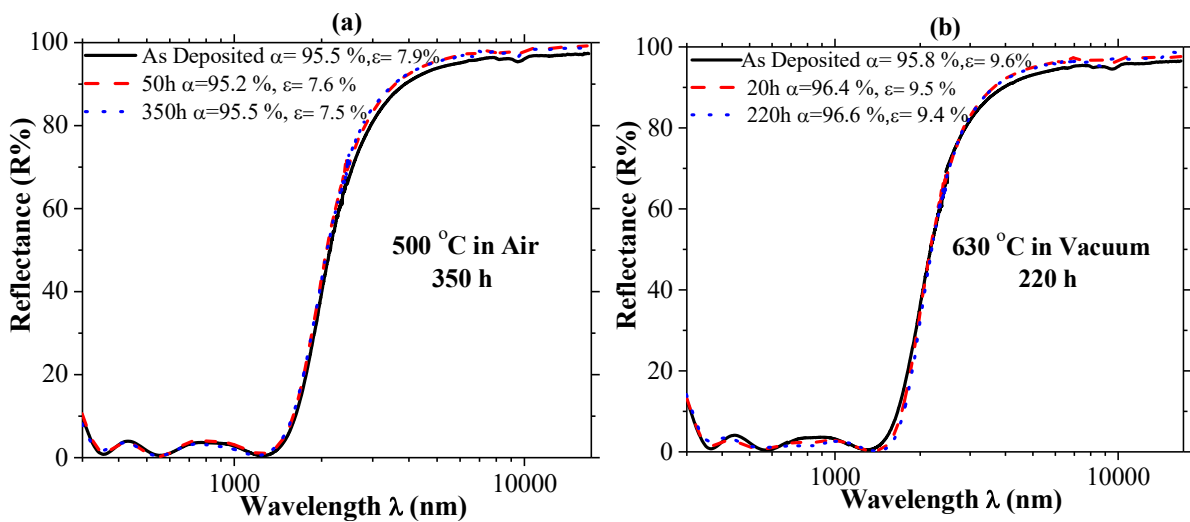
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The thermal stability and oxidation resistance of the tandems were tested by subjecting them to annealing tests in air, at 500°C , and in vacuum, at 630°C , for 350 h and 220 h, respectively. After each step of annealing, the absorptance (α) and the thermal emittance (ε) were calculated from the reflectance curves. Fig. 5a shows the reflectance curves of as deposited, after 50 h and after 350 h air annealing, with their solar absorptance and thermal emittance at 500°C after each step shown in the legend. As seen in the figure, the coatings show an excellent thermal stability and no changes reported in the reflectance curve, solar absorptance and thermal emittance values in the second annealing step (of 350 h), which reflect a very good thermal stability. However, after the first step of annealing, small changes in the reflectance curve especially for wavelengths greater than 2000

194 nm due to tungsten phase structure change (α and β phases), which is normal and fully studied in
 195 the previous studies (AL-Rjoub et al., 2019) (Chen et al., 2005) (O’Keefe and Grant, 1996)
 196 (Antonaia et al., 2010). The vacuum annealing was performed in two steps, 20 h and 220 h, as
 197 shown in Fig. 5b, the tests insure a very good thermal stability. Although those results were
 198 obtained at higher testing annealing temperatures in air and in vacuum than the one of the coating
 199 based on AlSiO_x:W(Dias et al., 2017), WSiAlN_x/WSiAlO_yN_x(A. AL-Rjoub et al., 2018) and/or
 200 CrAlSiN_x/CrAlSiO_yN_x(A AL-Rjoub et al., 2018) (Al-rjoub et al., 2019), they showed better
 201 thermal stability. Moreover, the emittance values calculated at 400 °C of this design is lower than
 202 the later.

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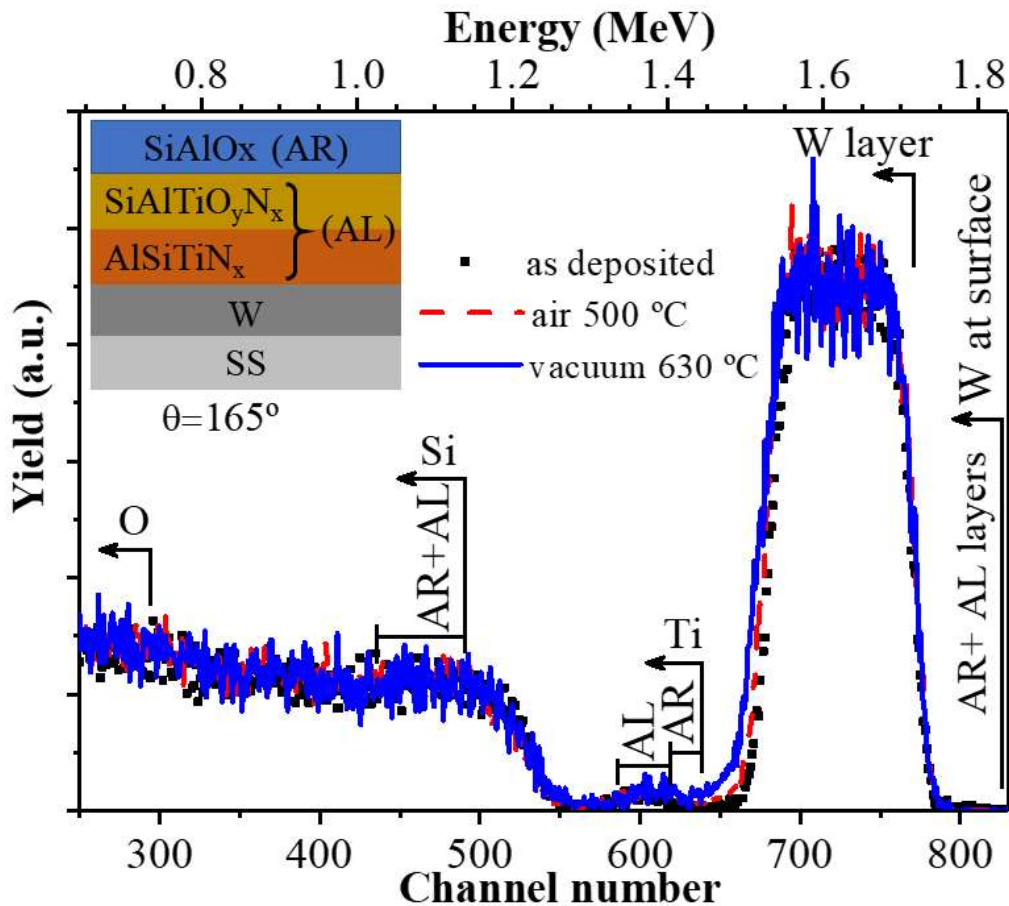


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205 **Fig. 5:** the reflectance curves of as deposited tandems and after annealing (a) in air at 500 °C (b) in vacuum
 206 at 630 °C, with measured values of the solar absorptance (α) and the thermal emittance (ϵ) (calculated for
 207 500 °C).

208 Rutherford back scattering (RBS) was also used to study the elemental depth profiles of the absorbers,
 209 as shown in Fig. 6. It shows three optical stack coatings deposited in same run and subjected to different
 210 thermal annealing, namely as deposited, after air annealing at 500 °C for 350 h and after vacuum annealing
 211 at 630 °C for 220 h. The composition of elements was determined from their front edges because of the
 212 signals can overlap as indicated in the figure. The analysis shows that the three as deposited samples have
 213 a small difference in the thickness of W layer. The relative heights of these front edges are correlated due
 214 to the relative concentrations of the different elements, and the position of the different elements, if located
 215 at surface sample, are indicated in the graph. It is difficult to distinguish between silicon and aluminum

216 because they have very close atomic masses. Nevertheless, the analysis shows a small difference between
 217 as deposited and vacuum annealed samples. After air annealing no changes were recorded for the coating
 218 reflecting an excellent thermal stability against diffusion and oxidation. For annealing in vacuum, a small
 219 changes of W depth profile were seen due to its small elemental diffusion to the stainless-steel substrate,
 220 which can be easily solved by adding a barrier layer as shown in the previous study (AL-Rjoub et al., 2019).



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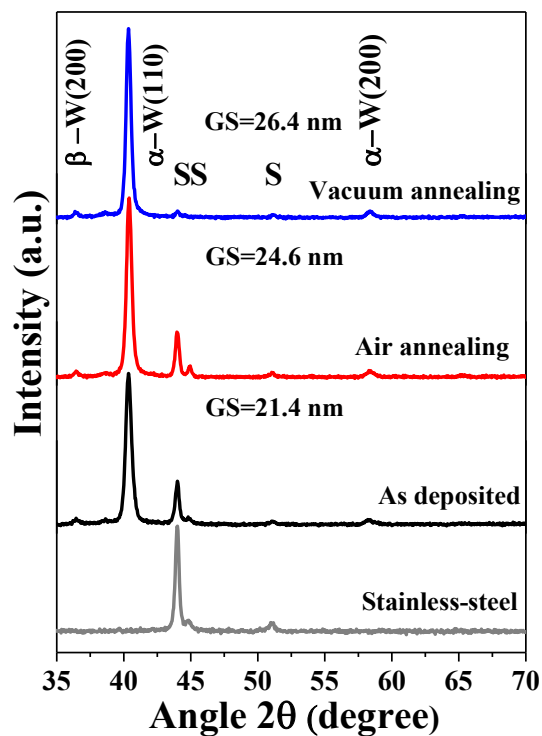
222 Fig. 6: The spectra of RBS of the stacks as deposited and after air thermal annealing at 500 °C and
 223 vacuum thermal annealing at 630 °C for 350h and 220 h, respectively.

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225 Further structural tests were applied on the coatings after the thermal treatments using XRD to
 226 analyze if it whether or not it has some changes due oxidation. Fig. 7 shows the XRD
 227 diffractograms of the three samples (as deposited, after air and vacuum annealing). As shown in
 228 the figure, no changes have been recorded after the thermal treatment , except small differences in
 229 the stainless-steel peaks and in the intensity of W peaks, which reflecting that the coatings showed
 230 a very good oxidation resistance and its thermally stable after annealing in air and vacuum at 500

231 °C and 630 °C, respectively. The W recrystallization induced by the thermal annealing also
 232 contribute to the tungsten phase structure change (AL-Rjoub et al., 2019) (Chen et al., 2005)
 233 (O’Keefe and Grant, 1996) (Antonaia et al., 2010), which is related with the reflectance increase
 234 in the infrared wavelength range resulting in a consequent emissivity decrease. The W grain sizes
 235 were calculated from the full width at half maximum (FWHM) of (110) tungsten peak and using
 236 the Scherrer equation, in as deposited state and after annealing. The grain size of as deposited
 237 sample was 21.4 nm, and a slight increase was found, 24.6 nm and 26.4 nm after the annealing in
 238 air, at 500 °C, and in vacuum, at 630 °C, respectively.

239



240

241 **Fig. 7:** X-ray diffraction of tandems before and after thermal annealing.

242 The structure based on AlSiTiN_x/SiAlTiO_yN_x showed better performance than the structure based on
 243 TiAlN_x/TiAlO_yN_x (Barshilia et al., 2007) (Rebouta et al., 2012b) (Du et al., 2013). In general the structure
 244 based on Me1(Me2)SiN/Me1(Me2)SiON, where Me1 is a transition metal element and Me2 is Al or another
 245 transition metal, revealed better performance than those based on Me1(Me2)N/Me1(Me2)ON. This
 246 behaviour was also reported for CrAl (A AL-Rjoub et al., 2018) (Al-rjoub et al., 2019), Cr (Zou et al.,

247 2015) and NbTi (Y. Wu et al., 2015), which means that the Si addition improves the thermal and chemical
248 stability.

249 Silicon usually assumes fourfold coordination and transition metal in respective nitrides (fcc lattice) are six
250 fold coordinated, and with high surface mobility during the deposition, Si atoms are segregated, allowing
251 the nucleation of the SiN_x phase, which forms a layer on the growth surface covering the nitride
252 nanocrystallites and limiting their growth, resulting in a two phase material with improved
253 properties (Vepfek and Reiprich, 1995) (Veprek and Veprek-heijman, 2008). Thus, the oxidation resistance
254 and barrier diffusion properties of transition metal nitrides and transition metal oxynitride films can be
255 improved by the addition of Si (Vepfek and Reiprich, 1995) (Thobor-keck et al., 2005) (Kim et al., 2006)
256 (Najafi et al., 2013)

257 Silicon in low concentration and with low surface mobility can be incorporated in solid solution
258 phase (Traverse et al., 2001) (Eriksson et al., 2014), but with increasing concentrations, the Si segregation
259 already occurs with the formation of an amorphous phase of either silicon nitride or free silicon.

260 With N deficiency and with relatively high Si content, metal silicides can be formed, which are stable at
261 high temperature due to their low vapour pressure and refractory nature. The presence of free Si has been
262 proved to be efficient in the oxidation resistance improvement, as reported by Llauro et al (Llauro et al.,
263 1998) demonstrating that TiNSi codeposits improves the oxidation resistance as compared to CVD TiN,
264 and for greater amount of titanium silicide, it has better improvement of oxidation behavior. The tested
265 samples had 2, 13 and 17 at. % of Si and 45, 35 and 32 at. % of nitrogen, respectively. The amorphous
266 phases are usually better to prevent the layer further oxidation than a polycrystalline layer, where grain
267 boundaries provide a number of diffusion paths for oxygen. With the formation of an amorphous layer the
268 number of diffusion paths decrease considerably leading to the passivation regime (Llauro et al., 1998).

269 In the design with two absorption layers (high and low absorption layers), it is necessary to adjust the
270 nitrogen content to obtain the desired optical properties. This usually means that the nitride layer is not
271 stoichiometric, having some nitrogen deficiency, resulting that part of the metals atoms are in the metallic
272 oxidation state. In the AlSiTiN_x layer, as shown in section 2.1.3, part of Ti and Si atoms are in metallic
273 oxidation state. In the structure based on $\text{WSiAlN}_x/\text{WSiAlOyN}_x$ layers (A. AL-Rjoub et al., 2018), the
274 nitride layer exhibited some W atoms in the metallic oxidation state and in the case of absorber stack
275 based on $\text{CrAlSiN}_x/\text{CrAlSiN}_x\text{O}_y$ (AL-Rjoub et al., 2019) absorption layers, the nitride also
276 revealed some Cr and Si atoms in the metallic oxidation state.

277 The Si content can also play an important role in the performance of this type of coatings. In case
278 of Si addition to CrN, Kim et al (Kim et al., 2006) reported that $\text{Cr}_{0.67}\text{Si}_{0.33}\text{N}$ revealed better
279 oxidation resistance than $\text{Cr}_{0.78}\text{Si}_{0.22}\text{N}$ (Kim et al., 2006). The performance of an absorber coating

280 based on CrAlSiN_x/CrAlSiN_xO_y exhibited a dependence with Si content(A AL-Rjoub et al.,
281 2018) (Al-rjoub et al., 2019) with the best performance for a Si content close to 20 at. % in a nitride
282 with about 27 at.% of nitrogen. The AlSiTiN_x/SiAlTiO_yN_x layers have 22 and 25 at.% of Si,
283 respectively, but with the nitride with 33 at. % of nitrogen. The Si and N contents can justify why
284 this design has a better performance than the one based on CrAlSiN_x/CrAlSiN_xO_y. The structure
285 based on WSiAlN_x/ WSiAlO_yN_x layers also exhibited excellent performance. The WSiAlN_x
286 layer had 20 at. % of Si and 29 at. % of nitrogen, while the WSiAlO_yN_x layer had 20 at% of Si.
287 These samples have excellent thermal and chemical stability and have in common a Si content of
288 about 20 at. % the nitride with around 30 at. % of nitrogen(A. AL-Rjoub et al., 2018).

289 **3. Conclusions**

290 A multilayered tandem coating of selective solar radiation absorption for high temperature
291 applications was designed and deposited by dc magnetron sputtering. The stack is based on four
292 layers of structure (W/AlSiTiN_x/SiAlTiO_yN_x/SiAlO_x); back reflector/high absorber/low
293 absorber/antireflection layer, respectively. The structural, optical and chemical properties of their
294 single layers were studied. With increasing nitrogen and oxygen partial pressures during
295 deposition, AlSiTiN_x and SiAlTiO_yN_x layers lost their metallic behavior and become more
296 transparent. The increase of the nitrogen and the oxygen contents, induced the formation of Si and
297 Al nitrides and oxynitrides, resulting in a decrement of their optical constants n and k. The two
298 absorbing layers AlSiTiN_x and SiAlTiO_yN_x are amorphous, as confirmed by the XRD analysis.
299 The XPS analyses confirm the formation of metals nitrides, oxynitrides, and oxides bonding.
300 However, the AlSiTiN_x, layer has some Ti and Si atoms in metallic oxidation state. The multilayer
301 coating was theoretically designed based on the obtained data from the single layers and then
302 deposited on stainless-steel substrates. The total thickness of as deposited coating is about 302 nm,
303 and shows simultaneously high average solar absorptance, $\alpha=95.5\%$, and very low emissivity, $\varepsilon=$
304 9.6% (calculated for 500 °C), together with high thermal stability at 500 °C, in air and at 630 °C,
305 in vacuum, for 350 h and 220 h, respectively. The thermal emissivity improved for all samples,
306 ensuring that the W back reflector layer maintained its reflectivity and it is well protected by the
307 other top layers. With these excellent results, we strongly recommend this choice of design and
308 materials for high temperature thermal applications, especially for CSP coating for electricity
309 production.

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