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Thermal charging performance of enhanced phase change material composites for thermal battery design^{\diamond}



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ABSTRACT

The design of effective thermal batteries with phase change materials (PCM) relies on the addition of materials with high thermal conductivity and specific surface area to increase the rate of thermal charging and discharging. This paper is a comparative study between two such enhancement materials, namely aluminum and graphite foams, saturated with phase change material. The rate of heat charging was measured for samples exposed to a constant heat flux boundary condition. The impact of aluminum foam was investigated as a function of PCM viscosity, heat flux magnitude, and aluminum pore density. The influence of graphite foam was considered by varying the heat flux magnitude and graphite bulk density. By using a standard experimental configuration, thermal charging enhancement of the foams was compared based on four metrics: composite latent heat, thermal conductivity, time to end of melt, and junction temperature between the heater and the sample. To relate all metrics, two dimensionless parameters were defined to compare diffusive transport and energy storage to junction temperature. It was shown that the thermal charging enhancement of graphite foams is superior to that of aluminum foams based on these performance metrics due to high thermal conductivity, low density, and small pore size.

1. Introduction

The use of thermal batteries to store and reuse waste heat has the potential to increase the efficiency of various systems such as cooling and heating in buildings, hot water heating, and electronics cooling [1]. The high storage density, or latent heat of fusion, combined with a nearly isothermal storage process in appropriate temperature ranges supports the use of solid-liquid phase change materials (PCMs) in such storage systems; however, the low thermal conductivity significantly limits the rate of energy storage. Furthermore, since the thermal conductivity of liquid PCM is typically about half that of the solid form, this liquid layer acts as insulation and increases the temperature of the heated surface, therefore decreasing the efficiency of the system.

To enhance the PCM, solutions such as extended surfaces, microencapsulation of the PCM, and introduction of high conductivity foams or nanoparticles within the PCM to increase the thermal conductivity of the composite have been proposed in literature [2,3]. Since extended surfaces are limited by relatively small surface area to volume ratios, microencapsulation is expensive, and nanoparticles introduce instability issues from settling in the liquid phase [4], the insertion of highly conductive porous foams offers an effective and stable enhancement method without the addition of considerable size or weight to the thermal battery. Two common materials used to form conductive foams are metal and graphite. Metal foams have favorable stiffness/ strength properties, high surface area densities, and a continuous interconnected pore structure [2,5]. Aluminum and copper are the most common metals used to create these open-cell porous foams that are then saturated with PCM to form thermal batteries [6,7]. Graphite foam composites can be formed with mesophase foam or expanded natural graphite flake saturated with PCM [8,9]. Mesophase foams have higher strength properties than foams made with expanded natural graphite flake, but they are also more dense.

Compatibility of metal and graphite foams with various PCMs must also be considered when used to enhance the PCM thermal performance. Not all metals are compatible with PCMs, while carbon-based materials possess high corrosive resistance resulting in chemical

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stability when in direct contact with most PCMs [2]. For example, aluminum and graphite are compatible with organic PCMs as suggested by limited corrosion and stable latent heat during cycling studies; however, nickel and stainless steel have stability issues with some organic PCMs [2,10–12]. Similarly, aluminum, copper, and steel corrode when exposed to salt hydrates [2,13,14], though various studies have demonstrated the stability of carbon-based materials with these inorganic PCMs [15–19].

Open-cell metal foams are classified based on pore density and relative density. Pore density (measured as pores per inch, PPI) describes the diameter of each pore. Typical values range from 10 PPI to 40 PPI. As the number of pores per unit length increases, the diameter of each pore decreases and the overall surface area of the foam increases. Relative density (α), defined as the density of the foam divided by the density of the solid parent material, identifies the amount of metal compared to the amount of PCM. Relative densities of most metallic foams vary from 3% to 13%. As the relative density increases, the redistribution of aluminum within the foam ligaments results in an increase in the surface area. Research with metal foams saturated with PCM typically describes the impact of these two parameters by measuring the decrease in melting time of the PCM [6,20] or the decrease in temperature at the junction of the heat sink and the latent heat thermal storage system [21,22]. Metal foams have been found to decrease the melting time of the PCM up to a factor of 10 and reduce the junction temperature by more than 50% [6,7].

The introduction of metal foam may impede natural convection in the liquid phase depending on orientation of the heat source, but the conductive foam network greatly improves the thermal charging as compared to solely relying on natural convection within a pure PCM. However, for foams with large pores, an additional complexity of natural convection within the pore must be considered in the design of the thermal battery. There is an inherent tradeoff between natural convection and conduction as a function of the relative density and pore density of the foam. As the surface area increases to promote conduction within the system, the size of the pore decreases, therefore, limiting the natural convection within the pore.

Lafdi et al. [21,23], Zhao et al. [6], and Tian and Zhao [24] studied PCM/metal foam composites exposed to constant heat flux conditions. The study conducted by Lafdi et al. [21,23] used a PCM with a melting temperature between 26°C and 29°C and a viscosity at 50°C of 0.003 Pa-s. Each aluminum foam was attached to a 0.3 W/cm^2 heated surface with thermal grease. In the work by Zhao et al. [6] and Tian and Zhao [24], the PCM melting temperature ranged between 48 °C and 62 °C, and the copper foams were sintered to the 0.2 W/cm² heated surface. Viscosity was not reported. These studies discuss the impact of pore density and relative density on the junction temperature and melting time or time to steady state. It was found that metal foams with a higher relative density decreased the junction temperature due to greater conduction through the composite. Regarding the impact of pore density on junction temperature, Zhao et al. [6] and Tian and Zhao [24] found that a higher pore density decreased the junction temperature, while Lafdi et al. [21,23] showed it increased the junction temperature. When studying the impact on the melting time, Zhao et al. [6] and Tian and Zhao [24] found that foams with a higher pore density resulted in a greater reduction of melting time, while a higher relative density increased the melting time. Though Lafdi et al. [21,23] do not discuss melting time, it is summarized that higher pore density and higher relative density increased the time to steady state due to inhibited convection within the pores. It is noted that the use of different viscosities, thermal interface materials, and heat fluxes in these studies may lead to the differences in reported results. Overall, these studies show that there can be a complex dependence of the thermal charging performance of PCM/metal foam composites on the geometry, constituent properties, and thermal boundary conditions. Thus, comparisons between reported experiments in the literature must be done carefully.

Thermal charging studies comparing graphite foams and aluminum foams also have been conducted. Chintakrinda et al. [25] studied the impact of mesophase foam and aluminum foam under high heat fluxes. A 9% relative density aluminum foam and 39% relative density graphite foam were compared using heat flux boundary conditions of 5.8 W/cm² and 11.6 W/cm². By comparing the time to full melt, it was found that the graphite foam outperformed the aluminum foam under both heat fluxes. However, the latent heat capacity of the composites was not discussed, which is an important factor in the total energy storage of the battery. The lower latent heat of the high relative density graphite foam reduces the time to full melt since the system is not capable of storing as much energy as the aluminum foam composite. In another study, Zhao and Wu [13] concluded that 5% relative density copper foams outperformed graphite foam made with 3 wt% expanded graphite flake due to the higher conductivity of the copper foam.

Though expanded graphite flake offers an improvement in thermal conductivity of the PCM, by compressing the flake, a significant increase in thermal conductivity can be realized [26,27]. This graphite composite is referred to as compressed expanded natural graphite (CENG) foam. Composites of CENG foam saturated with PCM are generally compared through composite thermal conductivity and latent heat measurements [9,27]. The amount of expanded natural graphite flake used in CENG is measured by bulk density, or the mass of graphite in the sample volume. PCM can be absorbed into the graphite matrix to form a highly conductive composite [27]. When the expanded graphite is compressed, closed pores can be formed that are impenetrable by PCM. Py et al. [9] found that the higher the CENG bulk density, the greater the volume of closed pores.

While latent heat of the PCM decreases with an increase in CENG bulk density due to the addition of graphite as well as air contained within closed pores formed during the compression process [9], the thermal conductivity of the PCM increases [9,12,27]. The thermal conductivity of composites formed with CENG bulk densities greater than 100 kg/m^3 is found to be anisotropic: thermal conductivity of the sample in the compression direction is less than in the direction perpendicular to the compression by as much as a factor of 7 [9]. This arises due to the preferred orientation of the graphite flakes transverse to the compression direction.

In addition to studying the influence of CENG bulk density on the storage properties of the PCM composite, applications such as thermal management of a battery pack and thermal storage in a solar hot water system have been considered. In these applied experiments [27–29], CENG bulk densities of 210 kg/m³, 150 kg/m³, and 70 kg/m³ were studied, respectively. These three experiments discuss thermal charging of the PCM/CENG composites, but comparison of the effectiveness of the CENG is only considered in terms of the thermal conductivity, while no consideration of the varying heat capacity is given.

Although all of these studies agree that the addition of metal and CENG foams improves the rate of thermal charging of PCM, a direct comparison between the experiments in the literature is often difficult. In addition to the use of different heat flux boundary conditions for the thermal charging, various relative or bulk densities, sample geometries, PCM viscosities, and performance metrics are reported on the tested systems. In metal foam studies that compare the impact of pore or relative density, long transients of thermal charging were exhibited due to heat flux boundary conditions less than 0.3 W/cm². However, these studies did not yield information on comparative performance under higher heat fluxes. Additionally, to the best of our knowledge, to date there are no CENG thermal charging studies for varying heat fluxes and bulk densities. Due to the lack of a standardized methodology for testing these thermal storage materials, it would be beneficial to perform comparisons of metal and CENG-based phase change composites using a standard experimental setup and orientation and over the same thermal boundary conditions to better understand the factors that govern thermal charging rates in PCM/foam composites.

In this work we present a comparative study on the thermal

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