

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

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5 August 2019

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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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Date: August 5, 2019

ACKNOWLEDGEMENTS

I would like to express my special appreciation and thanks to my supervisors Prof. Michael Cortie, Dr Matthew Arnold, Dr Angus Gentle and Prof. Geoff Smith for providing me a unique opportunity to become a part of their research team and their ongoing support during these years. I deeply appreciate the experience I have received working closely with them and the professional knowledge and expertise they have shared with me, which helped me to grow a lot as a research scientist! A special thanks to Angus Gentle and Geoff McCredie for the thorough technical training and assistance they have provided, making the lab environment a very professional and friendly place. A very big thank to Geoff Smith for his stimulating discussions and a great deal of encouragement in the process of growing my scientific expertise. He shared his wealth of experience with me during our discussions and I have learnt a lot from him! A very big thank to Matthew Arnold for his encouragement, constructive criticism and guidance through my PhD journey. And, finally, I am very grateful to my principal supervisor, Michael Cortie, for his constant support and sympathy, being a highly professional academic advisor, who helped me all the time during my research and thesis writing. Also, I would like to thank Katie McBeam and Mark Berkahn for their assistance with the SEM and XRD training.

A special and sincere gratitude to Dr Marie Manidis for her tremendous moral support and becoming a good friend of mine, who lit up the moments of my PhD life with her generosity, sincerity, sharp intelligence and just a wonderful personality.

A special thanks to my family, sister, mom and dad, my godfather and my brother-in-law for being with me all these time despite separation of thousands of kilometers and being an infinite source of love, inspiration, support and strength and encouraging my scientific journey.

Also, I thank my fellow labmates and colleagues for becoming the part of my PhD journey and being very supportive during these times, sharing their experience and creating a family like environment at the work-place we shared altogether: Marc Gali, Aaron Colusso, Matthew Tai, Sujeewa de Silva, Chris Wolf, Igor Aharonovich, Daniel Pasin, Alba Santin Garcia, Mahmoud El Safadi, Smith Panicer, Minh Nguyen, Blake Regan, Daniel Totonjian, Matthew Cappaddona, Behnaam Mlk, John Scott, Chris Elbadawi, Mehran Kianinia, Noah Mendelson, Jacqueline Loyola-Echeverria, Mark Barash, Anirudha Saha, Olivier Lee, James Bishop, Trong Toan Tran.

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:SiO₂ 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 Ti_{1-x}Al_xN-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)

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<http://onlinelibrary.wiley.com/doi/10.1002/solr.201700092/full>

[Accessed: 7 September 2017].

M. BILOKUR; A. GENTLE; M. ARNOLD; M. B. CORTIE; G. B. SMITH (2017)

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M. BILOKUR; A. GENTLE; M. ARNOLD; M. B. CORTIE; G. B. SMITH (2015)

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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 ($>500^{\circ}\text{C}$). 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-AuAl₂:AlN/AlN/SiO₂, Al/AlN/Au-AuAl₂:AlN/AlN/ Au-AuAl₂:AlN/AlN/SiO₂, Pt/AlN/TiAlN/ AlN/SiO₂, Pt-Ta:SiO₂/Ta:SiO₂/AlN/SiO₂ and Ta-Ta:SiO₂/Ta:SiO₂/AlN/SiO₂ 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 400°C . 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 AuAl₂, 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 AuAl₂ nanoparticles in the AlN host. The alloying process was thermally induced at 200°C and was finalised at 500°C , 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 500°C without major change. Such stability has apparently not been achieved before for Au-based solar absorbers. Furthermore, the AuAl₂ 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 $\text{Ti}_x\text{Al}_{1-x}\text{N}$ system were also considered due to their known diffusion barrier properties, high thermal tolerance, and very suitable optical properties. The composition of $\text{Ti}_x\text{Al}_{1-x}\text{N}$, (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 600°C but this was successfully suppressed in the present work by an application of an AlN diffusion barrier. The whole $\text{Ti}_x\text{Al}_{1-x}\text{N}$ -based stack, despite some structural modifications upon heating up to 900°C, 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 $\alpha_s > 97\%$. 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 250°C-500°C 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:SiO₂ layers with different Ta content, which showed not only an efficient light absorptance with $\alpha_s = 97.6\%$, but also preservation of its value up to 900°C 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:SiO₂ cermet-based absorber on a Pt reflector showed good thermal stability up to 1000°C, 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°C 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.