Numerical and Experimental Study of Sputtered Mo-Al_2O_3 Nano Composites for Photothermal Energy Harvesting

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***Abstract*— We report on the numerical and experimental study of the optical spectra of Mo-Al_2O_3 nanocomposites that were grown using sequential DC and RF sputtering. The measured spectra were compared with the ones calculated using FDTD simulations to analyze the prediction accuracy of the approach.**

Keywords—solar selective absorbers, ceramic-metal nano composites, cermet, molybdenum nanoparticles, FDTD.

I. Introduction

Ceramic-metal nano composites (NCs) are fundamental materials for solar selective absorbers (SSA) with high absorptance of solar radiation and low infrared emittance which are critical for photothermal energy harvesting [1]. Accurate prediction of the optical characteristics of an NC with multilayer coatings is essential to design efficient SSAs [2]. Recently, utilization of FDTD technique has been reported for simulation of the optical properties of NCs with noble metal (Ag and Au) nanoparticles and SiO_2 as dielectric host [3], [4]. More recently, importance of particle-particle interactions as well as the effects of the spatial distribution of the particles in determination of the reflectance (R) and the transmittance (T) of nano composites have been clearly demonstrated by FDTD calculations [3], [4]. It was also demonstrated that the spatial disorder of NCs nanoparticles (randomness) reduces the quality of resonant behavior in R and T spectra of the amorphous metamaterials similar to NCs [5]. To the best of our knowledge, FDTD simulations of Mo-Al_2O_3 nano composites have not been reported in the literature.

This work aims to study the optical properties of Mo-Al_2O_3 NCs through FDTD simulations considering the effects of sputtering on the particle distribution and material characteristics and compare with the experimental data.

II. Experiemntal Details

A commercially available FDTD solver (Lumerical 2020) [6] was used for numerical modeling of the cermet structures. In the simulation setup, we have used periodic boundary conditions for the x and y-directions, while the PML boundary condition (64 PML layers) was used for z-direction. A broadband plane wave (200nm-1700nm) was injected in the z-direction for all simulation cases. To definite the geometrical setting and precise calculations, spatial mesh grid size was set to $\Delta x = \Delta y = \Delta z = 0.25$nm. Two monitors (transmission and reflection monitors) were placed above and below the sample to record the spectral data. For the simulation setup, we have designed the cermet structure by embedding Mo nanoparticles in alumina (Al_2O_3) and placing on a glass substrate. We have used two different sets of Mo optical properties in the FDTD simulations. The first set was the optical parameters for pure bulk Mo provided in the literature whereas the second set was calculated from the measured optical spectra of the sputtered Mo thin films in this study. The numerical analysis was done using custom codes by exporting the data from the corresponding monitors.

For the experimental verification study, two magnetron sputtering guns (Gencoa SW50200), each of which was able to operate in DC or RF mode were used to deposit the Mo-Al_2O_3 nano composites. The power of each magnetron gun was adjusted to have the desired growth rate and metal volume fraction (30%). The Mo and Al_2O_3 ceramic targets, had the purity of 99.95% and 99.99 %, respectively. The specular borosilicate glass substrates were mounted on a cylindrical holder that rotated with an adjustable circular rotation speed. Each pass of the substrate, conduced to deposition of material with no more than 0.2 nm thickness, which is smaller than the interatomic distance between Mo atoms (of about 0.2 nm). This condition guaranteed that Mo nucleated as isolated nanoparticles. We measured the NCs thickness as 50 ± 5 nm using an Alpha step 200 Tencor Instruments profiler. The cermets reflectance and transmittance spectra were measured using a QEX10 Solar Cell Quantum Efficiency Measurement system with high repeatability. The material analysis was performed using a Phillips CM-200 200 kV Transmission Electron Microscope (TEM).

III. Results and Discussions

For the FDTD simulations we have considered two particle distribution schemes with varying metal volume

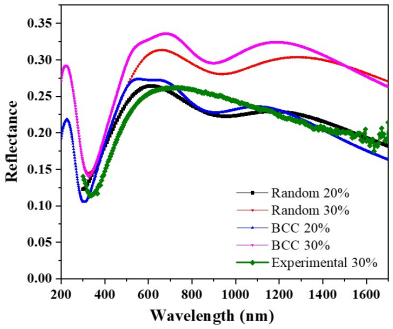

Fig. 1. Comparison of FDTD (*bcc* and random particle distribution) and experimental reflectance. FDTD used bulk Mo optical parameters and metal volume fraction f=20% and 30%.

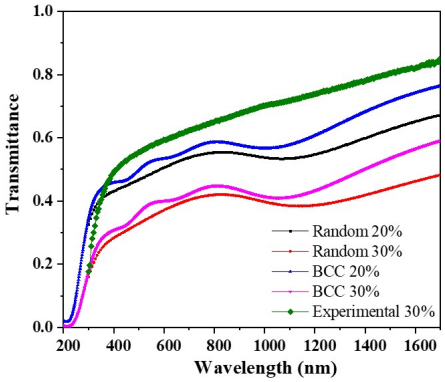

Fig. 2. Comparison of FDTD (*bcc* and random particle distribution) and experimental transmittance. FDTD used bulk Mo optical parameters and metal volume fraction *f*=20% and 30%.

fractions (f): i) body centric cubic (*bcc*) lattice arrangement and ii) random distribution of the nanoparticles. In all cases, we assumed a monodisperse distribution with the diameter of 2 nm based on the TEM analysis results. This has been proved to be a good approximation when particle diameter is considerably smaller than the wavelength of the incident radiation [3]. In the case of *bcc* lattice, we changed the value of f by changing the distance between particles around 3 nm. Fig. 1 shows the comparison between the experimental reflectance and FDTD reflectance calculated using the optical parameters for bulk Mo metal volume fraction *f*=20% and 30%. We observe that the experimental reflectance is better fitted for f = 20%, for both random and *bcc* cases. The random case is very similar to the *bcc* case, but with a less prominent broad dip. The reflectance minimum is much better described for the *bcc* case. For the transmittance shown in Fig. 2, we also have the best fit for f =20%. We note that the behavior of transmittance at low wavelengths is well described for FDTD simulations using bulk Mo optical parameters. Fig. 3 and 4, present the effect of the optical parameters extracted for sputtered Mo on the FDTD calculated reflectance and transmittance. In the case of reflectance, the agreement between the curves is remarkable for f = 20%, except that the reflectance minimum in the measured spectrum is not well reproduced as the calculated one. This discrepancy may be stemmed from the fact that the disorder considered in the particle positions or the optical properties of Mo nanoparticles are not accurately defined in the simulations. In

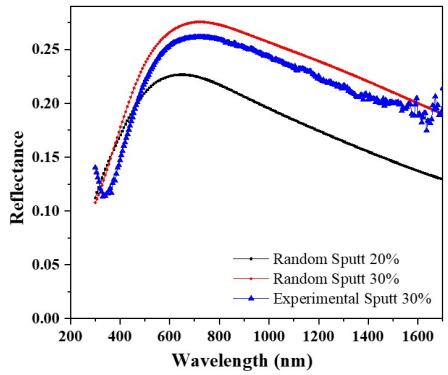

Fig. 3. Comparison of FDTD (random particle distribution) and experimental reflectance. FDTD used optical parameters of sputter Mo and metal volume fraction *f*=20% and 30%.

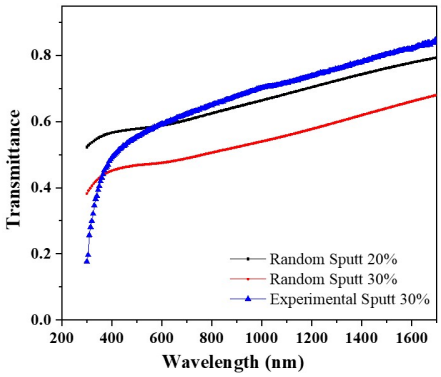

Fig. 4. Comparison of FDTD (random particle distribution) and experimental transmittance. FDTD used optical parameters of sputter Mo and metal volume fraction *f*=20% and 30%.

the case of transmittance, we observe a good fit above 500 nm. The discrepancy below 500 nm can be related to the same effect in the case of reflectance minimum or the inaccuracy of the optical parameters used for Mo, which were calculated using the experimental spectral of a sputtered Mo thin film. The optical properties of Mo nanoparticles in our nanocomposite non-necessarily present the same optical properties as a sputtered Mo film.

IV. Conclusion

We have shown that FDTD simulations can accurately predict the reflectance and transmittance of sputtered Mo-Al_2O_3 nano composites even with the strict monodisperse assumption. The results show that use of experimentally obtained optical parameters for Mo significantly improves the accuracy of the simulations. We have also observed that use of random particle distribution considerably improves the accuracy of the calculated R and T spectra, but some characteristics as such as reflectance minimum are better reproduced using *bcc* particle arrangement. The presented results can help designing more efficient solar selective absorbers.

Acknowledgement

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References

[1] J. Y. Lu, A. Raza, N. X. Fang, G. Chen, and T. Zhang, "Effective dielectric constants and spectral density analysis of plasmonic nanocomposites," *J. Appl. Phys.*, vol. 120, no. 16, p. 163103, Oct. 2016.

[2] M. R. Nejati, V. Fathollahi, and M. K. Asadi, "Computer simulation of the optical properties of high-temperature cermet solar selective coatings," in *Solar Energy*, 2005, vol. 78, no. 2, pp. 235–241.

[3] C. Etrich, S. Fahr, M. Keshavarz Hedayati, F. Faupel, M. Elbahri, and C. Rockstuhl, "Effective Optical Properties of Plasmonic Nanocomposites," *Materials (Basel).*, vol. 7, pp. 727–741, 2014.

[4] M. D. Thoreson, "Fabrication and realistic modeling of three-dimensional metal-dielectric composites," *J. Nanophotonics*, vol. 5, no. 1, p. 051513, Jan. 2011.

[5] C. Helgert *et al.*, "Effective properties of amorphous metamaterials," *Phys. Rev. B*, vol. 79, no. 23, p. 233107, Jun. 2009.

[6] "Nanophotonic FDTD Simulation Software - Lumerical FDTD." [Online]. Available: https://www.lumerical.com/products/fdtd/. [Accessed: 08-Apr-2020].