Optical thin film stacks integrating spectral and angular control of solar energy and thermal radiation

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STATEMENT OF ORIGINAL AUTHORSHIP

I, Maryna Bilokur, declare that this thesis is submitted in fulfilment of the requirements for the award of PhD degree, in the School of Mathematical and Physical Sciences, Faculty of Science, University of Technology Sydney. This thesis is wholly my own work unless otherwise referenced or acknowledged. In addition, I certify that all information sources and literature used are indicated in the thesis. This document has not been submitted for qualifications at any other academic institution. This research is supported by the Australian Government Research Training Program.

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THESIS FORMAT

The thesis has been prepared using conventional structure, comprised of a series of chapters. There are seven chapters with an abstract outlining the main results that have been achieved in the conducted research. Chapter 1 is dedicated to an introduction into the topic of the research (concentrated solar thermal applications), with a review of the principal physical phenomena behind photo-thermal conversion and spectrally-selective mechanisms to enhance solar-to-heat conversion. The chapter is concluded with the literature review, analysing the spectrally-selective surfaces developed to date and the key problems they face if applied in high temperature concentrated solar thermal applications.

Chapter 2 revises the methodology that has been used, explaining the principal work and mechanisms of device operation and physical processes behind it.

Chapter 3, Chapter 4 and Chapter 5 describe the main results that have been achieved during my PhD research. Chapter 3 describes new findings in the family of spectrally-selective surfaces based on noble metal cermets. Chapter 4 reflects on the semiconductor based approach of the solar absorber coatings using TiAlN family as the main solar absorbing component. Chapter 5 demonstrates a novel spectrally-selective solar absorber that is comprised of a multilayer stack with two $Ta:SiO_2$ cermets.

Chapter 6 analyses the deposition conditions of one of the main components of the spectrally-selective surface, an infrared back reflector leading to the highest possible ramp up in reflectance starting from 2500 nm.

Finally, Chapter 7 is devoted to a discussion and conclusion of the main findings with some overlook for the future work.

LIST OF PUBLICATIONS

***M. BILOKUR*; A. GENTLE; M. ARNOLD; M. B. CORTIE; G. B. SMITH (2017)**
High temperature optically stable spectrally-selective Ti1-xAlxN-based multilayer coating for concentrated solar thermal applications. *Solar energy materials and solar cells*. Volume 200, 15 September 2019, 109964. Available from:
https://www.sciencedirect.com/science/article/pii/S0927024819302934
[Accessed: 23 July 2019].

***M. BILOKUR*; A. GENTLE; M. ARNOLD; M. B. CORTIE; G. B. SMITH (2017)**
High Temperature Spectrally Selective Solar Absorbers Using Plasmonic $AuAl_2$:AlN Nanoparticle Composites. *Solar RRL*. 1(10). WILEY-VCH Verlag GmbH & Co. KGaA, Berlin, Germany. Available from:
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***M. BILOKUR*; A. GENTLE; M. ARNOLD; M. B. CORTIE; G. B. SMITH (2017)**
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https://www.spiedigitallibrary.org/conference-proceedings-of-spie/10356/103560C/An-advanced-plasmonic-cermet-solar-absorber-for-high-temperature-solar/10.1117/12.2273598.short [Accessed: 30 August 2017.].

***M. BILOKUR*; A. GENTLE; M. ARNOLD; M. B. CORTIE; G. B. SMITH (2015)**
Optical properties of refractory TiN, AlN and (Ti,Al)N coatings. In – "*SPIE 9668, Micro+Nano Materials, Devices, and Systems*", Sydney, Australia, December 7-9. University of Sydney. Available from:
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[Accessed: January 2016].

CONTENTS

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Abstract

The objective of this project is to enhance the efficiency of photo-thermal conversion by improving the optical and other properties of solar-absorbing surfaces. Designing a suitable coating for these surfaces involves a delicate balance between thermal stability, reflectance and emittance. As an added complication, it is necessary to have a coating with a spectral response that switches from highly absorptive in the visible and near-IR range to reflective at longer wavelengths. Despite extensive prior investigations in this area, there are still several problems that remained unsolved — in particular the maintenance of structural integrity and optical response of solar absorbers operating at higher temperatures (>500ºC). The results of the present work are highly relevant to various kinds of high temperature concentrated solar power (CSP) applications as well as to thermo-photovoltaic (TPV) systems.

A number of advanced new spectrally-selective solar absorbers: Al/AlN/Au-$AuAl_2$:AlN/AlN/SiO_2, Al/AlN/Au-$AuAl_2$:AlN/AlN/ Au-$AuAl_2$:AlN/AlN/SiO_2, Pt/AlN/TiAlN/ AlN/SiO_2, Pt/Ta:SiO_2/Ta:SiO_2/AlN/SiO_2 and Ta/Ta:SiO_2/ Ta:SiO_2/AlN/SiO_2 were investigated. All were produced by magnetron sputtering, and their optical properties and thermal stability assessed.

This work has shown that the Au-based solar absorbing structures are strongly oxidation-resistant, however, their exploitation in CSP applications is currently limited due to coarsening and agglomeration of the Au inclusions in the dielectric host temperatures greater than 400ºC. A solution to this problem is proposed : the Au nanoparticles in the cermet layer are allowed to alloy with Al. This converts them to the intermetallic compound $AuAl_2$, which is considerably more resistant to coarsening than pure gold. This was achieved by an introduction of the Al substrate to serve both as an IR-reflecting layer and as a source of the Al species to form more structurally and temperature stable $AuAl_2$ nanoparticles in the AlN host. The alloying process was thermally induced at 200ºC and was finalised at 500ºC, where alloying of all Au inclusions present in the matrix was achieved. The resultant new structure was able to endure 168 hours annealing in vacuum at 500ºC without major change. Such stability has apparently not been achieved before for Au-based solar absorbers. Furthermore, the $AuAl_2$ formation was shown to be also beneficial for the solar absorptance (α_s) enhancement, leading to an increase in α_s by 3%, from its initial 92% to a final 95%, while preserving low emittance.

Spectrally-selective coatings based on the$Ti_xAl_{1-x}N$ system were also considered due to their known diffusion barrier properties, high thermal tolerance, and very suitable optical properties. The composition of $Ti_xAl_{1-x}N$, (effectively, the Ti/Al ratio) was selected to achieve a maximized solar absorptance of the overall stack. A tandem absorber, which included top anti-reflective layers, was tested on a stainless-steel substrate in order to see how the stack design would serve in parabolic trough-based power plants that used stainless steel pipe to carry the heat-transfer fluid. The diffusion of the Fe present in stainless steel into the coating is known to normally start at 600ºC but this was successfully suppressed in the present work by an application of an AlN diffusion barrier. The whole $Ti_xAl_{1-x}N$-based stack, despite some structural modifications upon heating up to 900ºC, preserved its optical integrity with solar absorptance remaining unchanged at 92%.

Finally, a new algorithm for designing a nearly ideal cermet-based spectrally-selective absorber was developed. This enabled achievement of α_s> 97%. There are only a few structures known to absorb solar energy with α_s in the 97-98% range, however, their optical performance is degraded in the range 250ºC-500ºC due to surface oxidation, diffusion of the back reflector into the coating, shape and/or phase transformation of the nanoparticles. The result may be a significant drop in solar absorptance down to 84%. The new algorithm was exploited in the present project to design a novel spectrally-selective coating, the heart of which was composed of two absorbing $Ta:SiO_2$ layers with different Ta content, which showed not only an efficient light absorptance with α_s = 97.6%, but also preservation of its value up to 900ºC with simultaneously improved spectrally-selective performance due to recrystallization of the Pt or Ta back-reflectors. These effects lowered thermal emittance to 0.04 and 0.15 from initial values of 0.18 and 0.21, respectively. The $Ta:SiO_2$ cermet-based absorber on a Pt reflector showed good thermal stability up to 1000ºC, with a minor solar absorptance reduction to 95%, but high enough for an enhanced photo-thermal conversion. This would appear to be an unprecedented degree of stability at 1000°Ç for a cermet-based solar absorber.

In summary, this project has resulted in the development of new stack designs for use in high temperature conversion devices. A new way of enhancing thermal stability in a Au-based coating has been discovered, and a new procedure for designing coatings demonstrated.