Numerical analysis of mechanical reliability of multi-coated phase change materials

Forner-Escrig, Josep; Navarrete, Nuria; Palma, Roberto; La Zara, Damiano; Goulas, Aristeidis; Valdesueiro, David; van Ommen, J. Ruud; Hernández, Leonor; Mondragón, Rosa

DOI
10.1051/e3sconf/202132102019

Publication date
2021

Document Version
Final published version

Published in
E3S Web of Conferences

Numerical analysis of mechanical reliability of multi-coated phase change materials

Josep Forner-Escrig[1], *Nuria* Navarrete[1], *Roberto* Palma[2], *Damiano* La Zara[3], *Aristeidis* Goulas[3], *David* Valdesueiro[4], *J. Ruud* van Ommen[3], *Leonor* Hernández[1] and *Rosa* Mondragón[1*]

[1]Department of Mechanical Engineering and Construction, Universitat Jaume I, Av. de Vicent Sos Baynat, s/n 12071 Castelló de la Plana, Spain
[2]Department of Structural Mechanics and Hydraulic Engineering, University of Granada, Campus Universitario Fuentenueva, Edf. Politécnico, 18071 Granada, Spain
[3]Department of Chemical Engineering, Delft University of Technology, 2629 HZ, Delft, the Netherlands
[4]Delft IMP B.V, Molengraaffsingel 10, 2629 JD, Delft, the Netherlands

Abstract. Nanoencapsulated phase change materials (nePCMs) are nowadays under research for thermal energy storage purposes. NePCMs are composed of a phase change core surrounded by a shell that confines the core when molten. One of the main concerns of nePCMs when subjected to thermal processes is the mechanical failure of the passivation shell initially present in commercial metallic nanoparticles. In order to overcome this issue, multi-coated nePCMs, based on the synthesis of an additional coating by atomic layer deposition, appear to be as a candidate solution. With the objective of studying the influence of the composition and thickness of the additional nePCM shells on their probability of failure, a numerical tool combining a thermomechanical finite element model with phase change and Monte Carlo algorithms is developed. This tool also allows including the uncertainty of material and geometrical properties into the numerical analysis to account for their influence in the mechanical performance of nePCMs. In the present work, the mechanical reliability of SiO_2 and Al_2O_3 coatings on $Sn@SnO_x$ nanoparticles is assessed by considering both deterministic and probabilistic failure criteria and Al_2O_3 coatings appear to have a better mechanical performance than their SiO_2 counterparts.

* Corresponding author: mondragn@uji.es

1 Nomenclature

Quantity	Symbol	SI Unit
Acceleration field	$\underline{\ddot{u}}$	m/s^2
Cauchy stress tensor	$\underline{\underline{\sigma}}$	N/m^2
Body force vector	$\underline{f}$	N
Heat flux vector	$\underline{q}$	W/m^2

2 Introduction

Energy generation and conversion is one of the major concerns that society is facing nowadays. This tendency falls within the global context of transition from conventional energy sources towards renewable ones. Among the wide variety of available renewable sources, the focus is put on solar energy due to the enormous amount of energy that the Sun constantly delivers to Earth.

However, the main issue with solar energy is their dependence upon weather conditions for energy generation. On this ground, thermal energy storage (TES) systems are under research in order to decrease the eventual gaps between energy supply and demand, which may lead to instabilities in the operation of the electric grid.

More precisely, in the field of solar thermal energy, nanofluids based on molten salts are being studied as a candidate technology for TES purposes. Nanofluids are colloidal suspensions of nano-sized particles in a working fluid [1]. The potential of nanofluids to store energy normally was associated to their sensible storage capability but recently nanoencapsulated phase change materials (nePCMs) were used instead as the solid component of nanofluids. The use of nePCMs enhanced the thermal storage capability of the suspension by means of the contribution of latent heat [2,3].

NePCMs are normally composed of a phase change core wrapped in a shell made of another material with a higher melting temperature than the core. With regard to metallic nanoparticles, which are the object of study of the present work, their shell is normally made of an oxide layer formed by passivation [4,5].

An issue encountered when subjecting nanofluids with nePCMs dispersed in it to thermal cycles is the mechanical failure of their shell, which results in the leak of the molten core of nePCMs and the loss of thermal storage capability of the nanofluid. With regard to the mechanical failure of nePCM shells, the main difficulty in their synthesis is to determine which material and/or geometrical parameters exert the most significant influence on the mechanical failure of shells. In some previous works [6], it was found that the shell thickness as well as other material parameters such as the melting temperature or the thermal expansion coefficient of the core were some of the variables having more influence on the mechanical failure of nePCM shells. Since the initial oxide layer thickness formed by passivation is hard to control, the technique of atomic layer deposition (ALD) [7] appears to be as a possible solution to synthesise an additional coating of controlled properties and thickness on nePCMs. These nanoparticles with two different encapsulations often receive the name of multi-coated nePCMs and Figure 1 shows an image of these experimentally synthesised multi-coated nePCMs.

With regard to the synthesis of the shell thickness surrounding the core, a compromise must be reached between their mechanical strength and energy storage capability. Consequently, in order to systematically assess the mechanical performance of different nePCMs, to incorporate the measurement dispersion of material and geometrical parameters into the numerical analysis and to gain deeper insight into the causes of the failure of nePCMs, a numerical tool combining a thermomechanical finite element model with phase change with Monte Carlo (MC) simulations is presented in this work. More concretely, the performance of the following nePCMs (core@inner_shell@outer_shell) experimentally synthesised by ALD is assessed: Sn@SnO, $Sn@SnO_2@SiO_2$ and $Sn@SnO@Al_2O_3$.

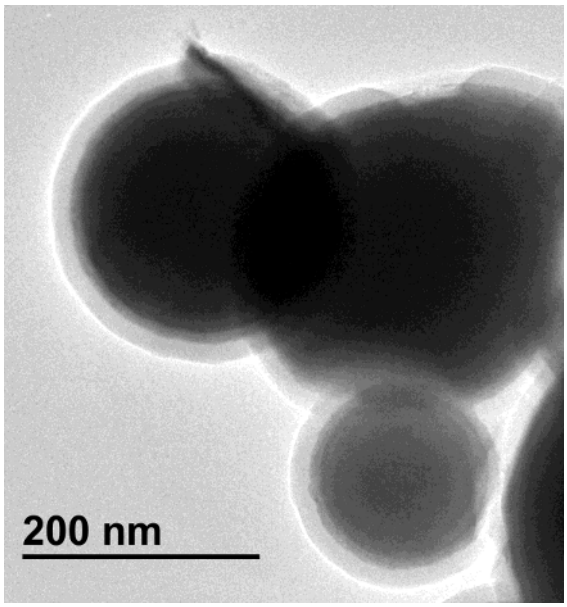


Fig. 1. Image of a multicoated $Sn@SnO@Al_2O_3$ nePCM synthesised by the technique of ALD.

3 Analysis and modelling

The numerical tool presented in this work combines a thermomechanical FE model with phase change with MC techniques.

The thermomechanical FE model was developed in a previous work [8] to study the thermal stresses

arising in nePCMs when they are subjected to thermal processes. The governing equations of the thermomechanical FE model with phase change are the linear momentum and energy balances, respectively:

$$\rho\ddot{\underline{u}} = \underline{\nabla}\cdot\underline{\underline{\sigma}} + \underline{f} \qquad (1)$$

$$\frac{dH}{dt} = -\underline{\nabla}\cdot\underline{q} + \dot{S} \qquad (2)$$

Notice that the angular momentum balance is automatically satisfied by the symmetry of the Cauchy stress tensor.

The regularisation scheme called as enthalpy method presented in that same previous work [8] is used to run the numerical simulations performed in the present study. Regarding MC techniques, they are a class of numerical algorithms that make use of statistical samples to approach the solution of a model in a probabilistic sense. The general procedure to perform a MC analysis consists in:

i) Defining the random parameters of the model and their distribution functions.
ii) Generating a sample of size N.
iii) Performing N evaluations of the model.

Then, outputs are obtained by post-processing the results obtained from simulations. Further detail concerning MC techniques used in the present work can be found in the work of Forner-Escrig [6].

The performance parameter considered for the analysis of mechanical reliability of the nePCM shells is their Probability of Failure (POF), which can be evaluated by means of MC techniques as:

$$POF = \frac{n\left[G(\hat{\xi} \le 0)\right]}{N} \qquad (3)$$

where $n\left[G(\hat{\xi} \le 0)\right]$ stands for the number of cases n for which a considered limit state function (the mechanical strength of the nePCM shell in this case) is not satisfied and N is the number of MC iterations.

The model used for the numerical predictions is composed of a single three-dimensional ellipsoidal nePCM and two different pairs of multi-coated nePCMs (core@inner_shell@outer_shell) are considered: Sn@SnO, Sn@SnO_2@SiO_2 and Sn@SnO@Al_2O_3, which were experimentally synthesised by ALD to precisely control the shell thickness of the nePCMs. For the use of MC techniques, the input parameters of the model (material and geometrical properties) are considered as random variables and their nominal values and standard deviations retained for the numerical simulation are presented in Tables 1 and 2.

Table 1. Summary of material and geometrical properties for Sn@SnO_2@SiO_2 nePCMs.

	Property	Values	σ(%)	Units
Core (Sn)	ρ_s	7280	5	kg/m^3
	ρ_l	6800	5	kg/m^3
	c_s	230	5	J/(kg·K)
	c_l	257	5	J/(kg·K)
	κ_s	65	5	W/(m·K)
	κ_l	31	5	W/(m·K)
	E	43.3	5	GPa
	υ	0.33	5	-
	α	$2\cdot10^{-5}$	5	1/K
	T_m	498.65	5	K
	L	60.627	5	kJ/kg
Inner shell (SnO)	ρ	7020	10	kg/m^3
	c	348.95	10	J/(kg·K)
	κ	40	10	W/(m·K)
	E	222.72	10	GPa
	υ	0.284	10	-
	α	$4\cdot10^{-6}$	10	1/K
	T_m	1900	10	K
Outer shell (Al_2O_3)	ρ	3970	5	kg/m^3
	c	919.38	5	J/(kg·K)
	κ	10	5	W/(m·K)
	E	370	5	GPa
	υ	0.24	5	-
	α	$8.2\cdot10^{-6}$	5	1/K
	T_m	2273.15	5	K
Geometry	a_{semi}	90	1	nm
	b_{semi}	90	1	nm
	c_{semi}	90	1	nm
	$e_{shell,i}$	12	1	nm
	$e_{shell.o}$	(1, 5, 10)	1	nm

Table 2. Summary of material and geometrical properties for Sn@SnO and Sn@SnO@Al_2O_3 nePCMs.

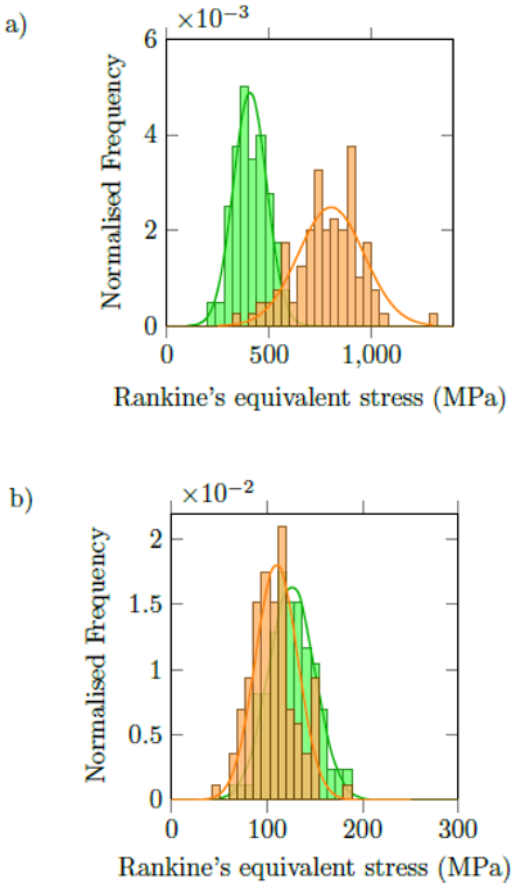
	Property	Values	σ(%)	Units
Core (Sn)	ρ_s	7280	5	kg/m^3
	ρ_l	6800	5	kg/m^3
	c_s	230	5	J/(kg·K)
	c_l	257	5	J/(kg·K)
	κ_s	65	5	W/(m·K)
	κ_l	31	5	W/(m·K)
	E	43.3	5	GPa
	υ	0.33	5	-
	α	$2\cdot10^{-5}$	5	1/K
	T_m	498.65	5	K
	L	60.627	5	kJ/kg
Inner shell (SnO)	ρ	7020	10	kg/m^3
	c	348.95	10	J/(kg·K)
	κ	40	10	W/(m·K)
	E	222.72	10	GPa
	υ	0.284	10	-
	α	$4\cdot10^{-6}$	10	1/K
	T_m	1900	10	K
Outer shell (Al_2O_3)	ρ	3970	5	kg/m^3
	c	919.38	5	J/(kg·K)
	κ	10	5	W/(m·K)
	E	370	5	GPa
	υ	0.24	5	-

	α	$8.2 \cdot 10^{-6}$	5	1/K
	T_m	2273.15	5	K
Geometry	a_{semi}	90	1	nm
	b_{semi}	90	1	nm
	c_{semi}	90	1	nm
	$e_{shell,i}$	12	1	nm
	$e_{shell.o}$	(1, 5, 10)	1	nm

With respect to boundary and initial conditions, the centre of the nePCM is mechanically fixed and a temperature value is prescribed on the external surface of the outer shell. Temperature is increased progressively with time varying from an initial value of 343 K until a value of 553.15 K, which is higher than the core melting temperature (498.65 K) of the aforementioned multi-coated nePCMs.

4 Results and discussion

With regard to the performance parameter, namely the POF of the nePCM shell, Figure 2 shows the stress distributions predicted by the numerical tool against a probabilistic failure criterion to also consider the influence of the tensile strength dispersion on the mechanical failure of the nePCM shells. POF is obtained by comparing the overlapping between stress distributions and failure criteria. For instance, for $Sn@SnO_2@SiO_2$ nePCMs, the probabilistic failure criterion is obtained by comparing the intersection between the stress distribution and a distribution of tensile strength (in orange in Figure 2) which determines the POF of nePCM shells.



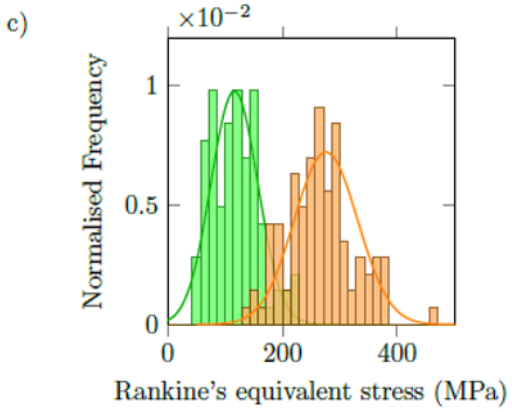


Fig. 2. Probabilistic failure criteria against stress distributions for nePCMs: a) Sn@SnO, b) $Sn@SnO_2@SiO_2$ and c) $Sn@SnO@Al_2O_3$

The values of POF from Figure 2 for Sn@SnO nePCMs is 2.27%, for $Sn@SnO_2@SiO_2$ nePCMs is 72.38% and the values for $Sn@SnO@Al_2O_3$ nePCMs is 1.66%, according to the probabilistic failure criterion. Numerical studies performed by also considering deterministic failure criteria show the need to consider probabilistic ones because by using a deterministic critetion slightly underestimates the POF of nePCM shells, which may result in a remarkable loss of energy storage capability of nanofluids.

The influence of the shell thickness of the outer shell is examined as well, as depicted in Figure 3. This Figure shows the evolution of the mechanical POF of the shells with increasing shell thickness. It can be observed that the POF of the inner shells (SnO and SnO_2) remains nearly constant regardless of the value of the outer shell thickness. However, remarkable variations of POF of the outer shells occur when varying their shell thickness. For instance, POF of SiO_2 reduces from an initial value of 87.56% until a value of 72.38%. The same trend is observed for Al_2O_3, which exhibits a drastic reduction in POF from a value of 31.71% until a value of 1.66%.

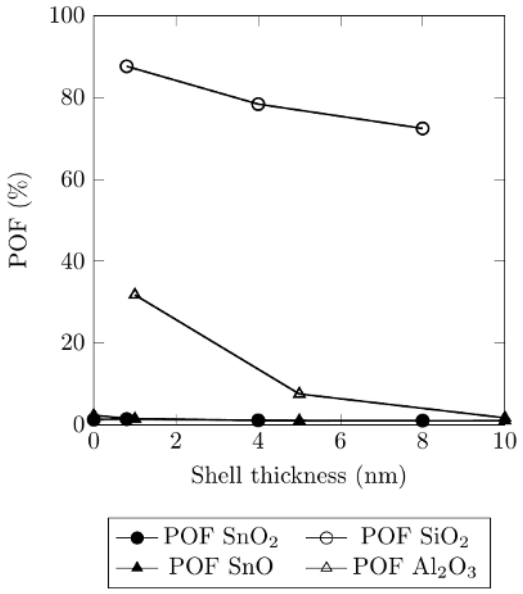


Fig. 3. Evolution of POF of shells with shell thickness of the outer coating

From these results it is noticed that Al_2O_3 coatings present better mechanical strength than their SiO_2 counterparts. Furthermore, it is observed that encapsulation of nePCMs can contribute to improve their mechanical reliability but the shell thickness cannot be continuously increased because this could jeopardise the colloidal stability of the nanofluid since nanoparticles with bigger size could agglomerate and settle.

5 Conclusions

This work presents a numerical tool combining a thermomechanical FE model with MC techniques, which is used to assess the mechanical reliability of different nePCMs.

From the numerical simulations, the importance of considering a probabilistic failure criterion is stressed in order not to overestimate the mechanical strength of nePCM shells. Furthermore, $Sn@SnO@Al_2O_3$ nePCMs exhibit the best mechanical performance since they are not expected to fail under thermal stresses while the POF of $Sn@SnO_2@SiO_2$ is still high even for the largest value of shell thickness considered in this study.

In short, tuning the shell thickness of nePCMs can contribute to enhance their mechanical performance although this thickness cannot always be increased in order not to guarantee the colloidal stability of the nanofluids. Therefore, in order to carefully study the mechanical reliability of nePCMs, the numerical tool presented in this work can be employed to run numerical simulations with the aim of reducing the number of experiments to be conducted to determine the optimal properties of nePCMs.

Acknowledgements
This research was partially funded by Ministerio de Economía y Competitividad (MINECO) of Spain through the project ENE2016-77694-R, by Generalitat Valenciana through the project PROMETEU/2020/029 and by the Universitat Jaume I through the project UJI-B2020-32. Josep Forner-Escrig thanks Ministerio de Economía, Industria y Competitividad of Spain and Fondo Social Europeo for a pre-doctoral fellowship through Grant Ref. BES-2017-080217 (FPI program).

References

1. S. Choi and J. Eastman, *Enhancing thermal conductivity of fluids with nanoparticles*, in Proceedings of the ASME International Mechanical Engineering Congress and Exposition, 12-17, November 1995, San Francisco, CA, USA, (1995).
2. S. Cingarapu, D. Singh, E. V.Timofeeva, and M. R. Moravek, Int. J. Energy Res., **38**, 51-59 (2013).
3. S. Cingarapu, D. Singh, E. V.Timofeeva, and M. R. Moravek, Renew. Energy, **80**, 508-516, (2015).
4. N. Navarrete, A. Gimeno-Furió, R. Mondragón, L. Hernández, L. Cabedo, E. Cordoncillo, and J. E. Juliá, *Sci. Rep.*, **7**, 17580 (2017).
5. N. Navarrete, R. Mondragón, D. Wen, M. E. Navarro, Y. Ding, and J. E. Juliá, Energy, **167**, 912 -920, (2019).
6. J. Forner-Escrig, R. Mondragón, and R. Palma, *Mechanical Reliability of Core-Shell Nanoparticles for thermal energy storage by Finite Element Method*, in Conference Proceedings of 1st Internationational Conference on Nanofluids (ICNf2019), 2nd European Symposium on Nanofluids (ESNf2019), 26-28 June 2019, Castelló de la Plana, Spain, (2019).
7. 7. N. Navarrete, D. La Zara, A. Goulas, D. Valdesueiro, L. Hernández, J. R. van Ommen, and R. Mondragón, Sol. Energy Mater. Sol. Cells, **206**, 110322 (2020).
8. J. Forner-Escrig, R. Palma, and R. Mondragón, J. Therm. Stress., **43**, 543-562 (2020).