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## Inverse method to describe crystallization of undercooled water in cold storage tank

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### Abstract:

The objective of this work is to a physical model to describe the thermal performance of cylindrical tank filled with spherical water capsules during the storage period. A particularity is encountered in this type of storage; it refers to the supercooling phenomenon caused by the delay in the crystallization of the water in the capsules. The originality of this model consists in evaluating the heat flux exchanged between the heat transfer fluid (HTF) and the spherical capsules by using experimental data (here, the temperature) and an inverse method. The inverse method is addressed with a genetic algorithm (GA). The heat flux determined by the inverse method for one case of flowrate condition has been applied for other flowrates to evaluate the temperature of HTF at the outlet of the tank during the storage period. The obtained results are then compared with experimental results from the literature. A good agreement was found between the numerical and the experimental results. The influence of the flowrate of the HTF on the shape of the heat flux released by the capsules during the storage period is also analysed and discussed.

**Keywords:** Latent storage, Supercooling, Crystallization, Inverse method, PCM

## 1. Introduction

Currently, latent thermal energy storage (LTES) using Phase Change Materials (PCMs) provides the opportunity for storing a significant amount of energy at an approximately constant temperature [1-2]. Such storage has a significantly higher energy volumetric density than that attainable by sensible storage [3-5]. In the past years, a number of latent thermal technologies with different phase-change materials have been experimentally and numerically investigated [6-8]. In these works, the main aspects that have been considered are design of storage systems, PCMs used, and methods to optimize flow and heat transfer during the charge and discharge operations [9-11]. Based on new research [12-15], spherical PCM capsules are very promising in LTES. Spherical capsules are recommended due to the advantageous relationship between stored energy and heat transfer surface and to their easy replenishment with a good porosity [14-16]. In most cases, LTES consist of cylindrical tanks filled with spherical PCM capsules allowing latent heat storage of large amounts of energy. Such systems include a fixed bed of PCM capsules (i.e. packed bed), which is traversed by a heat transfer fluid (HTF) to accomplish thermal charging, or discharging by convective heat transfer [17].

A number of physical models have been developed to evaluate and investigate the energy performance in a packed bed employing spherical PCM capsules as a storage medium [18-21]. Gracia and Cabeza [22] presented a detailed study concerning different physical models to analyse flow and heat transfer inside a latent storage system comprising PCMs confined in spherical capsules. According to these authors, there are two main groups of numerical models: the homogeneous model and the heterogeneous model. The first one regards the PCM capsules as an equivalent homogeneous medium with a single local temperature [23-24]. This model is rarely used since the assumption of a single local temperature for both solid and liquid media is only valid when the PCM capsules have a conductivity close to that of the HTF or when they have a small diameter [25-26]. The heterogeneous model considers thermal gradients within the capsules. For each of these physical approaches, there are one- or two-dimensional models depending whether or not the radial gradient in the packed bed is accounted for.

Three basic heterogeneous models can be identified: Schumann's model, Concentric Diffusion Model (*CDM*) and Continuous Solid Phase Model (*CSPM*) [22]. The Schumann model is unidimensional and considers the heat transfer between the HTF and PCM capsules [27].

However, this model presents some limitations as it ignores axial heat conduction in the HTF and the PCM, and radial diffusion in the capsule [28-29]. The CDM is a two-phase heterogeneous model that has the particularity of considering the thermal conduction inside the PCM [30-31]. It is important to note that once the PCM starts melting within the particle, two modes of heat transfer are occurring: conduction in solid phase and natural convection in liquid phase. However, due to the complexity of simulations, natural convection is not taken into account in this model. However, an effective conductivity can be included to improve the model [32-33]. The CSPM considers the spherical capsules as a continuous medium and not as a medium composed of independent PCM capsules. As a result, the thermal conduction within the PCM is not taken into account. However, axial and/or radial conduction in the PCM capsule can be considered [34-35]. It is interesting to note that the analysis of latent heat energy storage containing encapsulated PCMs with detailed numerical models can be time consuming. Due to this limitation, simplified models, which can rapidly predict the transient thermal performance of the packed bed, are highly required in order to integrate them into the dynamic simulation software of energy systems. However, simplified models suffer from the accuracy of prediction due to multiple hypotheses.

In advanced simulations of flow and heat transfer in LTES, the impact of supercooling is generally omitted. This approach is possible if the supercooling is negligible in comparison to the temperature variations in the packed bed, but for important supercooling, the obtained information can be erroneous. In this case, supercooling needs to be addressed in the numerical model. None of the models described above includes supercooling. In addition, most models described in the literature do not consider the random crystallization of PCM in spherical capsules [36-38]. Despite the introduction of additives or nucleation catalysts can reduce it [36-37], supercooling cannot be completely avoided and it is important to address it.

Supercooling has two potential concerns: (i) the crystallization temperature may be outside the temperature region monitored by the system, and (ii) the energy consumption of the compressor to cool the PCM can be higher. In addition, the random nature of this phenomenon complicates the management of the latent storage system [36-37].

Several PCMs are concerned by the supercooling phenomenon, which for industrial applications is challenging since it blocks the liberation of the stored latent energy. However, the supercooling feature offers an attractive interest to store thermal energy for prolonged times with no loss of heat, as the undercooled liquid is retained at normal ambient temperature. The latent heat stored can be discharged by activating the crystallization of

undercooled PCM. Numerical studies demonstrate that the performance of latent heat installations is improved when supercooling effects are exploited [39].

Water is commonly accepted as a PCM in cold storage applications thanks to its characteristics of high-latent heat, availability and manageability. However, there are three phenomena that occur during cooling and crystallization of water in spherical capsules: density inversion, supercooling phenomenon and ice's volume expansion.

A few studies have been carried out on the numerical simulation of the crystallization process of a PCM in a storage tank [31, 40-44]. They have only mentioned the supercooling phenomenon and relatively few of them have focused on its modelling and its impact on the overall performance of the storage system. Bedecarrats et al. [45-46] and Kousksou et al. [47-48] are among the few authors to have developed a detailed physical model to investigate the effect of supercooling on the thermal behaviour of a latent storage system. These authors developed concentric diffusion models to investigate in detail supercooling, flow and heat transfer during charging and discharging modes for two storage tank positions (vertical and horizontal). These models are verified and validated using experimental results [46]. Numerical results indicate that the performance of the latent heat storage system can be optimized by judicious choice of HTF flow rate and packed bed position.

Physical modelling of flow, heat transfer and stochastic behaviour of the crystallization of supercooled water contained in spherical capsules randomly distributed in a latent thermal storage system is challenging. Some of the principal phenomena to be modelled during charge mode include the following:

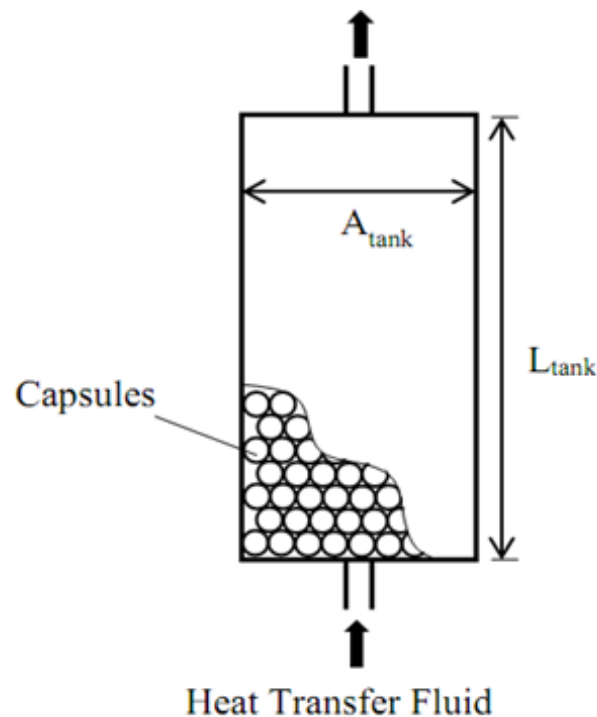
- Heat transfer between HTF and spherical capsules.
- Contact between capsules.
- Water density inversion during the cooling process.
- Volume expansion of ice inside spherical capsules.
- Solid-liquid interface movement.

Such phenomena makes the physical modelling of the storage tank during the crystallization process very complicated. The objective of this work is to suggest an inverse method to characterize the overall performance of the storage tank avoiding the need for modelling heat transfers between the HTF and the spherical PCM capsules. This method consists in evaluating the heat flux released by PCM in the HTF by comparing the numerical results of

the physical model with the experimental data (here, the temperature). The inverse method is addressed with a genetic algorithm (GA).

## 2. Description of the latent energy storage system

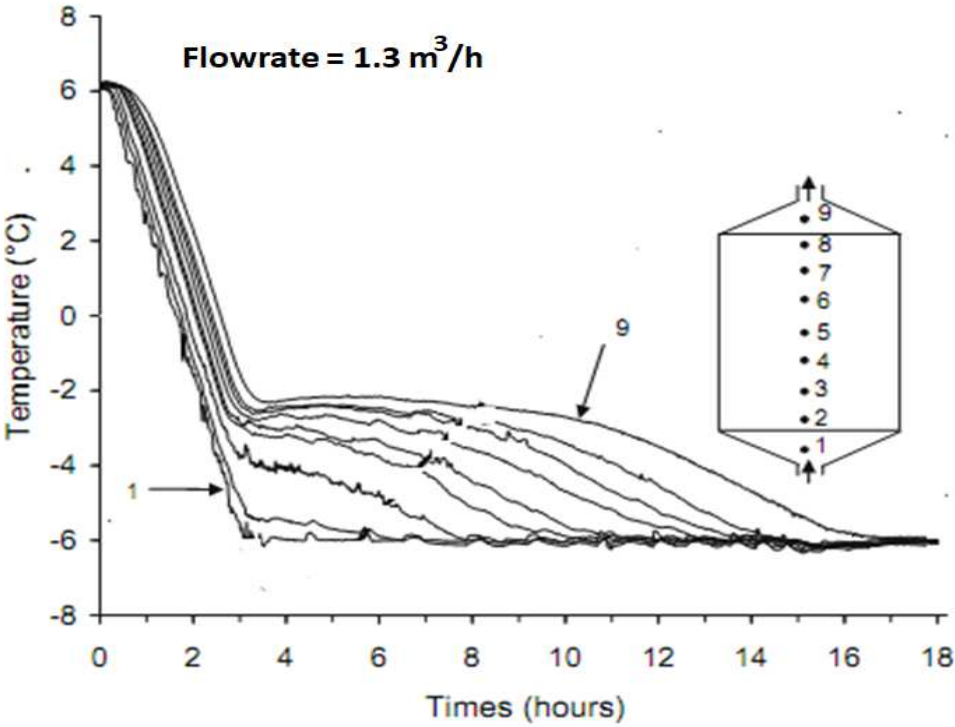
**Fig.1** illustrates the picture of the storage tank, which was studied by Bedecarrats et al. [45-46] and Kousksou et al. [47-48]. Its useful height is  $L_{\text{tank}} = 1.42$  m and its internal diameter is  $D_{\text{tank}} = 0.94$  m. It contains 2400 spherical capsules containing water as PCM and with a diameter of  $D_{\text{PCM}} = 77$  mm. The tank is insulated with polyurethane foam. The tank is initially at temperature  $T_0$  higher than the melting temperature of ice. During the charging phase, the temperature of the HTF (solution of monoethylene glycol) at the inlet of the tank decreases continuously until it reaches a value called the final inlet temperature. The heat transfer fluid flows from the bottom to the top of the tank.



**Fig.1:** Packed bed

**Fig.2** shows the experimental inlet and outlet temperatures obtained by Bedecarrats et al. [45]. The HTF volume flow rate is equal to  $1.3 \text{ m}^3/\text{h}$ . The initial temperature of the tank is  $6.1^\circ\text{C}$ , the cooling rate of the HTF is  $\beta = -4^\circ\text{C} / \text{h}$  and  $-6^\circ\text{C}$  is the final temperature of the heat

transfer fluid at the inlet of the tank. Between  $t=0$  s and  $t=30$  min the HTF temperature at the outlet of the tank remains practically constant. This time corresponds to the time required to allow the HTF to circulate through the whole tank. Between  $t=1$ h and  $t=3$ h, the fluid temperature at the outlet of the tank decreases according to the inlet temperature. This period corresponds to sensible storage. Between  $t=3$ h and  $t=10$ h, the fluid temperature at the outlet of the tank remains at around  $-2.2^{\circ}\text{C}$ . This part corresponds to latent storage (i.e. the crystallization of water in the capsules). One can note that the degree of supercooling in this case is around  $2.3^{\circ}\text{C}$ . From  $t=10$ h to  $t=17$ h, the HTF temperature at the outlet of the tank decreases to reach the HTF temperature at the inlet of the tank. This period corresponds to sensible storage. It can be concluded that the storage time with a flow rate of  $1.3\text{ m}^3/\text{h}$  is around 17 hours. It is interesting to note that the analysis of the HTF temperature curves between the inlet and outlet of the packed bed permits to confirm that the HTF temperature is only dependent on the tank height. As a result, HTF flow in the packed bed appears to be similar to a piston flow.



**Fig.2:** Experimental HTF temperature at various positions inside the tank [45]

**3. Mathematical formulation of direct and inverse problem**

As described in section 3, during the charging process, the cold HTF circulates from the bottom to the top of the cylindrical tank vertically in order to neglect the natural convection in the HTF. To simplify the direct physical model, a number of hypotheses were proposed as follows:

- The storage tank is well insulated so that heat loss to surrounding areas is omitted.
- As has been shown in experimental study [45], the flow in the tank is unidirectional (x) and incompressible.
- The spherical PCM capsules are considered as a source term in the HTF.

$$\rho_f c_f \frac{\partial T_f}{\partial t} + \rho_f c_f u_f \frac{\partial T_f}{\partial x} = \lambda_f \frac{\partial^2 T_f}{\partial x^2} + \Phi(x, t)$$

(1)

where  $T_f$  is the HTF temperature,  $c_f$  is the specific heat capacity of the HTF,  $\rho_f$  is its density and  $\Phi$  is the rate of heat production per unit volume caused by the spherical PCM capsules. The rate of heat production relies on the state of the PCM in the spherical capsules: i.e. when the PCM is liquid, solid or if both phases coexist.

Based on the experimental study described by Bedecarrats et al. [45] the boundary conditions completing equation (1) can be written as follows:

$$T_f(x=0, t) = \beta t + T_o, \quad \beta < 0$$

(2)

$$\left( \frac{\partial T_f}{\partial x} \right)_{x=L_{\text{tank}}} = 0$$

(3)

where  $\beta$  is the cooling rate and  $T_o$  is the tank temperature at  $t = 0$ . During the cooling process,  $T_f(x=0, t)$  decreases linearly until reaching  $T_s$ . Then,  $T_f(x=0, t)$  is maintained at  $T_s$ .

**Eq.1** can be solved using a fully implicit finite volume method to describe flow and heat transfer inside the HTF (see **Fig.3**). The first-order upwind scheme was applied to address convective terms, and diffusion terms are discretized using the second order of the central



differential scheme. Therefore, the resulting expression of the system of energy equations is as follows:

$$A_{i,f} T_{i,f}^{t+\Delta t} = A_{i+1,f} T_{i+1,f}^{t+\Delta t} + A_{i-1,f} T_{i-1,f}^{t+\Delta t} + C_{i,f} \quad (4)$$

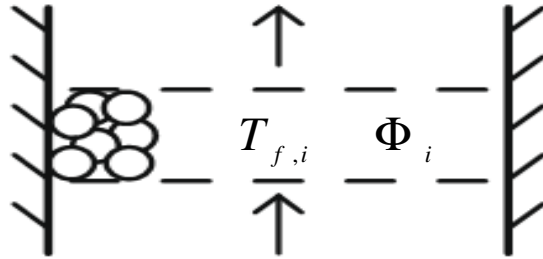
where

$$A_{i,f} = \frac{(\rho c)_f}{\Delta t} + \frac{2\lambda_f}{\Delta x^2} \quad (5)$$

$$A_{i+1,f} = \frac{\lambda_f}{\Delta x^2} - \frac{\rho_f c_f u_f}{2\Delta x} \quad (6)$$

$$A_{i-1,f} = \frac{\lambda_f}{\Delta x^2} + \frac{\rho_f c_f u_f}{2\Delta x} \quad (7)$$

$$C_{i,f} = \frac{(\rho c)_f}{\Delta t} T_{i,f}^t + \Phi_i \quad (8)$$



**Fig.3:** Scheme of the mech

In this paper, **Eq. 4** can be solved iteratively by using a Tridiagonal Matrix Algorithm (TDMA) method [49]. The computation procedure was carried out by using Fortan 90.

For the inverse model, it is supposed that the source term  $\Phi$  in **Eq.1** is unknown while the thermophysical properties of the HTF are known. The objective of the inverse problem is to determine at each instant the source term  $\Phi$  by using experimental temperature [45]. So, the determination of  $\Phi$  from experimental data can be constructed as an optimum control problem, which aims at minimizing the cost function  $f(\Phi)$  :

$$f(\Phi) = \sum_{i=1}^{N_{cv}} \left( \frac{T_i^{\text{exp}} - T_i^{\text{num}}}{T_i^{\text{exp}}} \right)^2 \quad (9)$$

where  $N_{cv}$  the number of control volumes in the tank,  $T_i^{\text{exp}}$  are the experimental temperatures and  $T_i^{\text{num}}$  are the temperatures generated from the numerical solution of the direct physical model.  $N_{cv}$  is chosen to have in each control volume a number of spherical capsules which is an integer. The solution of the inverse problem is obtained by minimizing  $f(\Phi)$ . Various optimization algorithms can be employed to address the optimization problem, **Eq.(9)**. In our case, the Genetic Algorithm (GA) was adopted for solving the inverse problem [50-51]. **Table 1** reports initial setting parameters used in GA for this study. In addition, the population size of GA is 30 and the algorithm is accomplished by 400 iterations.

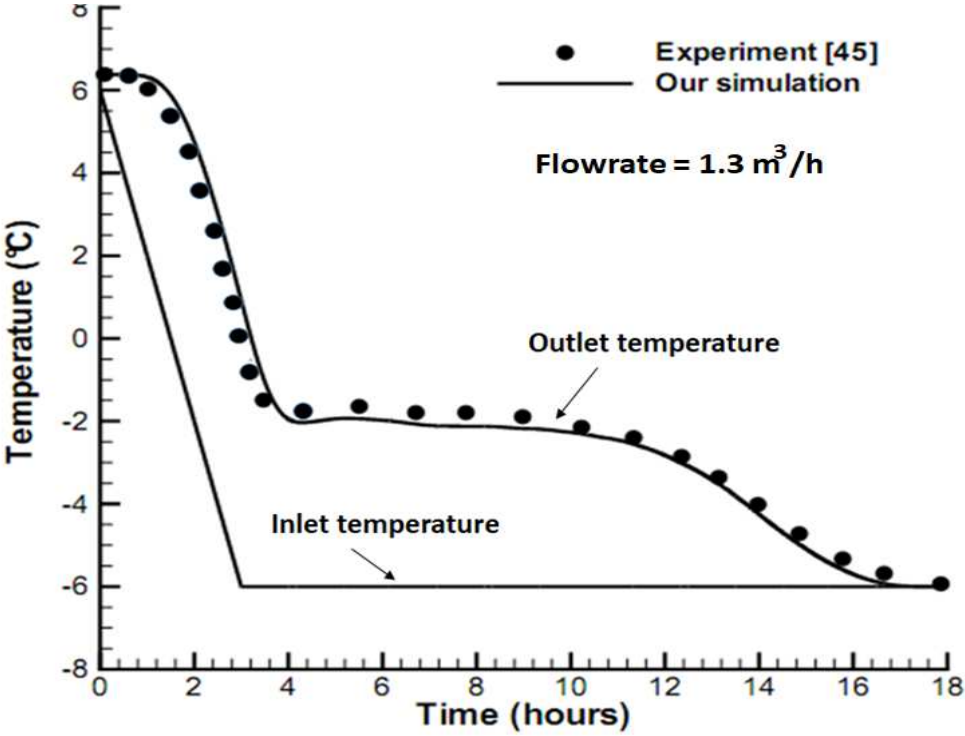
**Table 1:** The specification of the Genetic algorithm

GA's parameters	Parameter value
Mutation percentage	40
Mutation rate	0.05
Crossover percentage	70
Crossover adjustment	0.05

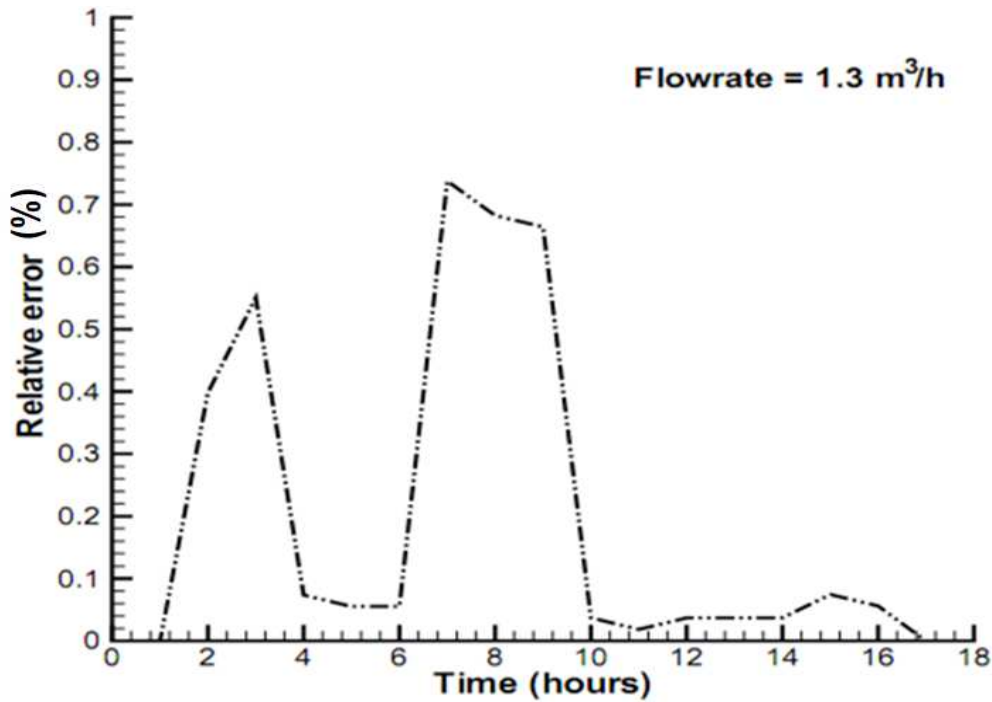
#### 4. Results and discussion

**Fig.4** shows the evolution of the numerical and experimental heat transfer fluid temperature and the rate of heat production per unit volume at the outlet of the tank as a function of time under the same experimental conditions of Bedecarrats et al. [45] (see **Fig.2**). We have presented on the same figure (i.e. Fig.3) the evolution of the HTF temperature at the inlet of the tank, which follows the law presented by equation 2. We have also presented in **Fig.5**, the relative error between the numerical results and the experimental data  $\left( \frac{T_{\text{exp}} - T_{\text{num}}}{T_{\text{exp}}} \right)$ . We can notice that the temperatures of the heat transfer fluid at the inlet and outlet of the tank are

similar to those obtained experimentally by Bédécarrats et al. [45]. Considering the obtained curves, we can conclude that the numerical temperature of the HTF at the outlet of the tank determined from the above physical formulation is in good agreement with that of the experiment.



**Fig.4:** Experimental and numerical heat transfer fluid temperature at the outlet of the tank



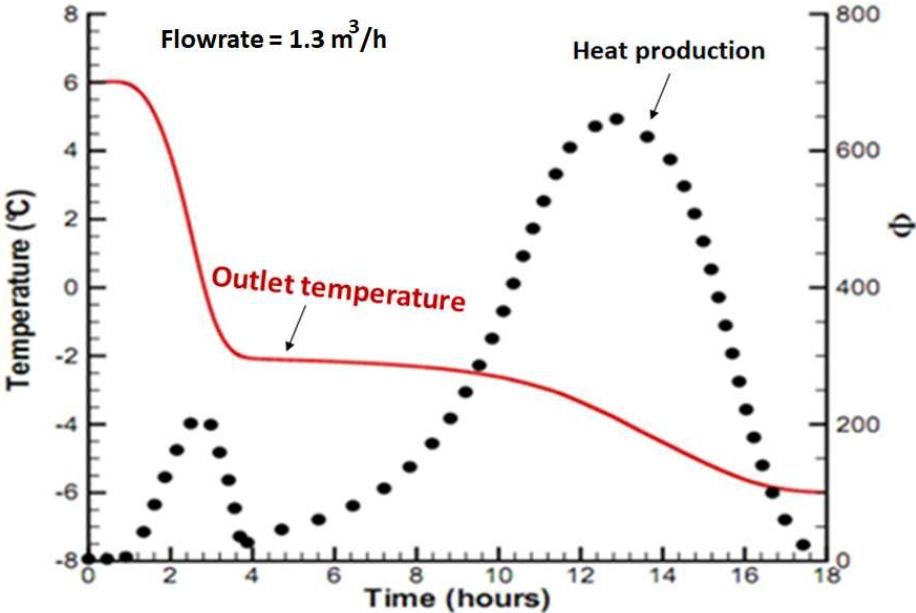
**Fig.5:** Relative error between the numerical results and the experimental data.

**Figs 6-a** show the variation of the rate of heat production per unit volume  $\Phi$  as a function of time at the outlet of the tank. By analysing this figure, one can identify and understand the different phases of the storage process. At the beginning of cooling, the HTF temperature at the inlet tank decreases in a linear way, while the HTF temperature and  $\Phi$  at the outlet tank remain approximately constant for a longer period of time. That time ("residence time") corresponds to the required time that the fluid crosses the full length of the tank. Then, the temperature of the HTF at the outlet tank decreases as a function of the inlet temperature and  $\Phi$  rises to reach a maximum value. During this period, thermal energy is stored in a sensible form since the PCM remains at a liquid state (i.e. water) inside the capsules.

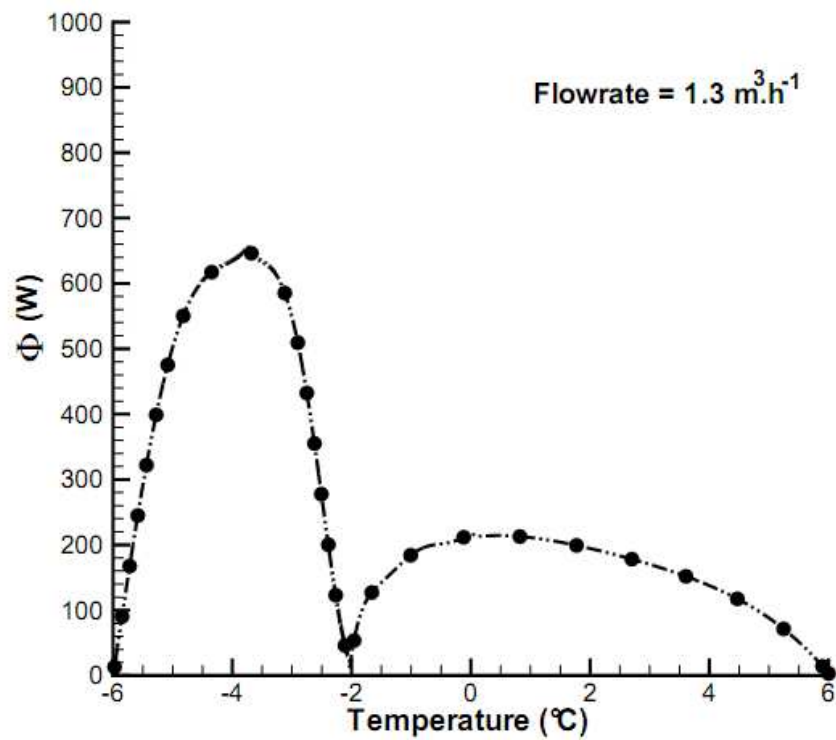
Between 3h and 4h, the PCM inside the spherical capsules is kept in liquid state despite the temperature of the HTF temperature ranging from zero to  $-2.1^{\circ}\text{C}$ . It can be concluded that the PCM is metastable throughout this stage. During this stage,  $\Phi$  decreases noticeably to attain a certain minimum value. During crystallization of the PCM, which starts around  $-2.1^{\circ}\text{C}$ , the HTF temperature decreases slowly and then forms approximately a plateau while  $\Phi$  increases sharply to reach a maximum value, which marks the end of PCM crystallization inside the capsules. After that maximum, the HTF temperature decreases rapidly to reach the inlet temperature. During this period, the heat flux  $\Phi$  released from the capsules diminishes, and

there is a sensible cooling of ice within the capsules. One can also conclude that the rapid increase in the heat flux  $\Phi$  during PCM crystallization in the capsules can be explained by the release of latent energy. This energy is absorbed by the HTF and stabilizes the temperature of that fluid in the tank. Finally, one can conclude that the analysis of the graph that represents  $\Phi$  versus time allows us to determine easily the beginning and the end of PCM crystallization in spherical capsules.

**Fig. 6-b** illustrates the evolution of the heat flux  $\Phi$  released from the capsules at each control volume as a function of the temperature of the heat transfer fluid. We notice that the heat flux released by the capsules has the same shape for all control volumes. During continuous cooling between 6°C and 0°C, all capsules store only sensible energy. Between 0°C and -2°C, the PCM remains liquid due to the supercooling phenomenon. During this period, the heat flux  $\Phi$  decreases to reach a minimum value at -2°C. As soon as the supercooling is broken, the heat flux  $\Phi$  increases to reach a maximum value. This increase is due to the crystallisation of the water in the spherical capsules. After that maximum, the heat flux  $\Phi$  released from the capsules diminishes, and there is only a sensible cooling of ice inside the capsules. It is interesting to note that from this curve (Fig.6-b) a correlation was found between the heat flux  $\Phi$  and the temperature of the heat transfer fluid. This correlation was introduced in Eq. 1 to evaluate the heat flux released by the capsules at each control volume.

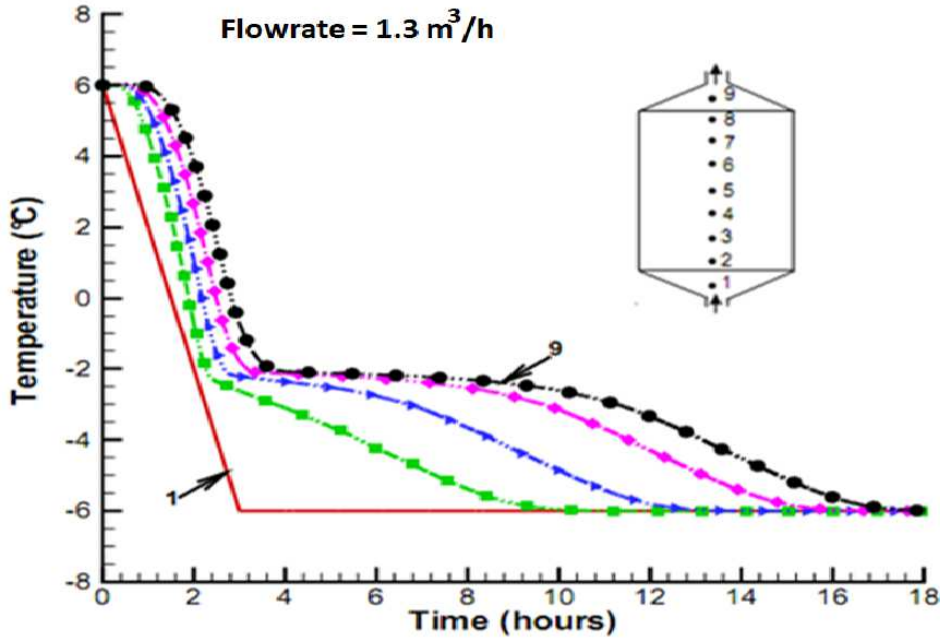


**Fig.6-a:** Heat transfer fluid temperature and heat production at the outlet of the tank



**Fig.6-b:** Heat production versus heat transfer fluid temperature

The physical model permits to describe the temperature evolution between the inlet and the outlet of the tank (see [Fig.7](#)). The stratification of the HTF temperature corresponding to a piston flow inside the tank can be confirmed. One can also notice that the PCM capsules near the inlet of the tank crystallize quickly compared to those close to the outlet of the tank. In our case, we suppose that the cold thermal storage is finished when the HTF temperature at the outlet of packed bed reaches that imposed at its inlet.



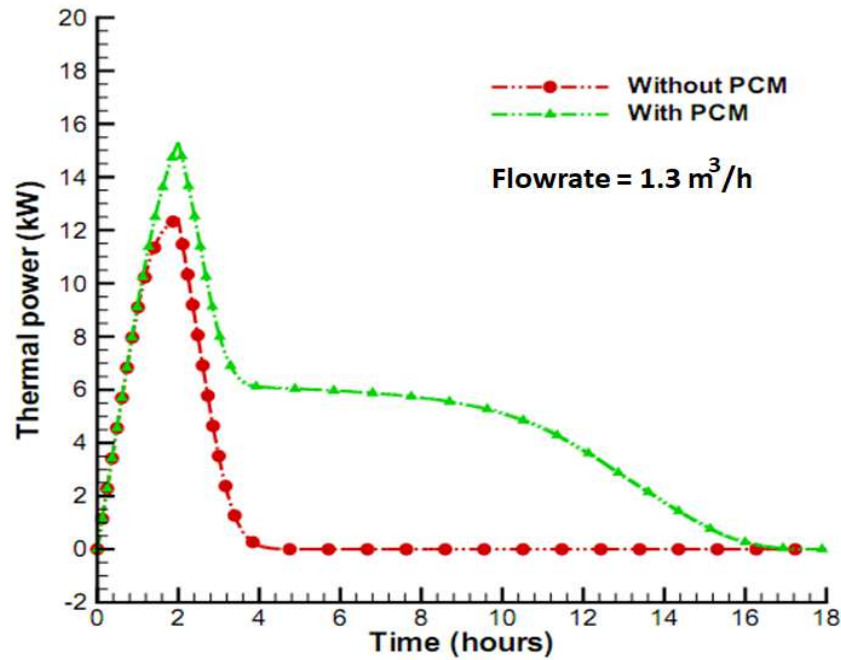
**Fig.7:** Numerical HTF temperature at various positions in the tank

The time dependent thermal power absorbed by the HTF during the storage mode may be calculated according to the following relationship:

$$P(t) = \rho_f c_f q_v (T_f(x=L, t) - T_f(x=0, t)) \quad (10)$$

where  $q_v$  is the flowrate of the HTF.

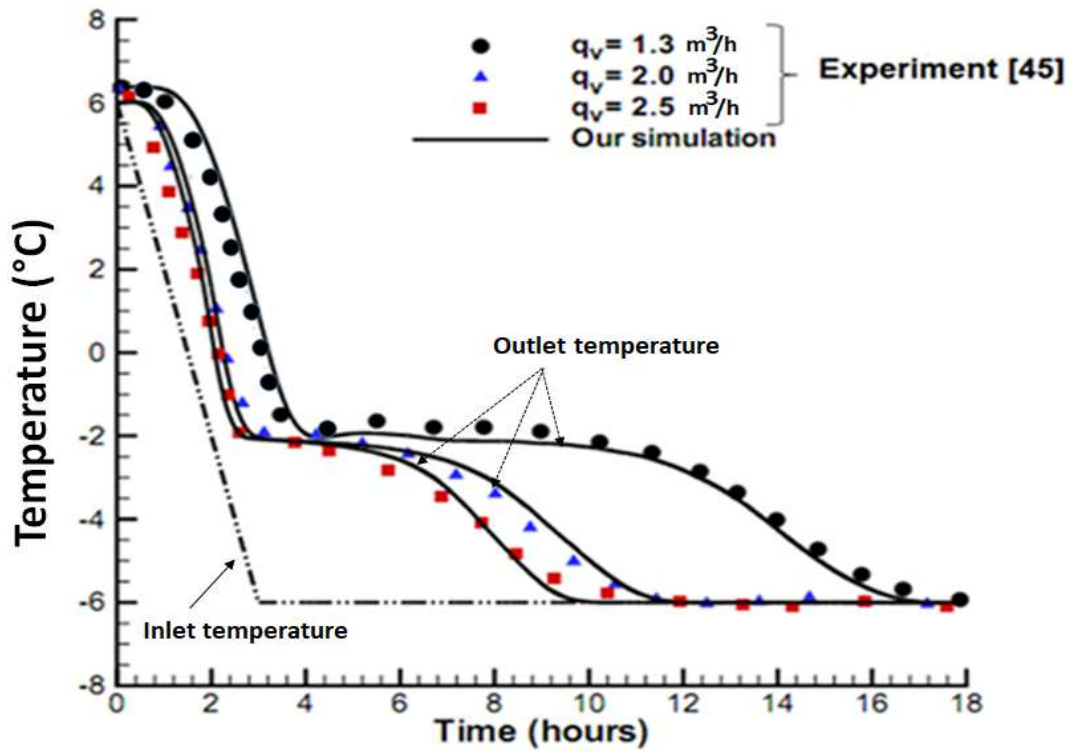
The evolution of thermal power  $P$  as a function of time during the storage phase is shown in **Fig. 8** for two cases: with and without PCM. We notice that at the beginning of the continuous cooling process, the thermal power  $P$  increases rapidly to reach a maximum. During this period, the HTF temperature at the outlet of the tank remains practically constant. Once a maximum is reached, the heating power decreases rapidly until it is cancelled in the absence of the PCM. When the tank is filled with capsules enclosing water, the thermal power decreases to stabilize during the crystallization process of PCM in the capsules. At the end of latent thermal storage, the thermal power decreases in order to be cancelled. One can notice that the use of PCM allows to store an important part of latent energy compared to a thermal sensible storage without PCM.



**Fig.8:** Thermal power  $P$  versus time during storage process

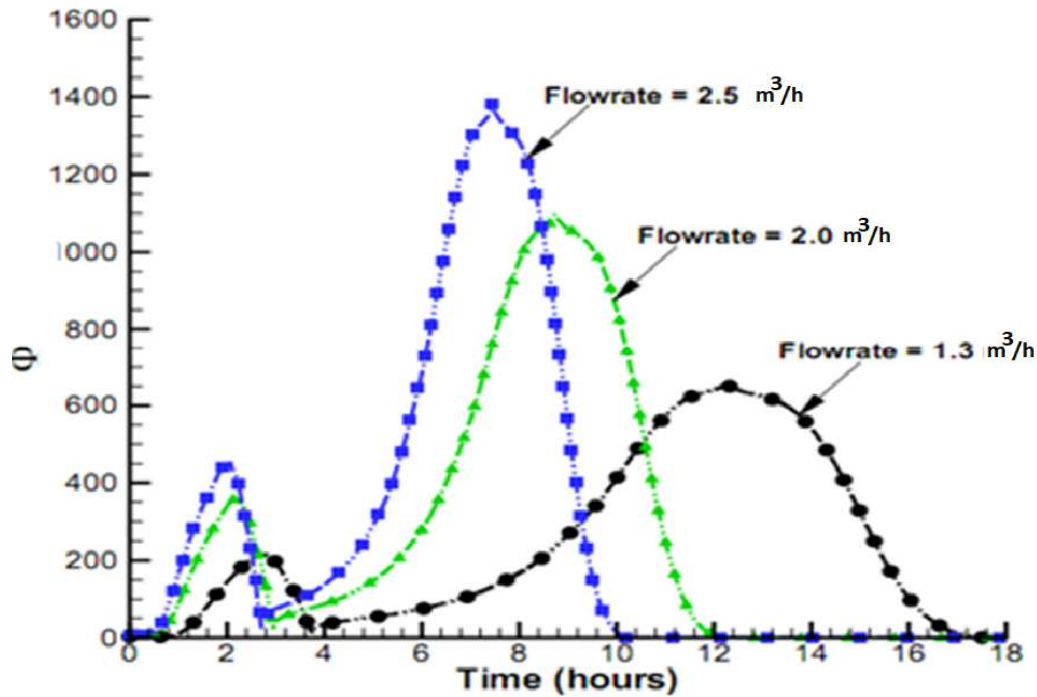
The heat flux determined by the inverse method for the flowrate of  $1.3 \text{ m}^3/\text{h}$  condition has been applied for other flowrates ( $2.0 \text{ m}^3/\text{h}$  and  $2.5 \text{ m}^3/\text{h}$ ) to evaluate the temperature of HTF at the outlet of the tank during the storage period. **Fig.9** presents the impact of the flowrate on the outlet temperature of the HTF at the outlet of the tank. On the same figure, we also compared the numerical results and the experimental data for different flowrates. We can observe that for the flowrates ( $2.0 \text{ m}^3/\text{h}$  and  $2.5 \text{ m}^3/\text{h}$ ), the numerical temperature of the HTF at the outlet of the tank determined from the physical model coupled with the inverse method is in good agreement with that of the experiment. We can also note that the storage duration is faster under higher flow rates. This result can be explained by the fact that increasing the HTF velocity inside the packed bed improves the heat transfer between the HTF and the spherical capsules. However, a certain compromise needs to be achieved between the storage time and the pressure losses across the tank.





**Fig.9:** Numerical HTF temperature at the outlet of the tank for various HTF flowrates

**Fig.10** displays the effect of HTF flow rate on  $\Phi$  at the outlet of the packed bed. As we can see, the crystallization time of PCM inside the capsules increases with decreasing HTF flow rate. This result confirms that the phase transition kinetics in the capsules depend on the heat transfer coefficient between the HTF and the spherical capsules. This coefficient increases with increasing HTF flow rate. When the crystallization of the MCP inside the capsules is completed, the flow decreases sharply and then cancels out.



**Fig.10:**  $\Phi$  at the outlet of the tank for various HTF flowrates

## Conclusion

In this work, a physical model is adopted to describe the heat transfer during crystallization of undercooled water confined in spherical capsules and randomly dispersed in a cylindrical tank. The heat flux exchanged between the heat transfer fluid (HTF) and the spherical capsules is calculated by using experimental data (here, the temperature) and an inverse method. The inverse problem is addressed with a genetic algorithm (GA). The heat flux determined by the inverse method for one case of flowrate condition has been applied for other flowrates to evaluate the temperature of HTF at the outlet of the tank during the storage period. The results obtained by the proposed model show a good agreement with experimental results from the literature. Work in progress to verify the validity of our formulation under other conditions and for other latent storage systems.

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