## **Electronic Supplementary Information**

# Magnetically-accelerated large-capacity solar-thermal energy storage within high-temperature phase-change materials

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### Theoretical simulation of charging process

#### **Fixed charging**

The numerical simulation of melting process was carried out by FLUENT software with the following assumptions: (1) the system is considered as one-dimensional and semi-infinite; (2) constant heat flux boundary condition (Neumann boundary condition) is applied to the left boundary; (3) heat losses due to radiation, convection, and evaporation are not considered; (4) free convection of melted PCMs is neglected; (5) physical properties of PCMs listed in Table S1 are isotropic and independent of temperature.

The one-dimensional heat conduction problem is solved by the transient thermal solver in the Solidification/Melting module of FLUENT. The governing energy equation can be described by the following partial differential equation:

$$\rho \frac{\partial H}{\partial t} = k \frac{\partial^2 T}{\partial x^2} \tag{1}$$

where  $H = h + \Delta H$ ,  $h = h_{ref} + \int_{r_{ef}}^{T} c_p dT$ ,  $\Delta H = \beta L$ ,  $\beta = \frac{T - T_{solid}}{T_{liquid} - T_{solid}}$ , and H is the total enthalpy, h is the sensible heat enthalpy, L is the latent heat,  $\beta$  is the liquid fraction ( $\beta = 0$  represents solid,  $\beta = 1$  represents liquid), k is the thermal conductivity,  $\rho$  is the density,  $T_{liquid}$  and  $T_{solid}$  are the temperature of liquid and solid, respectively, while T is the transient temperature. The latent heat of molten and phase-transition temperatures are obtained from DSC measurement. The reported heat capacity and density for the 40 wt% KNO<sub>3</sub>-60 wt% NaNO<sub>3</sub> eutectic molten salts are used.<sup>1, 2</sup> To solve this equation, the following boundary and initial conditions are imposed. Boundary conditions:

$$-k\frac{\partial T}{\partial x}|_{x=0} = q_0$$
, where  $q_0$  is the constant heat flux (2)

$$-k\frac{\partial T}{\partial x}\Big|_{x=\alpha} = 0$$
(3)

The same adiabatic boundary conditions are applied to the top and bottom surface.

Initial condition:

 $T_{t=0} = T_0$ , where  $T_0$  is the initial temperature (4)

With the continuous heating at x = 0, the temperature sequentially increases. When the temperature reaches the phase change point of the PCMs, the solid PCMs starts to melt generating a solid-liquid interface in between.

#### **Movable charging**

A 'step-by-step' model based on energy conservation within a controlled volume was developed to simulate the melting process of PCM under movable heating with the following assumptions: (1) the system is considered as one-dimensional and semi-infinite; (2) the energy distribution of the incident light in the melted liquid PCM region follows the Beer–Lambert law; (3) during the charging process thermal conduction in the PCMs can be neglected based on the fact that the conduction heat flux is much smaller than the heat generation induced by solar radiation; (4) heat losses due to radiation, convection, and evaporation are not considered; (5) physical properties of the PCMs listed in Table S1 are isotropic and temperature-independent.

The whole PCM is divided into small slices ( $\Delta x$ ) in *x* direction and the slices were melted one by one. The attenuation of incident solar energy in the melted liquid region follows the Beer–Lambert law:

$$I = I_0 exp^{[i0]}(-\alpha x) \tag{5}$$

where  $\alpha$  is the absorption coefficient of liquid PCM and  $I_0$  is the power density of the solar beam. Based on the solar radiance distribution, an average absorption coefficient of 0.125 cm<sup>-1</sup> was used.<sup>3</sup>

For the first slice in the solid region (the *n*-th slice), we assume the incident light induces a uniform heating within this controlled volume. By neglecting heat conduction, the following energy conservation can be written as:

$$I_0 exp[-\alpha(n-1)\Delta x]t_n = \rho\Delta x [L + c(T_m - T_0)]$$
(6)

where  $\Delta x$  is the step size (0.01 cm),  $T_{\rm m}$  is the melting temperature. The left side of Eq. (6) represents the solar energy absorbed in the controlled volume. The right side is the energy needed to melt the solid PCM in the controlled volume including both the sensible heat and the latent heat. After melting the *n*-th slice, the solid-liquid interface moves to  $x_n = n\Delta x$ , and the illumination time  $t(x_n)$  is sum of total time to melt the first *n* slices that is given by:

$$t(x_n) = \sum_{i=1}^n t_i = \frac{\rho x_n [L + c(T_m - T_0)]}{n I_0} \cdot \frac{1 - exp(x_n \alpha)}{1 - exp(\alpha x_n/n)}$$
(7)

Table S1. Thermophysical properties of molten salt and parameters for theoretical simulation

Material	Molten salt	
properties	Solid	Liquid
ho (kg m <sup>-3</sup> )	2200	1900
<i>c</i> (J kg <sup>-1</sup> K <sup>-1</sup> )	1210	1470
$k (W m^{-1}K^{-1})$	0.5	0.48
$\alpha$ (cm <sup>-1</sup> )	-	0.125
$L (J \text{ kg}^{-1})$	121000	
$T_m$ (°C)	210	
$T_0$ (°C)	25	
<i>P</i> (W m <sup>-2</sup> )	30000	
$I_0$ (W m <sup>-2</sup> )	30000	

#### **References:**

- 1 B. Dudda and D. Shin, Int. J. Therm. Sci., 2013, 69, 37-42.
- 2 R. Serrano-López, J. Fradera and S. Cuesta-López, *Chem. Eng. Process.*, 2013, 73, 87-102.
- 3 M. Tetreault-Friend, L. A. Gray, S. Berdibek, T. McKrell and A. H. Slocum, *Sol. Energy*, 2017, **153**, 238-248.



**Fig. S1.** SEM characterization of pristine iron mesh and magnetic photothermal mesh (MPM) solar absorber. (a) SEM images of iron mesh showing large pore size and unprotected rough surface. (b) SEM of MPM-1 showing reduced pore size and smooth surface. (c) SEM image of MPM-3 showing full blockage of the pores.



**Fig. S2.** Comparsion of corresion resistance. (a) Photographs of as-cleaned pristine iron mesh before and after exposure in humid air and molten salts for 24 h. (b) Photographs of MPM-2 before and after exposure in humid air and molten salts for 24 h.



**Fig. S3.** TGA curves of MPM-2 and neat polydimethylsiloxane (PDMS). The observed slight weight loss of the MPM-2 sample around 400 °C is due to the decomposition of PDMS.



**Fig. S4.** Transmittance spectra of iron mesh and magnetical photothermal mesh (MPM) samples. The increased loading of graphite particles leads to thicker coating layer on the surface of MPM and smaller optical transmittance of the MPM solar absorber.



Fig. S5. Temperature evolution profiles of iron mesh and MPM under 4 kW m<sup>-2</sup> solar radiation.



**Fig. S6.** Schematic of solar-thermal energy harvesting within molten salt through fixed and movable charging.



Fig. S7. Schematic simulation model for charging processes. (a) Fixed charging. A constant heat flux ( $I_0$ ) is applied on one side to transfer into the molten salts and induce solid-to-liquid phase transition. (b) Movable charging. A constant flux of solar energy ( $I_0$ ) is applied on one side to penetrate through the melted salts and continuously advance the solid-to-liquid phase transition in a step-by-step manner with a step size of  $\Delta x$ .



**Fig. S8.** Simulated charging interface movement of neat molten salts charged under fixed charging and magnetically movable charging mode. The same energy flux of 30 kW m<sup>-2</sup> was applied for both fixed and movable charging.



**Fig. S9.** Repeated movable optical charging through manipulating MPM within the motlen salts.



**Fig. S10.** Temperature distribution of the liquid molten salts after continous magneticallymovable optical charging.



Fig. S11. Magnetically-accelerated optical charging of molten salts embedded with heattransfer pipelines. (a) Schematic charging the molten salts by tailoring the shape of mesh-

structured solar absorber. (b) Temperature distribution of the charged sample after 30 kW m<sup>-2</sup> solar illumination for 16 min. (c) Advancement of charging interface.