PCM/ graphite foam composite for thermal energy storage device

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Abstract. Numerical studies are proposed to predict and investigate the thermal characteristics of a thermal storage device consists of graphite foam matrix saturated with phase change material, PCM. The composite (graphite foam matrix saturated with PCM) is prepared by impregnation method under vacuum condition, and then is introduced into a cylindrical shell and tube device while it experiences its heat from an inner tube fluid. The two-dimensional numerical simulation is performed using the volume averaging technique; while the phases change process is modelled using the enthalpy porosity method. A series of numerical calculations have been done in order to analyze the influence of fluid operating conditions on the melting process of the paraffin/ graphite foam. The results are given in terms of temperature or liquid fraction time history in paraffin/ graphite foam composite, which show that the heat transfer rate of the device is effectively improved due to the high thermal conductivity of graphite foams. Therefore, paraffin/ graphite foam composite can be considered as suitable candidates for latent heat thermal energy storage device.

1. Introduction
High Efficiency thermal storage device is an important requirement for heat recovery, solar energy utilization and seasonal storage since it can bridge the time gap between energy requirements and energy use.

There are sensible heat and latent heat (i.e., phase change) stores in the thermal energy storage device. Thermal storage utilizing solid–liquid phase change materials (PCMs) is one of the most attractive thermal storage techniques due to its inherent advantage of simplicity and reliability. The major advantages of phase change stores are their large heat storage capacity and their isothermal behavior during the charging and discharging process. Therefore, how to select a suitable PCM should be considered in the high efficient latent thermal storage device. Among the various organic or inorganic PCMs, normal paraffin is an excellent PCM for low temperature thermal energy storage due to its large latent heat, good stability, no subcooling degree and no toxicity [1]. Moreover, various paraffin waxes that both the melting point and the heat of fusion increase with their increasing chain length, so it’s available in a selection of melting point ranges to make a good match between melting range and system operating temperature.

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Although paraffin waxes exhibit desirable properties as PCMs, they present a low thermal conductivity below 0.4 W/(m K). This property increases the thermal resistance of the growing layer of the molten/solidified medium and thereby may cause the surface heat flux decreases, reducing the rate of heat storage further in the thermal storage device.

In order to offset the low thermal conductivity of the PCM, the device must be designed with an adequate heat transfer area and the material must have suitable heat transfer coefficient. Therefore, the structures of the device have been researched, such as spherical capsule [2], unfinned or finned cylinder [3], which leads to reduced effective energy storage capacity. The low thermal conductivity of paraffin wax can also be enhanced, such as like inserting fins [4], metal matrices [5].

Some authors have put forward to disperse high-conductivity particles inside PCM, such as expanded graphite [6], metal nanoparticles [7], to prepare composite PCMs and measure the effect of containing particles on the thermal conductivity of the composite PCM. The thermal conductivity of the composite PCM was found to increase with the loading of metal nanoparticles. Nevertheless, these enhanced methods add significant weight and cost to the storage devices.

Some other authors have used paraffin supported within the porous structure of silica catalyst [8] or activated carbon [9]. The results have found that the mean pore size of the support could be a critical parameter: too small pore size could decrease the latent heat storage capacity. On the other hand, if the pores are too big, there will not be enough capillary forces to retain the wax liquid phase inside the matrix. However, these two porous supports did not improve the effective thermal conductivity [10].

In addition to the aforementioned methods, inserting PCMs into a porous media appears to be an attractive choice to enhance heat transfer. A number of experimental and theoretical investigations on this problem have been conducted in the past two decades. For example, Lafdi et al. experimentally investigated the phase-change heat transfer within a PCM composite permeated with aluminum foam [11]. Li M et al. [12] gave one of the preparation, structure and thermal energy storage property of capric-palmic acid/attapulgite composite PCMs; Zhao et al. [13] experimentally and numerically studied the paraffin melting in metallic foam. The results showed that the paraffin embedded in metal foam (copper or aluminum) was found to be more effective in improving the heat conduction of PCM.

In recent years, development of high thermal conductivity, graphite foams opened a new horizon to thermal storage and thermal management applications. Graphite foams present advantages as the bulk thermal conductivities varying correspondingly with density from 40 to 150 W/m K, densities ranging from 0.2 to 0.6 g/cm³, and good mechanical properties and chemical inertness. It’s good compatibility with many surfaces and has a porous structure with a high porosity, which can reach 95% of the volume consist of void spaces and provides a high ratio of surface area to volume. So Graphite foam can be filled with various materials. On the other hand, graphite foam is known to be superior to porous metallic like aluminum, copper or nickel due to having higher thermal conductivity than conductive metals. The above advantages make graphite/PCMs composites desirable for latent heat thermal storage applications. Py et al. [14] proposed a supported PCM made of paraffin impregnated by capillary forces in a graphite matrix. They found that the thermal conductivity of the composite was equal to that of the sole porous graphite matrix. Moreover, the composite presented the same anisotropy with respect to the compression axis. Zhong et al. [15] characterized the thermal performance of paraffin wax/graphite foam composites using experimental measurements of thermal diffusivity and latent heat, experimental results indicated noticeable improvements in thermal diffusivity of the composite compared to that of pure PCM, especially with lower porosity of the foam. A numerical study has been carried out by Lafdi et al. [16] to investigate the thermal performance of graphite foam with different porosities saturated with the PCM (paraffin wax). The energy absorption rate for graphite/PCM composite was compared with that of pure PCM and found a significant improvement in the energy absorption rate.

Thermal characteristics of a thermal protection system consisted of graphite foam saturated with PCM were investigated [17-19]. The results illustrated that the higher the porosity the more stability of the thermal performance of the matrix composite.
Considering the above-reviewed papers, it can be concluded that utilizing highly-porous, highly-conductive graphite foam is one of the most effective methods of improving thermal conductivity and heat transfer of phase change processes.

In the present work, we focus on the shell-and-tube thermal energy storage device, which the PCM occupies the space between the tube and the shell, and the heat is transferred from the heat transfer fluid (HTF) which flows through a single tube, is a highly attractive option in the energy storage applications. In the last few years, several authors numerically or experimentally analyzed the paraffin wax melting process in a shell-and-tube type storage device, for example, in [20], the temperature distribution and melting times of the pure paraffin wax were studied. Here, a new kind of PCM/graphite composite prepared by commercially available graphite foam under the brand name POCO-HTCTM being infiltrated with paraffin wax is introduced as heat storage material in the device. The thermal characteristics of the proposed device are investigated numerically. The numerical model is based on volume averaging technique and the enthalpy porosity method is used to model the phase change process in the shell-and-tube type storage device. Moreover, parametric study is performed to analyze the effects of influential factors on thermal performance of the proposed device.

2. Experimental

PCM used in the experiments is a kind of commercial paraffin. Thermophysical properties of the paraffin wax are measured and listed in table 1. The latent heat is 173.4 kJ/kg, the thermal conductivity is 0.21 W/mK as solid phase and 0.12 W/mK as liquid phase, and the specific heat is 2890 J/kg K. the melting process starts at 46.45°C (319.6K) and ends at 48.68°C (321.83k).

| Properties                      | PCM1 | Paraffin | Cupper foam | Graphite foam |
|---------------------------------|------|----------|-------------|---------------|
| Density (kg m⁻³)                | 1000 | 747      | 8920        | 500           |
| Specific Heat (J Kg⁻¹K⁻¹)       | 1828 | 2890     | 390         | 700           |
| Thermal conductivity (W m⁻³K⁻¹) | 1.568(solid) | 0.21(solid) | 401         | 150           |
| Latent heat (kJ kg⁻¹)           | 220  | 173.4    |             |               |
| Melting temperature (K)         | 331-333 | 319.6-321.83 |           |               |

![Figure 1. SEM image of graphite foam.](image1.png)  ![Figure 2. SEM image of PCM/graphite foam composite.](image2.png)

The graphite foam used in these experiments is POCO-HTCTM open pore graphite foam provided by POCO company in USA, which has the average pore diameter of 400 ms, total porosity of 72.8%,
density of 0.5 g/cm³, heat capacity of 0.7 kJ/kg·K and the average heat conductivity coefficient of 150 W/m·K.

Two graphite foam cylindrical disks are fabricated to be introduced as the solid matrices of the PCM/graphite foam composites. The graphite foam cylinders have 150 mm height, 126 mm and 25 mm in outer diameter and inner diameter, respectively. Each one of the graphite foam is heated under vacuum in a molten paraffin wax to achieve maximum wax absorption.

Each one of the graphite foam and paraffin wax are filled into a beaker, then is heated to 335K (above the melting point of the paraffin wax 321.83 K) in a vacuum oven for about 5h, after the graphite foam is submerged in the melted paraffin wax to achieve maximum wax absorption, the vacuum oven is cooled until the paraffin wax is solidified under normal atmosphere. Mass ratio of the paraffin wax in the composites is about 51.2%. The morphology of the graphite and PCM / graphite foam composites are observed by means of scanning electron microscopy (SEM, JSM-7500F) at room temperature. As illustrated in figure 1, there are some unconnected areas between pores in graphite foam. It can be known from figure 2 that some of the PCMs have been impregnated to the pores of the graphite foam.

The two composites are introduced into a vertical cylindrical concentric tube, which has inside radius (r) of 12.5mm and outside radius (R) of 63 mm, and the height (H) of 300 mm. The outer tube is well insulated using a suitable layer of Styrofoam. As presented in figure 3, the heat transfer fluid (HTF), water, flows through the inner tube and exchanges heat with PCM of the composite. During charging process hot water heats the PCM, and when PCM melts, the heat is stored. During discharging process, the PCM solidifies and the stored heat is delivered to the cold fluid, which is the working principle of thermal energy storage device.

3. Physical model

The thermal energy storage device used for the present analyze is shown in figure 3. It consists of an inner tube, outer tube and an annulus space filled with paraffin wax/graphite foam composites. The outer tube is insulated. Due to the symmetry, only a half of this domain will be considered in numerical simulation, as shown in figure 4, and the device is formulated in two-dimensional axi-symmetric coordinates. The volume averaged energy equation with phase change is used to model the heat transfer process inside the PCM/graphite composite with phase change.

The enthalpy porosity theory is employed to predict the phase change process of PCM/graphite foam in the numerical model. The thermal properties of the paraffin and graphite foam are listed in table 1.
- Thermophysical properties of the HTF, the tube wall and the PCMs are independent of the temperature;
- Natural convection in the liquid phase of PCM is caused by density variation, which is based on the Boussinesq assumption;

Accordingly, the governing equations used here are as follows:
- for the HTF

**Continuity:**

\[
\frac{\partial (\rho, u)}{\partial t} + \frac{\partial (\rho, v)}{\partial x} = 0
\]  

(1)

Where \( u, v \) is the flow velocity.

**Momentum:**

\[
\frac{\partial (\rho, u_i)}{\partial t} + \sum_{j=1}^{2} (\rho, u_i u_j) = \frac{1}{x_j} \left[ \mu \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) \right] - \frac{\partial p}{\partial x_i} + S_i
\]  

(2)

Where \( p \) is an effective pressure, and \( \mu \) is the viscosity. Subscribe \( i \) and \( j \) represents \( r \) and \( x \) coordinates. \( S_i \) is source term.

**Energy:**

\[
\frac{\partial (\rho, T_f)}{\partial t} + \frac{\partial (\rho, u_f T_f)}{\partial x} = \frac{\partial}{\partial x} \left( k_f \frac{\partial T_f}{\partial x} \right) + S_f
\]  

(3)

For the numerical solution of the phase change, the enthalpy-porosity model developed by Voller [21], which implemented in FLUENT, is applied. In this model, the entire domain is considered as a porous medium where \( \gamma \) takes the value of 1 in the liquid phase, 0 in the solid phase and \( 0 < \gamma < 1 \) in the mushy region between solid and liquid. When PCM and porous media are mixed as composite materials, the flow pressure loss due to the solid PCM and the porous media can be determined by the momentum equations as equation (2), but the source terms are expressed as:

\[
S_t = - \rho g \beta (T - T_{ref}) + Av \left( \frac{\mu}{\alpha} + C_2 \frac{1}{2} \rho_0^2 |\gamma| \right)
\]  

(4)

\[
S_r = Au \left( \frac{\mu}{\alpha} + C_2 \frac{1}{2} \rho_0^2 |\gamma| \right)
\]  

(5)

Where \( \beta \) is a thermal expansion coefficient, and \( \alpha \) and \( C_2 \) are the permeability and the inertia coefficient of graphite foam, respectively. The function \( A \) is the “porosity function”, is written as [22]

\[
A = \frac{c (1 - \gamma)^2}{\gamma + \omega}
\]  

(6)

Where \( \omega = 0.001 \) is a small computational constant used to avoid division by zero, and \( C \) is a constant reflecting the morphology of the melting front. This constant is a large number, usually \( 10^5 \). The value of \( C = 10^5 \) has been used in the present study.

The equilibrium thermal model is applied to simulate the heat transfer process of PCM in graphite foam, which treats that the liquid PCM and the graphite foam have the same temperature. The energy equation for the device can be described as:

\[
\frac{\partial (\rho, \varphi, h)}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left( k_{eff} \frac{\partial T}{\partial r} \right) + \frac{k_{eff}}{r} \frac{\partial^2 T}{\partial x^2} - \varepsilon^{(d)} \rho L \frac{\partial \varphi}{\partial t}
\]  

(7)
Where $\varepsilon$ is the porosity of graphite foam and $L$ is the latent heat of the PCM, $h$ is the sensible enthalpy and can further be written as:

$$h = h_{ref} + \int_{T_{ref}}^{T} c_{eff,n}dT$$  \hspace{1cm} (8)

The effective thermal conductivity $k_{eff}$ and effective density $\rho_{eff}$ in equation (7) are calculated as the volume average of the conductivities and density of porous material and PCM, respectively, which are denoted by:

$$k_{eff,n} = k_s - (k_s - k_{p,n}) \cdot \varepsilon$$  \hspace{1cm} (9)

$$\rho_{eff,n} = \rho_s(1-\varepsilon) + \rho_{p,n} \cdot \varepsilon$$  \hspace{1cm} (10)

Where $\rho_s$ and $\rho_p$ are densities of graphite foam and PCM, respectively; $k_s$ and $k_p$ are thermal conductivities of graphite foam and PCM, respectively. Subscribe $n$ represents liquid or solid state of PCM.

The initial and boundary conditions are:

The initial temperature of PCM is at melting temperature $T_m$, and the HTF enters the inner tube at a constant temperature which is a little higher than $T_m$ and causes melting of the PCM in the graphite foam. So the velocity of HTF is specified at the inlet, and pressure outlet condition is set at the outlet. The outer tube is insulated.

4. Computational methodology

The model for the numerical study is created using pre-processor software GAMBIT 2.3.16. Meshing of the numerical model is generated and the boundary is applied at appropriate surfaces. Equations of the HTF and the PCM, with initial and boundary conditions have been solved as one domain. The computational domain is resolved with a fine unstructured mesh near the wall to resolve the boundary layer and an increasingly coarser mesh in the rest of the domain in order to reduce the computational time. The GAMBIT model is then exported to FLUENT for problem solving. Time step of 0.05s is conducted to provide a stable convergent solution and accuracy of results. The FIRST ORDER UPWIND differencing scheme has been utilized for solving the momentum and energy equations, whereas the PRESTO scheme is adopted for the pressure correction equation. The PISO algorithm has been applied for the velocity-pressure coupling. The under-relaxation factors for the velocity components, pressure correction and thermal energy were 0.5, 0.3 and 1 respectively. The converged results are assumed to be reached when the maximum relative changes of all velocity, temperature and enthalpy values between consecutive iterations are less than $10^{-6}$. Grid-dependence test is carried out by 10000 elements, 18000 elements and 25000 elements and the maximum difference of melt fraction at an identical time is within 0.01% between using 18000 elements and 25000 elements with a time step of 0.05s. Therefore, 18000 elements with a time step of 0.05 s are used in the computations considering both accuracy and computing time are suitable for the accuracy of solution.

5. Numerical model validation

The proposed numerical model has been checked by comparison between numerical results and existing experimental data obtained by [23]. In this experiment, a shell and tube thermal energy storage device, similar to that proposed in the present work as presented in figure 3, with PCM1 and copper foam with porosity of 95% filling the annual space and a HTF flowing through the inner tube, is used.

The thermal properties of the PCM and porous materials are given in table 1. The melting of the PCM1 is studied experimentally for HTF inlet temperatures above the initial temperature of the PCM, $T_{cin} = 343$ K. In this experiment, temperature was measured at representative locations point 2 ($r=24$ mm, $x=215$ mm). Figure 5 shows the comparison of measured and calculated PCM1 temperature at
point 2. As can be seen in figure 5, a good agreement between present predicted and experimental results is observed. Minor discrepancies between predictions and measurements were observed. The reason for this can be ascribed to the measurements uncertainties and model simplifications. Therefore, the present numerical model can be accurately used for the study of the shell-and-tube thermal energy storage device proposed.

![Figure 5. Comparison of experimental and calculated PCM temperature at point 2.](image)

6. Results and discussion

After the validity of the numerical model has been checked, the numerical analysis of the heat transfer during charging of the shell-and-tube thermal energy storage device has been performed. A series of numerical calculations have been carried out in order to analyze the transient heat transfer and the effect of graphite foam on the heat transfer of PCM. Initially, PCMs are solids and their temperatures are assumed to 303 K, PCM is paraffin with the properties presented in table 1, and graphite foam has the porosity of 0.728. Analysis has been performed for various HTF operating conditions: the HTF inlet velocities from 0.35 to 0.5 m/s, and inlet temperatures of the HTF varied from 330 to 345 K.

6.1. Temperature distribution and liquid fraction

In comparison with the device with pure PCM, figure 6 gives the temperature distribution at 500 s, 1500 s and 2500 s for the case of HTF temperature of 345 K and inlet velocity of 0.35 m/s. In the melting process of PCM, Temperature distributions at 500 s, 1500 s and 2500 s in shell and tube thermal storage device with pure PCM and PCM/graphite foam composites are given in figures 6(a) and 6(b), respectively. It can be seen for pure paraffin, isothermals is almost vertical and temperature distribution is not uniform, which is much higher near the inner tube wall than that of outer tube wall, due to its low thermal conductivity leading to slow heat conduction. However, for the PCM/graphite foam composite, its isothermals of top are tilted to the outside wall gradually with the increase of melting time because liquid phase of paraffin of high temperature rises with the melting of paraffin wax and solid phase of paraffin wax of low temperature sinks resulting from the gravity and natural convection, and temperature distribution of phase transition zone tend to be homogeneous with time increase. In the melting process, paraffin of upper and near the inner tube wall melt first and that of lower and away from the inner tube wall melt finally.

Figure 7 shows that liquid fraction of PCMs at 500 s, 1500 s and 2500 s in melting process. It also can be learnt that heat transfer can be greatly enhanced by graphite foam compared with the melting of pure paraffin wax in shell-and-tube thermal energy storage device. Influence of the HTF inlet temperature on the melting time is shown in figure 8. It can be seen that the melting time decreases with the increase of HTF inlet temperature.
Influences of the different HTF inlet velocity on the melting time of the PCM are also presented in figure 8, the melting time decreases with the flow velocity increase when the HTF inlet temperature is constant. when HTF inlet temperature is 330 K, the melting time is 6725, 6484.85, 6283.85 and 6111.7 s corresponding HTF inlet velocity of 0.35, 0.4, 0.45 and 0.5 m/s, respectively, which means the melting time difference between adjacent velocity is 240.15, 201, and 172.15s, while when HTF inlet temperature is 345 K, the melting time is 3112.5, 3011.35, 2927.85 and 2857.3 s for the corresponding flow velocity of 0.35, 0.4, 0.45, 0.5 m/s, and the melting time difference of adjacent velocity is 101.15, 83.5 and 70.55s, so the influence of velocity on melting time decreases gradually with the increase of HTF inlet temperature.
6.2. Influence of the temperature difference on melting of the composites

The temperature difference between HTF inlet temperature and PCM initial temperature, \( \Delta T \), on liquid fraction of PCM is illustrated in figure 9. It can be observed that the liquid fraction of PCM/graphite composite changes over time at different \( \Delta T \), and the bigger the temperature difference causes the faster variation of liquid fraction while HTF inlet velocity is equal to 0.35 m/s. It can also be noted that increasing the temperature difference results in decreasing of the melting time of paraffin/graphite composite in the thermal energy storage device.

Figure 10 shows the transient thermal response of the point \((r=25.25 \text{ mm}, x=157.5 \text{ mm})\) in PCM/graphite composite to the different temperature differences. It is observed that thermal response is identical despite of the temperature different. In each case, the temperature rises quickly at the beginning due to the higher heat transfer temperature between the HTF and PCM/graphite composite, then gradually to a steady-state temperature of 320 K where PCM begins to melt by absorbing heat. After all the PCM become liquid state, the temperatures of the composites begin to rise again by absorbing heat from the HTF, until to the heat balance between the HTF and the composites. However, with the temperature difference decreases, phase change point is lagged and time of phase change is prolonged, so the melting time of phase change is longer. As given in figure 10, for example, time of phase change range is from 625 s to 1750 s when the temperature difference is 42 K. while time difference is 32 K, it’s from 875s to 2695 s, that is 1820s for melting of PCM.

6.3. Influence of HTF inlet velocity on melting of the composites

Figure 11 gives the liquid fraction of PCM changes with time under different flow velocity when HTF inlet temperature is set as 345 K. Results show that the bigger flow velocity is, the faster melting rate of paraffin wax is. But the influence of HTF inlet velocity on liquid fraction is not obvious.

Figure 12 shows temperature changes of five points in the composites under the conditions of HTF velocity of 0.35 m/s and inlet temperature of 345 K. Five points are selected as: point 1 (25.25 mm, 157.5 mm), point 2 (31.5 mm, 157.5 mm), point 3 (19 mm, 157.5 mm), point 4 (25.25 mm, 236.25 mm) and point 5 (25.25 mm, 78.75 mm). It can been seen from figure 12 that the temperature of composites rise and heat stores in the form of sensible heat before the composites reaching the melting point; on the contrary, temperature of composites keep at melting point and heat stores in the form of latent heat; while all PCMs melting completely, it again stores heat by means of sensible heat, so the temperature continue to increase. Results show that temperatures of these points are identical, but temperature of point 4 increases first, then point 3, point 1 and point 2 and finally temperature of point 5 rises. Because point 4 locates in the upper of phase change region and absorbs heat from HTF first. Therefore, it’s proved that paraffin wax of the composite near the top and the inner tube wall melts first, then melting front gradually to other areas.

**Figure 10.** Transient thermal response of a point in PCM/graphite foam composites

**Figure 11.** Influence of inlet velocity on liquid fraction of PCM.
7. Conclusion
Graphite foam are introduced as the porous medium while paraffin wax was introduced as the occupied PCM into the matrix pores. The PCM/graphite foam composite is prepared and introduced into a tube and shell thermal energy storage device, where paraffin/graphite foam composites as the PCMs fill the shell side and water as HTF circulating inside the tube, has been numerical analyzed in this paper. Compared with the results of the pure PCM, the phase change heat transfer can be greatly enhanced by using graphite foam in device. Moreover, several simulation calculations have been conducted in order to study the effects of operating parameters: HTF inlet temperature and velocity on melting process of the composites. It can be concluded that HTF inlet temperature plays a significant role for reducing the melting time and liquid fraction, but the influence of flow velocity on melting process is small although increasing velocity can reduce the melting time.

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Nomenclature

| A  | [-] | Porosity function | Special characters: |
|---|---|---|---|
| C | [-] | Constant | α [m²] | Permeability |
| ε | [J kg⁻¹ K⁻¹] | Specific heat | ρ [kg m⁻³] | Density |
| C₂ | [-] | Inertia coefficient | β [-] | Thermal expansion coefficient |
| g | [m s⁻²] | Gravity acceleration | ε [-] | Porosity of foam material |
| h | [J kg⁻¹] | Sensible enthalpy | γ [-] | Liquid fraction of PCM |
| k | [W (mK)⁻¹] | Thermal conductivity | μ [kg (ms)⁻¹] | Dynamic viscosity |
| L | [J kg⁻¹] | Latent heat of fusion | |
| p | [Pa] | Pressure | eff | Effective expression |
| u | [m s⁻¹] | Velocity in r-direction | | |
| v | [m s⁻¹] | Velocity in x-direction | f | Fluid (Phase change material) |
| t | [s] | Time | in | Inlet |
| T | [K] | Temperature | ref | Reference value |
| Tm | [K] | Melting temperature | s | Graphite foam |
| x | [m] | Cartesian axis direction | |
| r | [m] | Radius | |
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