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INNOVATIVE AND ADVANCED MATERIALS RESEARCH FOR HIGH TEMPERATURE SOLAR RECEIVERS

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Abstract

Achieving high temperatures in solar receivers is crucial for efficient and competitive solar thermal power plant operation. However, high temperature receivers enabling working temperatures beyond 1000 ºC are still subject of research projects. The fundamental problems observed are related to material durability and reliability. Thus, basic material research forms the core requirement regarding the engineering of advanced solar receivers that will most probably enable a highly efficient conversion of solar energy into electricity, paving the way for a broad establishment of clean and renewable energy supply. This work focuses on the presentation of fundamental research of high performance materials that allow the later development of high temperature and highly efficient solar receivers. It presents optical and thermal aspects of advanced materials.

Key-words: Advanced solar receiver materials; high temperature ceramics and metals; high temperature experiments

1. Introduction

Solar thermal power is a highly promising way of providing renewable electricity as it directly harnesses the abundant amount of solar energy incident on planet earth (Abbott, 2010), and additionally, features the important possibility of thermal energy storage and hybridization, enabling dispatchable power generation. One of the current technological challenges is the development of efficient ways of solar energy collection for its transformation into high quality heat, being able to efficiently run state-of-the-art thermodynamic power cycles for electric power generation.

The majority of today's CSP plants are based on the parabolic trough collector technology that has been established on commercial level since the 1980's (SEGS plants in California, USA(Kolb, 1994)). Back then, annual solar-to-electric conversion efficiencies achieved values up to 10.6% (Kolb, 1994) and are nowadays still not higher than 14 to 15% (Price, 2003). Achieved peak solar-to-electric conversion efficiencies are in the range of 20 to 25% (Reddy et al., 2013). Fundamentally, this efficiency limitation is due to the limited operating temperature (≈ 400 ºC) of the applied heat transfer fluid (thermal oil) (Solutia-Inc., 2008).

Clearly, the move to other heat transfer fluids that enable higher operating temperatures is a must. Viable options are for example molten salts (upper limit at about 600 ºC (Bradshaw and Carling, 1987)) and air. Another possibility is the direct steam generation, where the working fluid of the Rankine cycle is directly evaporated in the solar receiver. However, also a solar collector/receiver technology change is inevitable,

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since higher receiver operating temperatures are only feasible with high area concentration ratios (Romero-Alvarez et al., 2007). Instead of the conventional parabolic trough technology, the power tower concept is in this context much more favorable.

Generally speaking, in order to make solar thermal power competitive with conventional power generation, the efficiencies of the solar receivers as well as that of the thermodynamic power cycles have to be maximized. Since the conversion efficiency of the thermodynamic power cycle is ideally constrained by the Carnot efficiency, the temperature at heat input must thus be kept as high as possible and the temperature at heat rejection as low as possible. Given the fact that the temperature at heat rejection is determined by the ambient temperature, which is usually quite high at typical CSP locations, the only feasible way of improving the conversion efficiency are high temperatures at heat input, i.e. high temperatures of the plant's solar receiver. However, the thermal efficiency of the solar receiver is governed by the heat losses to the relatively cold environment. Thus, given that the specific heat losses (per area) increase highly non-linear with increasing receiver operating temperature (especially the radiative losses), advanced and innovative receiver designs are indispensable and area concentration ratios must be high (concentrated solar power).

On the one hand, when thinking of the optimization of thermodynamic power cycles, al lot of fundamental research work has already been accomplished for the conventional power technology and the current technology available needs in principle only slight adaptions for the application in solar power generation. Thus, an obvious step in engineering is the application of combined cycles in solar power generation, i.e. the application of solar driven Brayton cycles combined with bottoming Rankine steam cycles (Kribus et al., 1998).

On the other hand, the development of high temperature receivers that reliably work in temperature ranges beyond 1000 ºC is still in its early stages and currently being investigated (Ho and Iverson, 2014). The fundamental problems observed are related to material durability and reliability. Thus, basic material research forms the core requirement regarding the engineering of advanced solar receivers that will most probably enable a highly efficient conversion of solar energy into electricity, paving the way for a broad establishment of clean, renewable and safe energy supply.

Some pioneering work can already be found in the literature, especially regarding accelerated aging tests that are important when it comes to the estimation of solar receiver service lifetime. For example, Rojas-Morín and Fernández-Reche (2011) performed accelerated aging tests for the solar absorber material candidate Inconel 625CF at a parabolic dish located at the Plataforma Solar de Almería. They created Stress-Life (S-N) fatigue curves, also known as Wöhler curves. They observed that their defined normal operational conditions fell within the high cycle fatigue region, however when moving to higher operating temperatures (above 800 ºC) thermal stresses easily approached the ultimate strength leading to material failure at a low number of cycles. Also Boubault et al. (2012) presented a relevant work about accelerated aging of solar absorber materials. They developed a two-dimensional model of a two-layer material (metal + paint coating) in order to determine the optimal conditions for accelerated aging via simulations. Several thermal indicators (temperature and temperature gradients) were analyzed in different configurations and boundary conditions (irradiance cycles) and the most appropriate loads for accelerated aging tests were selected. To confirm the simulation results with experimental data, they developed an experimental solar accelerated aging facility. Further experiments with Inconel 625 are described in their subsequent work (Boubault et al., 2014). The typical irradiance patterns to which the material samples were exposed were square-shaped pulses with constant period and amplitude. The solar absorptance of the coating, its thermal conductivity, and the thermal contact resistance between the coating and the substrate were determined as characteristic parameters that determine the state of aging of the material.

The objective of this article is the presentation of some fundamental material research activities regarding promising candidate materials for high temperature solar receiver design for the power tower technology. As already mentioned in previous works (Rojas-Morín and Fernández-Reche, 2011; Boubault et al., 2012; Boubault et al., 2014), solar receiver material must withstand relatively harsh operating conditions, first of all, due to elevated operating temperatures (> 1000 ºC) of future designs (aiming for highest possible conversion efficiencies of the thermodynamic power cycle), and secondly, high heat fluxes of typically > 1 MW/m^2 that in addition show large and sudden variations (due to passing clouds) causing thermal fatigue. This paper aims at exploring the suitability of high-temperature intermetallics, refractory steels, superalloys, as well as dense and porous silicon oxycarbides for the possible application in such harsh and demanding conditions. Similarly to the abovementioned scientific contributions, this paper aims at performing material aging tests. Furthermore, optical properties of the candidate materials will be analyzed.

This work is structured as follows: Section 2 presents candidate materials which may be suitable for solar receiver designs. Section 3 will present some laboratory-scale high-temperature and thermo-shock experiments, applying on the one hand a tubular oven and on the other hand a Fresnel lens. Section 4 will deal with the evaluation of optical properties of relevant material samples. Section 5 concludes.

2. Materials under consideration

2.1. Exploration of high-temperature intermetallics in comparison with refractory steels and superalloys (at CSIC-CENIM)

Heating and oxidation experiments were carried out on a wide range of metallic and intermetallic alloys. The materials may be classified into several different groups as: (a) refractory steel; (b) iron aluminides; molybdenum silicide or boro-silicide intermetallics; and (c) nickel-base superalloy. These alloys were prepared by a variety of methods depending on whether there were any problems of chemical segregation during solidification. The refractory Ni-Cr-Fe steel was designated ET-45 (supplied by Schmidt-Clemens and prepared by centrifugal casting). The iron aluminides had Al contents in the range 20-40 atomic percent and were prepared by drop-casting or mechanical alloying, with grain sizes in the range 1 μm to 1 mm. Molybdenum silicides were prepared by mechanical alloying as a $MoSi_2$-15%SiC composite or a Mo-Si-B alloy containing a body centered cubic (alpha) Molybdenum matrix with high content of T2 and Mo_3Si phases. The Ni superalloy was a spray-formed Ni_3Al-rich alloy with high Cr content.

2.2 Exploration of dense and porous silicon oxycarbides (SiOC) (at CSIC-ICV)

Advanced dense and porous silicon oxycarbide (SiOC) material samples have been prepared at CSIC-ICV. A preceramic organic-inorganic hybrid material was synthesized by the sol-gel process, and then it was pyrolyzed under inert atmosphere at 1100 ºC to obtain a porous SiOC material. At the same time, the dense SiOC material was obtained by the application of the conventional ceramic route over the above mentioned hybrid. Thus, the hybrid material was attrition milled and pyrolyzed under inert atmosphere at 1100 ºC and finally the SiOC powders were then shaped and densified at high temperature. A description of the processing technique can be found in the work of Mazo et al. (2013).

3. Laboratory-scale high-temperature and thermo-shock tests

3.1. Laboratory high-temperature tests

The resistance to high temperature has been tested by exposing the sample to 1000 ºC during 100 cycles. The temperatures, dwelling time at 1000 ºC, and heating and cooling rates of a high temperature cycle are given in Table 1. The equipment available for this project is a small tubular oven (Fig. 1) with a maximum temperature up to 1400 ºC.

Figure 1: Tubular oven used for high temperature tests at CSIC-ICV

Table 1: High temperature test cycle (T_i, T_f: initial and final temperatures; v: heating and cooling rate; t: time; t_{ac}: accumulated time)

T_i (ºC)	T_f (ºC)	v (ºC/s)	t (s)	t_{ac} (s)
25	800	70	11	11
800	1000	5	40	51
1000	1000	0	600	651
1000	600	11	36	687
600	400	4	50	737
400	200	1	200	937

The high temperature experiments were performed with three different samples: SiC (used as reference: Ref), and a porous (S1) or a dense (S2) SiOC laboratory prepared materials. Figure 2 shows how the samples were placed within the tubular furnace and the appearance of the samples during the high temperature treatment. Figure 3 collects the photographs of samples before and after high temperature cycles. It is observed that as we reported in a preceding work of Sallaberry et al. (2015) (accepted for publication in the SolarPACES Proceedings 2014) the Ref sample crushed after the third cycle so the subsequent test were carried out only with SiOC samples. On the other hand, it is observed that both porous and dense SiOC samples (S1 and S2) remain visually unchanged during the 10 cycles and furthermore remain unchanged even after 100 cycles.

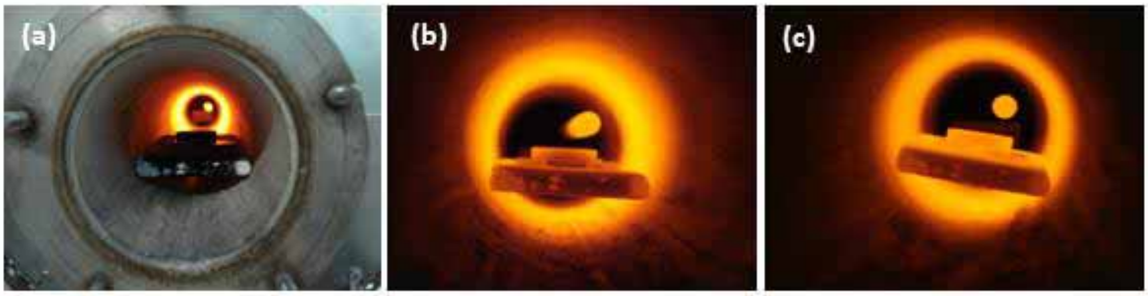

Figure 2: Details of tubular furnace during the high temperature tests. (a) Starting the test. (b and c) Sample at 1000 ºC for 0 and 600 s.

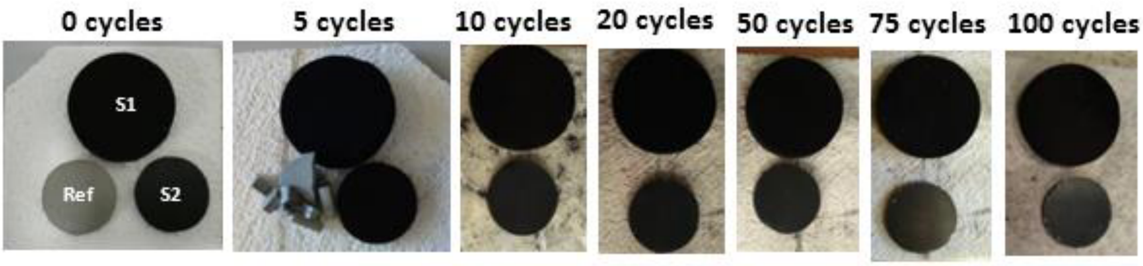

Figure 3: Photographs of materials before and after high temperature tests (cycle of Table 1).

During high temperature treatments under air atmosphere the SiOC materials experiment several processes due to an oxidative thermal degradation reaction. At low temperatures these materials experiment a weight loss associated basically to the thermal decomposition of the C_{free} phase reaction (1). Above 800 ºC the SiOC matrix can be decomposed according to reaction (2) and as a result a weight gain is observed. Finally, at higher temperatures the SiC can be oxidized and a new weight gain is also observed following reaction (3). (Bois et al., 1995; Chollon, 2000; Parmentier et al., 2001)

$C_{free} + O_2 \rightarrow CO_2 + H_2O$ (1)

$SiOC + O_2 \rightarrow SiO_2 + CO_x$ (2)

$SiC+O_2 \rightarrow SiO_2+CO_x$ (3)

For the SiOC materials prepared in our study although after 10 cycles it was not observed any change in weight or dimension for both S1 and S2 samples (Sallaberry et al., 2015), during the subsequent cycles the S1 sample experimented both weight and dimensional losses close to 4 and 2 %, respectively, associated to C_{free} phase oxidation (reaction 1) but the S2 sample remained unaltered.

On the other hand, the evaluation of the material characteristics was completed by the measure of the solar reflectance with a spectrophotometer (Perkin Elmer Lambda 40) with integrating sphere, from 400 nm to 1100 nm. As can be seen in Figure 4, S1 samples' reflectance values are practically not influenced by the performed temperature test cycles except for that treated after 100 cycles where the reflectance changes. However in the case of S2 sample, between 0 and 10 cycles the reflectance values show a decrease but from to 10 to 100 cycles test samples' reflectance does not change at all.

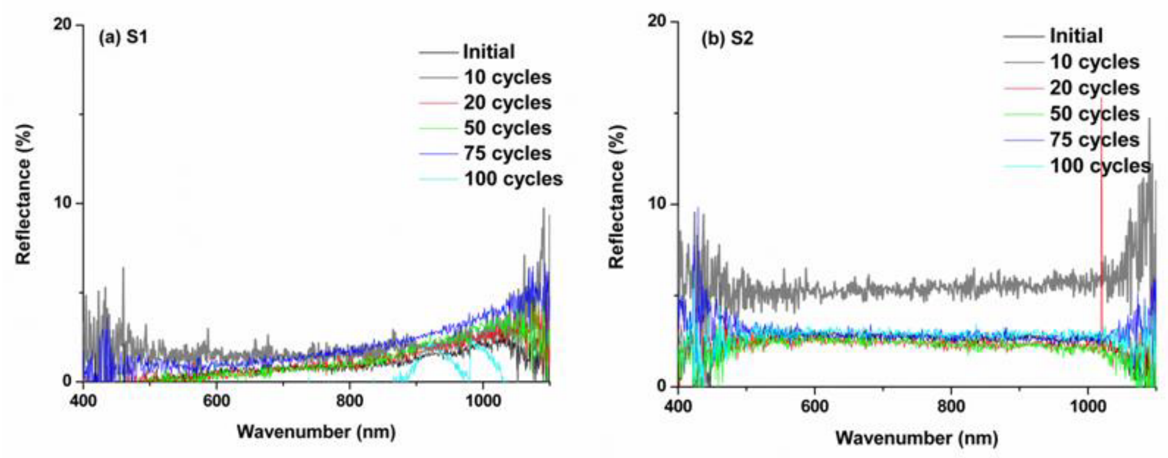

Figure 4: Reflectance results (a) Ref (b) S1 (c) S2

3.2. Thermo-shock tests using a Fresnel lens

The equipment available for this project is a Fresnel lens mounted on a solar tracker (Fig. 5). The lens is positioned on an aluminum frame with a polar axis orientation. The lens movement from east to west is controlled automatically by a computer and the solar height is hand positioned. The physical characteristic of the lens were previously reported (Padilla et al., 2014). The study considers the use of a wide range of ferrous and nickel-base refractory alloys, as well as some intermetallic alloys for high-temperature receiver applications. The materials may be classified into different groups as: a) refractory steel; b) iron aluminides, molybdenum silicide or boro-silicide intermetallics, and c) nickel-base superalloys.

Samples of 1 cm square section and 5 mm thick were subjected to thermal shock receiving concentrated solar radiation during heating/cooling cycles. Sample temperature while cycling was registered with a chromel/alumel thermocouple introduced in a side hole in the sample. Discovering the lens initiates the shock cycle by concentrating solar radiation fast heating up to 1200 ºC; after maintaining 10 min at this temperature, the shock cycle ends by fast cooling down to 200 ºC covering the lens to interrupt solar radiation on the sample. This temperature can be considered correct to within about ± 10 ºC, as evidenced by the temperatures reached during repeated thermal cycles for identical solar heating conditions. It is obvious that the Fresnel lens facility is capable of producing sufficiently high temperatures, as well as strong thermal shocks, that it can be used for the accelerated aging comparison of potential collector materials, as will be described in the following paragraphs.

Figure 5: Pictures of the Fresnel lens used for thermal shock tests installed on solar tracker at CSIC-CENIM

An example of the variation of temperature during the thermal shocks using the Fresnel lens is shown in Figure 6. As can be observed, both the heating rate and the cooling rate vary depending on the temperature range selected. So, from ambient temperature to 800 °C, a heating rate of 50-70 °C/s was registered. This is the maximum value of heating rate, because for higher temperatures, for 800 to 1000 ºC the heating rate decreases to values of 3-5 °C/s. Concerning the cooling, the highest rate occurred from high temperature to 600 ºC, with cooling rate of 9-11 °C/s. The cooling rate decreases for 600-400 °C to 2-4 °C/s and for the range 400-200 °C to 1 ºC/s. The cooling rate is much lower than the heating rate. Basically, the heating or cooling rates are governed by the heat loss to the ambient. In order to increase the thermal shock, the cooling rate could be increased by an external supply of cool air, i.e. active cooling.

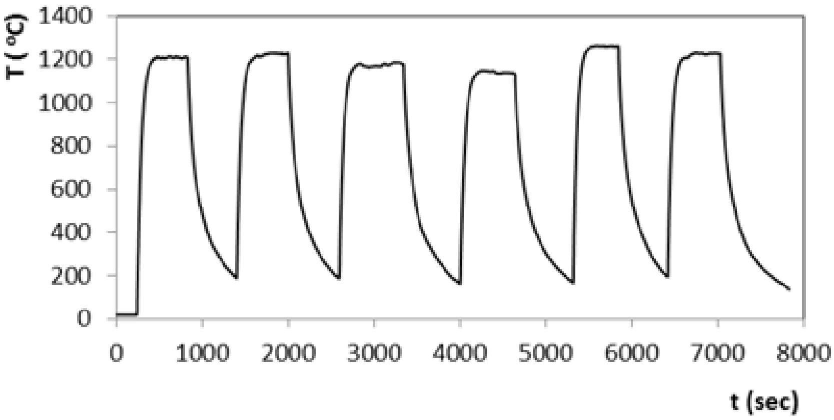

Figure 6: Example of a typical temperature (T) cycle achieved with the Fresnel lens.

Following 6 cycles of solar heating and cooling, the samples were removed for examination by optical and scanning electron microscopy. When the nominal temperature reached 1200 ºC or above, most of the samples showed a small crater-like region of sample melting. Temperatures at the front center clearly exceeded the solidus for these materials, of the order of 1300-1400 ºC for most of the materials. More relevant tests are carried out using a slightly lower solar radiation density, and are confirmed by furnace cyclic heating at temperatures of 100-1150 ºC. Under these conditions the materials showed oxidation to an extent that depended very strongly on the specific material. Oxidation of the refractory steel was small, as was that of the superalloy. The iron aluminides showed behavior that depended strongly on the Al content: for Al contents near 20% (with or without Cr) there was strong oxidation and loss of scale; for Al contents of 35-40% the oxidation was extremely fine, with no influence of the grain size. Both Mo intermetallics suffered extensive oxidation with spallation of the oxide.

From these preliminary studies, it is clear that all metallic alloys and intermetallics suffer from rapid oxidation and often oxide loss at temperatures in the range about 1000-1200 ºC, but nevertheless may be expected to withstand such cyclic oxidation with only minor damage at slightly lower temperatures. The possible interest of such alloys (compared with ceramics) for high temperature use lies in the favorable combination of physical and mechanical properties, especially good conductivity and moderate modulus combined with good toughness and (sometimes) good compatibility with the oxides formed. Further investigation is clearly necessary to evaluate such possibilities.

4. Evaluation of optical properties of relevant material samples

4.1. Influence of different surface textures on the optical properties of metallic surfaces

In order to evaluate the usefulness of surface textures for the application at solar receiver materials, the solar reflectance of different samples with various surface texture patterns has been analyzed. To do so, the hemispherical reflectance curve was measured with a spectrophotometer with integrating sphere (Gooch&Housego, 2011), from 300 nm to 2500 nm. Then, the solar reflectance value, ρ_S, was calculated integrating over the solar spectrum. This solar spectrum was based on the spectral energy distribution from the standard ASTM G173 (DNI + circumsolar). The solar absorptance of the material, which should be as high as possible for a good receiver, is calculated as given in Eq. (4), for an opaque material.

$$\alpha_S = 100 - \rho_S \text{ (in \%)} \quad (4)$$

The results of the reflectance measurements for four samples with two different materials (Inconel 718 and CrCo), and applying a surface finish (abrasive blasting) or not, are presented in Table 2.

Table 2: Reflectance and absorptance results for two different materials

Type	Surface finish	Reflectance ρ_S [%]	Absorptance α_S [%]
Inconel 718	with	53	47
Inconel 718	without	46	54
CrCo	with	60	40
CrCo	without	31	69

Considering these measurements, it can be said that CrCo without surface finish has the best optical properties regarding the application at solar receivers.

Furthermore, six samples with different textures (saw 30, 40, 60º and dents), with or without surface finish were also tested (see Figs. 7 and 8).

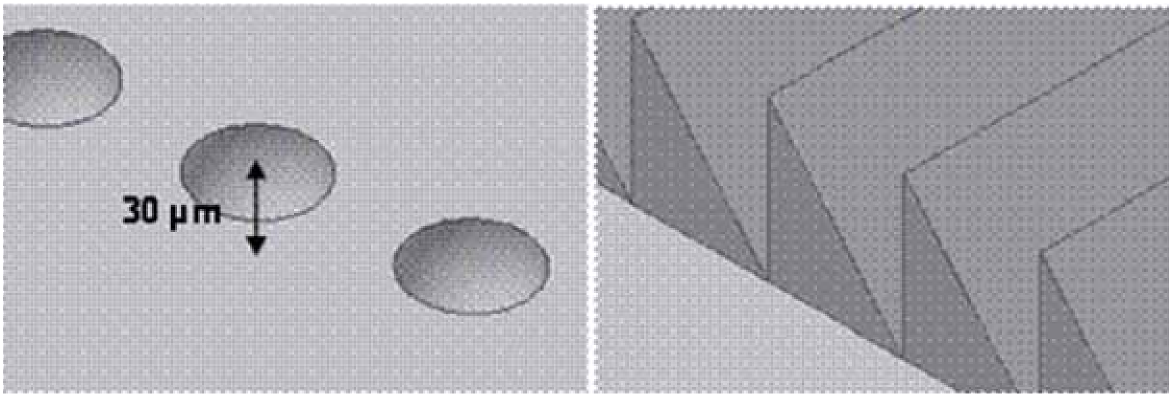

Figure 7: Sample texture concepts (left: dents, right: saw)

Figure 8: Saw and dent samples

The results of the reflectance measurements for those six samples indicate that the lowest reflectivity value can be obtained with a saw-type texture with 30º angle. This reflectance value achieved is about 27.6%.

4.2 Characterizing optical properties of ceramic materials

Finally, optical properties of SiOC ceramic materials, manufactured by CSIC-ICV, were measured and compared to the commercial painting Pyromark (Helling-GmbH, 2008; Ho et al., 2013) widely used on solar receivers in order to increase solar absorptance (see Fig. 9 for examined samples). The results of the reflectance measurements for those four samples are presented in Table 3.

Figure 9: Ceramic samples vs. Pyromark

Tab. 3: Reflectance and absortance results for 4 ceramic samples

Samples	Reflectance ρ_S [%]	Absorptance α_S [%]
P11-35-1 1008	6.36	93.64
P11-34-1 998	6.42	93.58
P11-33-1 1002	5.65	94.35
P11-32-1 1004	6.95	93.05

Pyromark	4.52	95.48

The material with the lowest reflectance would be the most recommended one for the use in a solar receiver. In this case, this would be: P11-33-1 1002 (reflectance 5.65%). However, the remaining samples are quite close and it can be said that all samples have an optical performance in the same order of magnitude as that of Pyromark paint (the chosen benchmark).

5. Conclusions

The present paper summarizes basic material research activities for high temperature solar receivers. After an introduction to promising receiver materials in Section 2, laboratory-scale high-temperature and thermo-shock tests have been performed indicating sample behavior.

Accelerated evaluation of potential materials for use as solar receivers is being carried out using a Fresnel lens system. The cyclic heating/cooling achieved can fracture oxide films forming on the material surface when heated and lead to much more accelerated damage than occurring during static heating as the same temperature. Such surface damage will cause a major reduction of the load-carrying capability of the component. Of the materials examined, a superalloy and a Ni-Cr-Fe refractory steel showed little surface oxidation damage. Iron aluminides show behaviour that depended on the Al content, with very extensive damage for alloys with low Al content, but damage much below that of the superalloy/refractory steel for alloys with high Al content. The Mo intermetallics suffered severe oxidation under the conditions used. Further work is required to refine these observations and improve the selection of suitable receiver materials

The laboratory high-temperature test was carried out over three different materials (Ref, S1 and S2). From this test, best results are obtained for the S2 sample (SiOC-dense material) which keep all its properties unaltered (appearance, weight, dimensions and optical properties).

Furthermore, the optical properties of two different materials (Inconel 718 and CrCo) have been compared. It can be concluded that CrCo without surface finish has the most promising optical properties achieving a solar absorptance of about 69%. In addition, the influence of different surface textures on the optical properties of metallic surfaces has been discussed. A saw-type texture with 30º angle has shown to provide the lowest reflectance value (27.6%) of the examined set of structures. Finally, selected ceramic materials have been evaluated regarding their optical properties with respect to the widely used Pyromark high temperature paint, showing that optical performance correlates quite well.

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