



# Sodium acetate trihydrate-chitin nanowhisker nanocomposites with enhanced phase change performance for thermal energy storage

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## ARTICLE INFO

### Keywords:

Chitin nanowhisker  
Latent heat  
Phase change materials  
Sodium acetate trihydrate  
Supercooling  
Thermal conductivity

## ABSTRACT

Inorganic phase change materials possess superior properties such as better thermal conductivity, higher latent heat of fusion, and cheaper price to their organic counterparts. However, their extreme supercooling during freezing and phase segregation during melting have led to challenges in their applications to thermal energy storage. In this paper, a series of sodium acetate trihydrate (SAT) nanocomposites filled with bio-derived chitin nanowhiskers (CNW) as the nucleating agent have been prepared and tested. Experimental results revealed that the bio-derived nanoparticles were an effective and stable nucleating agent to suppress SAT's supercooling. Furthermore, smaller SAT particle size ( $< 53 \mu\text{m}$ ), optimal CNW content (1 wt%) as well as presence of surfactant and thickening agent were advantageous to the phase change performance of SAT-CNW nanocomposites. It must also be noted that the heating temperature during the melting cycle should be limited to  $70 \text{ }^\circ\text{C}$  or below to retain the nanocomposite's capability to efficiently store and release thermal energy. To enhance the nanocomposite's effective thermal conductivity, different loadings of hexagonal boron nitride (hBN) and graphene nanoplatelets (GNP) were added. It was found that 1 wt% of hBN was appropriate to enhance the nanocomposite's heat transfer ability by 35% without compromising its low supercooling degree. The thermal properties of the nanocomposites were also analyzed by differential scanning calorimetry. Thermal analyses indicated that although adding nucleating agent, thickening agent, surfactant, and thermally conductive filler would decrease the heat of fusion of SAT, the nanocomposite's thermal storage ability was superior to other organic phase change materials. Thermal cycling results also revealed the thermal reliability of the SAT-CNW nanocomposites. In short, this work reported an appropriate formulation of SAT nanocomposites filled with bio-derived nucleating agents that exhibited promising phase change performances.

## 1. Introduction

The world's reliance on fossil fuels as the main energy source has had many negative environmental impacts. Each stage of extracting, refining, distributing and consuming fossil fuels has caused many problems not only on human health but also on the entire ecosystem. Water and air pollution, land degradation, as well as political crises due to unbalanced distribution of fossil fuel resources are only a few devastating consequences of extensive consumption of fossil fuels [1]. Since renewable energy can be a potential way to partially solve the problem, a wide search for alternatives to fossil fuels has been started for years and is still continuing [2]. The goal is to minimize the negative impacts of energy consumption on the environment. Solar radiation, with different intensities depending on geographic locations, is readily available over the world. It is applicable for numerous applications such as thermal collectors, which can be used for heating a fluid (e.g., water) for industrial and residential purposes [3,4]. By using a solar heater in a

20-year period, it is possible to eliminate carbon dioxide emission by 50 t. There are two types of thermal collectors: sensible heat collector and latent heat collector. The former works based on the temperature change of a heat transfer fluid (e.g., water), without phase change, as the energy storage medium during charging and discharging. In contrast, latent heat collectors store and release energy when the energy storage medium undergoes a phase transition at a desired temperature. Phase change materials (PCM) can store and release latent heat. For those undergoing solid-liquid phase transition, the energy is stored in them during melting and released while freezing. The latent heat collectors generally can store five to fourteen times more heat per unit volume than sensible heat collectors [5].

Among different types of PCM, organic PCM are more commonly used for thermal energy storage applications. Paraffinic petroleum-based and non-paraffinic vegetable-based materials (e.g., stearic acid and palmitic acid) are two main groups of organic PCM. Paraffinic petroleum-based PCM possess respectable amount of latent heat,

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limited supercooling, low reactivity, as well as good thermal and mechanical stability. However, their drawbacks include lower thermal conductivity, higher flammability, and higher material cost. Recent studies have demonstrated that microcapsules of palmitic acid in a bio-based polymer have comparable thermal properties and thermal stabilities to its paraffinic counterparts and hence can be a good alternative [6].

Inorganic PCM show promising properties such as higher latent heat of fusion, higher thermal conductivity, and lower cost than organic PCM. Examples of inorganic PCM include salts, salt hydrates, and metal alloys. Salt hydrates show corrosiveness when contacting metals. Therefore, their uses with metal containers are not suitable for long-term applications [7]. Supercooling and phase segregation are other disadvantages of salt hydrates. Phase segregation occurs after salt hydrates have undergone repeating melting and freezing cycles, during which their water and salt components dissociate due to their density difference. This leads to an undesired change in the thermophysical properties of salt hydrates and challenges in their crystallization [8]. Supercooling happens when a material stays in its liquid form when its temperature drops to below its freezing temperature. This may be due to the inability of the material to form crystal nuclei. In other scenarios, a salt hydrate may remain in a metastable condition because the crystal nuclei formed are smaller than the critical size required to initiate crystallization [9]. Therefore, additional amount of energy is needed to overcome the energy barrier to release its latent heat of fusion [10]. Unlike salt hydrates, metal and metallic alloy PCM do not suffer from these disadvantages; however, their high melting points have restricted their applications in thermal energy storage [7].

The values of latent heat of fusion of salt hydrates are approximately twice as high as that of organic PCM. Together with their desirable operating temperature ranges, they have drawn much attention by researchers and the industry. In particular, sodium acetate trihydrate (SAT) is an inorganic salt hydrate that is non-toxic, inexpensive, widely available, and thereby has a high potential for thermal energy storage. With its suitable melting point (i.e., 58 °C) and high latent heat of fusion (i.e., 258 J g<sup>-1</sup>), SAT can be a good candidate to be used in solar heating systems [11]. However, it suffers from severe supercooling and phase segregation. It has been shown that this material can be cooled down to -40 °C without freezing [12]. In this context, extensive researches have been conducted to investigate the uses of additives to overcome their problems of phase segregation and supercooling.

Ryu et al. [13] indicated that a super absorbent polymer made from acrylic acid copolymer can effectively prevent the phase segregation of salt hydrates with a high number of water molecules. They show that carboxymethyl cellulose (CMC) could circumvent phase segregation of salt hydrates with a low number of water molecules. Mao et al. [14] compared the performance of CMC, gelatin, and polyacrylamide as a thickening agent for SAT. They revealed that both CMC and gelatin were effective to inhibit phase segregation in SAT, and thereby helped SAT to retain stable phase change performance. Furthermore, polyvinyl alcohol, silica gel, and sepiolite are other candidates that have been used as thickening agents for salt hydrates [15].

Typical strategies to suppress supercooling include the introduction of an external nucleating agent or a “cold finger” in salt hydrates [16]. Nucleating agents are foreign particles or sites that provide surfaces and suppress the energy barrier for crystal nucleation of PCM. There are typically three classes of nucleating agents: isomorphous, isotypic and epitaxial. The first two classes have similar crystal structures and lattice parameters to the PCM. Isotypic nucleating agents also have nearly identical chemical structures to the PCM. In contrast, epitaxial nucleating agents have different crystal structures, lattice parameters, and chemical structures from the PCM. They enhance crystal nucleation by providing a surface for the deposition of the crystal nuclei and suppressing the energy barrier to initiate crystal nucleation [17]. The cold finger method provides a heat sink in the PCM while the substrate is being heated. During the cooling process, this cold region can initiate

crystallization and suppress the supercooling of PCM [18].

Various external nucleating agents have been researched to investigate their effectiveness to suppress the supercooling of SAT. Mao et al. [19] used 6.2 wt% of disodium hydrogen phosphate dodecahydrate as the nucleating agent and 3 wt% expanded graphite as a filler to enhance the PCM's effective thermal conductivity. They managed to decrease the supercooling degree to 2 °C. Other nucleating agents such as sodium sulfate, strontium sulfate, and Borax have also been studied as nucleating agents for different salt hydrates. They were accompanied by a thickening agent to avoid the phase segregation of salt hydrates [20]. Nevertheless, it has been shown that the uses of salts or salt hydrates as nucleating agents may result in deterioration of the PCM's phase change performance after repeating heating-cooling cycles due to their instability. As a result, researchers have tried to use other chemicals with higher stability as the nucleating agent [19]. Hu et al. [21] introduced 5 wt% of aluminum nitride nanoparticles into SAT and suppressed its supercooling. Cui et al. [22] added 0.5 wt% of nano-copper to SAT, which simultaneously prevented the supercooling and enhanced its effective thermal conductivity.

This work investigated the possibility of using a bio-based nanoparticle, chitin nanowhisker (CNW), as a nucleating agent to suppress the supercooling of SAT. Chitin, produced from renewable and bio-based sources such as crustacean shells or biowastes derived from aquatic organisms, is the second most abundant semicrystalline polysaccharide after cellulose [23]. It possesses many advantages over traditional inorganic nanoparticles including high abundance, nontoxicity, biodegradability, low density, and easy modification [24]. Unlike salt hydrates, it shows a great thermal stability under 250 °C [25]. In the form of nanowhiskers, it provides large specific surface area to potentially serve as a nucleating agent for SAT. To the knowledge of the authors, no research paper has studied bio-derived nanoparticles as nucleating agents to suppress supercooling of inorganic PCM. In an effort to enhance the phase change performance of SAT, a series of parametric studies were conducted to investigate the effects of CNW content, SAT particle size, initial heating temperature, surfactant content, as well as thermally conductive filler type and content on the supercooling degree of the SAT-CNW nanocomposites. CMC was used as the thickening agent to eliminate the phase segregation of SAT. The effective thermal conductivity of the samples was also measured to evaluate the heat transfer properties of SAT-CNW nanocomposites, with and without the addition of thermally conductive filler, during the heating and cooling cycles. Multiple measurements of each material composition were made to ensure the proposed formulation can yield reproducible phase change performances.

## 2. Experimental

### 2.1. Materials

Sodium acetate trihydrate (SAT, BioXtra, purity > 99%, Sigma Aldrich) was used as the inorganic PCM. Sodium carboxymethyl cellulose (CMC, average molecular weight of 250,000, Sigma Aldrich) and sodium dodecyl sulfate (SDS, ReagentPlus, purity > 98.5%, Sigma Aldrich) were selected as the thickening agent and the surfactant, respectively. Chitin nanowhisker (CNW, BOCO Technology Inc.), with its physical properties shown in Table 1, was the nucleating agent being

**Table 1**  
Physical properties of chitin nanowhisker (CNW).

Property	Value	Unit
Density	1450	kg m <sup>-3</sup>
Length	200–500	nm
Width	10–20	nm
Specific Surface Area	72	m <sup>2</sup> g <sup>-1</sup>

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