

# Effect of blending of medium-temperature phase change material on the bitumen storage heat

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## ABSTRACT

This study aims to investigate the feasibility of using D-Mannitol as a phase-change material (PCM) to increase the energy storage capacity and improve the thermomechanical characteristics of the modified bitumen. D-mannitol (Dm) was incorporated into Bitumen (Bm) using a high-speed shearing method. The results showed that the integration of PCM enhanced the physical characteristics of the basic bitumen, with the modified bitumen having a melting point close to that of D-mannitol ( $T_m = 164\text{ }^\circ\text{C}$ ). The D-mannitol was found to crystallize during cooling, indicating that it can store or release heat in the latent form. The specific heat capacity of the modified bitumen was proven to increase gradually as the PCM content (2, 4, and 8 wt%) increased. The maximum temperature regulation effect of the modified bitumen was observed when the PCM content reached 8 wt%, resulting in a temperature reduction of up to  $9\text{ }^\circ\text{C}$ . This study provides a basis for the utilization of D-Mannitol as a form of energy storage in bitumen-based pavement applications.

## 1. Introduction

Bitumen's thermoplasticity, viscoelasticity, and adhesiveness make it a perfect binder for asphalt mixes, used worldwide to build roads and other infrastructure. It is solid at ambient temperature and requires elevated temperatures during mixing and paving processes [1,2]. Furthermore, to ensure that bitumen is in a workable state, it must be stored at high temperatures exceeding  $150\text{ }^\circ\text{C}$  [3]. So, bitumen can be stored at elevated temperatures for an extended period of time exceeding one week. This prolonged storage duration leads to increased fuel consumption and consequent greenhouse gas emissions.

The reduction of the energy consumption and environmental impact of bitumen-based materials has been a subject of interest among researchers. For example, Amina et al. [4] explored the storage of bitumen at ambient temperatures. Despite its cost-effectiveness, this method is impractical since reheating bitumen becomes extremely difficult and time-consuming. Chemical or physical additions have also been studied to improve Bitumen's properties and lower its application temperature. Li et al. [5] explored the viability of incorporating liquid isocyanate-based additives in the process of bitumen modification. The utilization of isocyanate-modified asphalt mixture was observed to reduce energy consumption, owing to the comparatively low temperatures required for asphalt mixing and mixture manufacturing. However, the potential environmental impact of isocyanate-modified bitumen must be investigated, due to the toxicity of isocyanate. On the other hand, although the addition of physical agents has been shown to

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lower the mixing temperatures of modified bitumen, it cannot effectively lower the mixing temperatures of modified bitumen [6].

Furthermore, in order to minimize fuel energy consumption during the bitumen preparation process, researchers have investigated renewable energy sources such as solar energy as a substitute for conventional fossil energy [7,8]. Solar energy is abundant, renewable, and eco-friendly. In a recent study, Bouya and al [9]. presented a hybrid system for maintaining the optimal temperature of 150 °C required for bitumen storage. In cases where solar energy proves inadequate, supplementary fuel energy is employed to meet the energy demands. Although promising, the findings of this work remain relatively costly, and the significant fluctuations in temperature, especially at night, lead to storage temperature inconsistency.

The present study highlights the increasing interest in exploring Latent heat storage (LHS) as an alternative method to mitigate the energy consumption and environmental consequences of bitumen-based materials. LHS has emerged as the most efficient method for storing thermal energy in recent years, due to its ability to provide significantly higher storage density while maintaining a smaller temperature differential between the storage and release of heat [10]. The utilization of heat during phase changes, particularly solid-liquid phase transitions, is a common practice in LHS. Phase change materials (PCMs) are commonly utilized as materials for LHS due to their ability to stably store a significant amount of energy [11]. Currently, there is ongoing research on numerous PCMs for various applications in the fields of construction, industry, and solar energy conversion. PCMs can be organic [12] or inorganic [13] and have different advantages, such as maintaining a desired temperature for a longer period. The characteristics of organic PCMs have been found to include low levels of corrosion, absence of phase separation, minimal supercooling, and non-toxicity. Sugar-alcohol phase change materials have been identified as promising options for heat storage at medium temperatures (90–200 °C) due to their high melting enthalpy. Only sugar alcohols are considered suitable for this purpose [14].

The objective of the current research is to investigate the potential of utilizing D-Mannitol as PCM to enhance the energy storage capacity and improve the thermomechanical characteristics of the modified bitumen through thermal and physical analyses. Mannitol was selected as the ideal substance for the study due to its high melting point of approximately 167 °C, significant latent heat, cost-effectiveness, and non-volatile nature [15,16]. Mannitol's high enthalpy of melting is reported to be 344 J/g, and its phase change temperature is considered appropriate at 166 °C. Mannitol/Bitumen blending approach presents a novel concept for decreasing the fuel energy consumption associated with bitumen storage applications.

## 2. Materials and methods

### 2.1. Materials

Bitumen with 35/50 paving grade was obtained from Bituma Company, and D-mannitol with a purity of 99 % was purchased from Carlo Erba Reagents as the high-temperature PCM. D-mannitol was chosen because of its melting temperature range of 167 °C, making it suitable for high-temperature energy storage applications in bitumen.

### 2.2. Blends preparation

The blend of bitumen and D-mannitol (BmDm) was prepared by high-speed shearing. The pure bitumen was preheated to 160 °C, then D-mannitol was added progressively under high shear at 1000 tour/min for 5 min (160°C–165 °C) and heated up to 170 °C under high shear for ~20 min. The blends of bitumen and D-mannitol with different content of bitumen (2, 4, and 8 wt%), were obtained.

## 3. 3 Characterization methods

**Differential scanning calorimetry analysis (DSC):** The thermal analysis of PCM-modified and unmodified bitumen was determined using DSC (TA Instrument Q100), where the heat flow versus temperature was measured for the second heating cycle at a heating rate of 10 °C/min. The thermal transitions were investigated using samples of 7–10 mg under a nitrogen flow of 100 mL/min. The cooling and heating ramps were conducted at a rate of 5 °C/min at a temperature range of 40 °C–200 °C. The test was conducted in



Fig. 1. Temperature regulation test of bitumen and modified bitumen.

accordance with ASTM D3418 and ASTM E1356 standards.

The specific heat capacity of the bitumen samples was measured using the DSC results. The DSC method measures the heat absorbed or released by a substance as it is heated or cooled, and from these measurements, the specific heat capacity of the substance can be calculated.

**Attenuated Total Reflectance Fourier Transform Infrared Spectroscopy (ATR-FTIR):** The chemical analysis of the modified and unmodified bitumen was determined using ATR-FTIR at 23 °C on a PerkinElmer Two. The collected spectra were scanned for 32 consecutive repetitions with three replications, over 400–4000  $\text{cm}^{-1}$  range with a resolution of 4  $\text{cm}^{-1}$ .

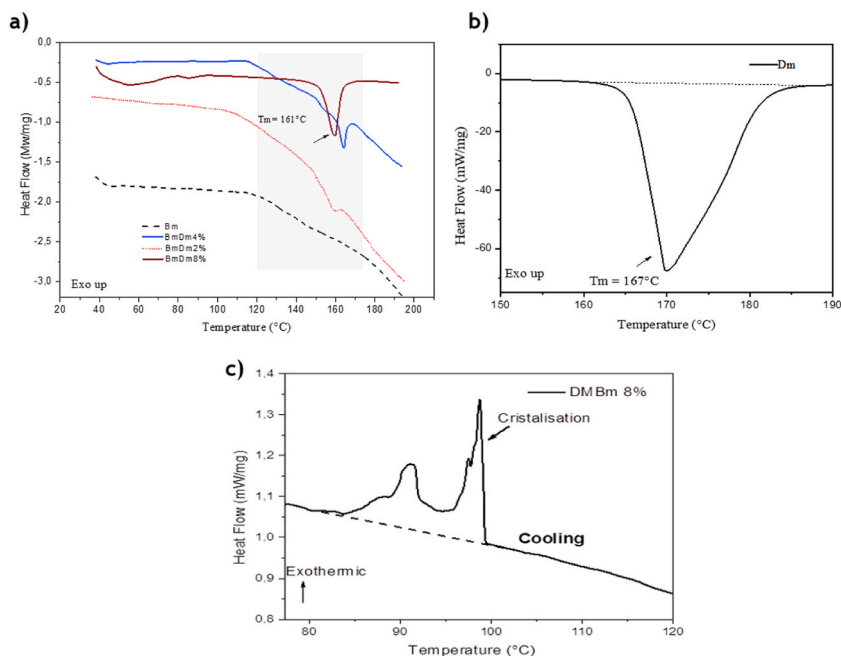
**Test of physical properties:** The physical properties of the modified and unmodified bitumen were tested according to European standards. The penetration at 25 °C, softening point, and ductility at 10 °C were tested according to NM EN 1426, NM EN 1427, and NF EN 13589 respectively.

**Temperature regulation test:** About 200 g of bitumen samples were prepared in aluminum crucibles (diameter: 70 mm; height: 50 mm). The samples were placed in an environmental test chamber and stored at 23 °C for 12 h. A calibrated Pt100 thermistor sensor was then inserted into the center of the bitumen sample (Fig. 1). The temperature of the test chamber was first adjusted to 170 °C and when the samples reached that temperature, it was then adjusted to 80 °C. The temperature changes of the bitumen samples were then recorded during the heating and cooling processes.

## 4. Results and discussion

### 4.1. Dynamic scanning calorimeter (DSC) results

The DSC analysis was used to identify the phase change performance of PCM in the blend of bitumen and D-mannitol (BmDm). The thermal behavior of pure bitumen (Bm), D-mannitol (Dm), and their blend with 2, 4, and 8 wt% was compared and the results are shown in Fig. 2 and summarized in Table 1. The modified bitumen showed a transition around 162–164 °C indicating the melting of the Dm during the heating process. The melting phase-transition temperature of pure Dm was 167 °C which decreased slightly from to 162 °C after blending it with bitumen. This result indicates that the addition of Dm in bitumen impacts its thermal behavior, which can be explained by the high-shear process affecting the crystallization of Dm. Except for the sample with 8 wt% Dm, the samples with 2 and 4 wt% didn't show any crystallization peak, therefore, only the sample with 8 wt% PCM showed a solidification peak at 80–100 °C around the crystallization temperature ( $T_c$ ) of the pure Dm ( $\sim 115$  °C) [17], which indicates that the Dm crystallizes during the cooling process, thus, it can store or release heat in the latent form. Therefore, Dm can be used as PCM in bitumen. However, the decreased crystallization temperature of Dm ( $T_c$  of pure Dm is 115 °C) indicates a leakage or loss of the PCM [17].



**Fig. 2.** DSC thermograms: (a) heating cycle of pure bitumen (Bm), bitumen/D-mannitol (BmDm) blends. (b) heating cycle of neat D-mannitol (Dm), and c) cooling cycle of BmDm8%.

**Table 1**  
Thermals characteristics of different bitumen samples.

Samples	Melting temperature [°C]	Latent heat of fusion [J/g]
DmBm2%	161	2.76
DmBm4%	164	5
DmBm8%	160	6.34
Bm	–	–
Dm	167	285

<sup>a</sup> Measured using DSC.

#### 4.2. Specific heat capacity of PCM-modified bitumen

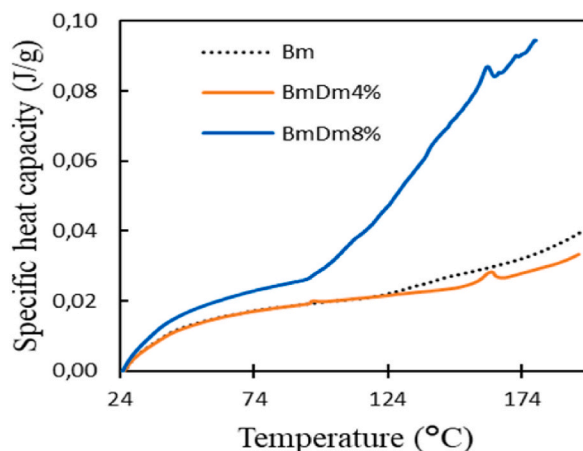
The specific heat capacity as a thermophysical parameter is very significant for the temperature control of phase change-modified bitumen. Fig. 3 shows the specific heat capacity (Cp) curves calculated at the onset of melting point obtained from the DSC test. As seen in the figure, the specific heat capacity of the unmodified and modified bitumen increases with increasing temperature. The specific heat capacity curve of BmDm (with 4 wt% Dm) coincided with the unmodified bitumen curve. However, with increasing the content of PCM from 4 % to 8 wt%, the heat-specific capacity of modified bitumen increases from 0.02 to 0.09 J/g°C with increasing temperature. Additionally, two small peaks in the curves of modified bitumen were observed at 160–162 °C. The heat capacity curves reflect the phase change temperature, which explains the observed peaks. PCM absorbed a large amount of latent heat, which caused a gradual increase of the specific heat capacity [10]. This is coherent with the DSC result during the heating process (Fig. 2) and indicates that Dm PCM may enhance the temperature control property of the bitumen binder.

#### 4.3. ATR-FTIR analysis result

The ATR-FTIR analysis was performed to investigate the chemical interactions between the bitumen and Dm. Fig. 4 shows the ATR-FTIR spectra of unmodified and Dm-modified bitumen samples. After incorporation of Dm in bitumen the absorption peak observed at 3320 cm<sup>-1</sup> represents the stretching vibration of the –OH functional group of Dm, and those at 2800–2950 cm<sup>-1</sup> are characteristic of the –CH group. The peak at 1028 cm<sup>-1</sup> represents the C–OH vibration group. With an increase in the amount of Dm from 2 % to 8 % in the blended bitumen, the absorption peak intensity of the hydroxyl group of Dm around 3320 cm<sup>-1</sup> and 1028 cm<sup>-1</sup> increased. Thus, by comparison with pure bitumen the characteristic peaks of the samples have not significant difference. This proves that there is no chemical interaction between Dm and bitumen.

#### 4.4. Physical properties of the modified bitumen

The physical properties of the modified and unmodified bitumen were tested, and the results are presented in Fig. 5a–c. The physical properties of the Bm/Dm blends with different amounts of PCM are better compared to pure bitumen. The addition of only 2 wt% of PCM to the bitumen improved its physical properties as seen in Fig. 5 for instance, the penetration ... decreases, the softening point increases, and the ductility decreases slightly but remains superior to 80 cm, which is the specification value of pure bitumen. With an increase in the amount of PCM in the blend from 4 % to 8 wt%, the penetrability, the softening point, and the ductility were also improved, however, remained practically comparable to the sample with 2 wt% PCM. These results are very interesting as they highlight the potential use of different contents of PCM without impacting the physical properties of bitumen. Additionally, this is



**Fig. 3.** Specific heat capacity curves of unmodified and Dm-modified bitumen samples.

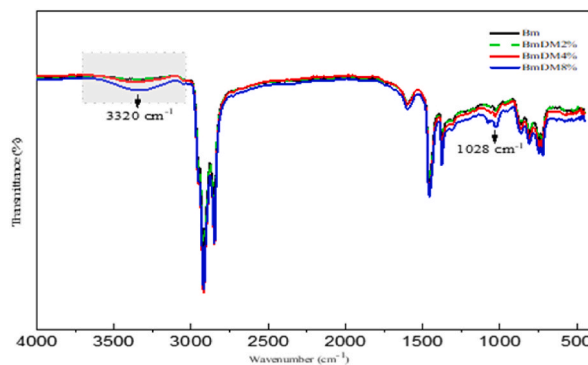


Fig. 4. ATR-FTIR spectra of bitumen samples.

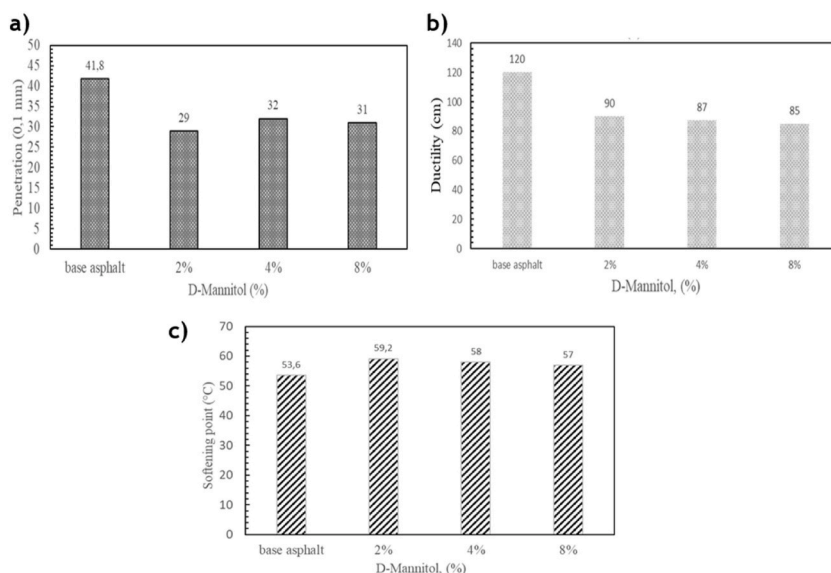


Fig. 5. Effect of PCM modification on penetration (a) ductility (b) and softening point (c) of the pure bitumen (base asphalt) and their blend with Dm.

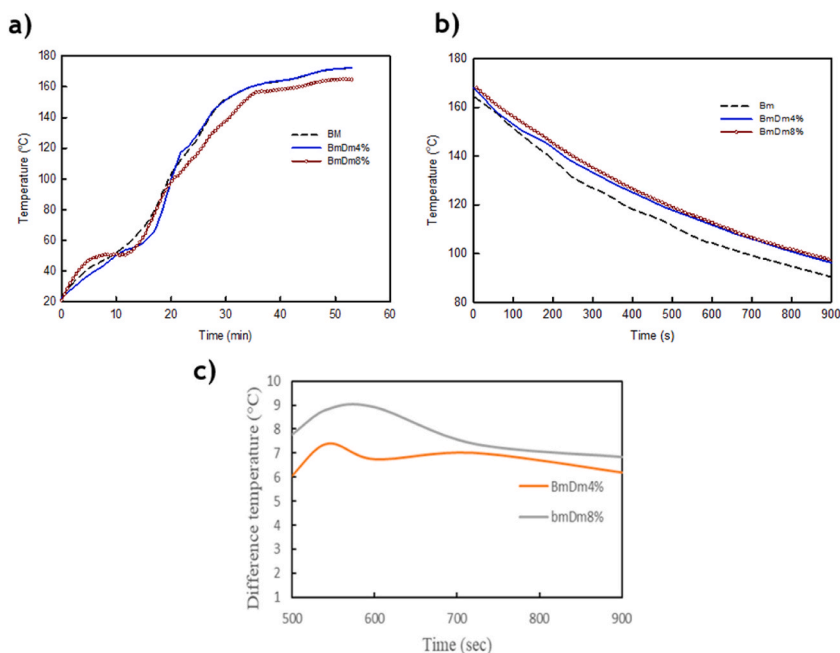
beneficial from an economic point of view as only small concentrations of Dm can be used to reach the desired properties.

#### 4.5. Temperature regulation test of PCM-modified bitumen during the cooling process

The temperature regulation test was employed to study the temperature effect of Dm on the liquid-solid transition of PCM-modified bitumen during the cooling and heating processes. As shown in Fig. 6a, the temperature of pure bitumen and Dm-modified bitumen rises gradually. Nevertheless, the pure bitumen heats faster than the modified bitumen, and the Dm concentration influences significantly this heating rate. For the sample with 4 wt% Dm, the trend of curves practically coincides with that of pure bitumen. However, for sample with 8 wt% Dm, a platform appears in the heating ramp graph (Fig. 6a) when the temperature reaches the melting point of Dm (about 162 °C), which means the phase transition and thermal energy storage processes. Subsequently, the cooling temperature curves (Fig. 6b) show a gradual drop in the temperature over time. for sample with 8 wt% Dm, there is a new inflexion appears near the solidification point of Dm from 110 °C, at this time the modified bitumen with PCM releases heat in the form of both sensible heat and latent heat. This result confirms that the latent energy absorbed under the melting of Dm is released. Therefore, pure bitumen releases heat only in the form of sensible heat. Since the cooling rate of the bitumen is faster than the cooling rate of the samples, so for temperature difference curves (Fig. 6c), with the increase of PCM from 4 to 8 wt% the temperature difference reaches 9 °C. This means that a large amount of stored heat has been released and it can be used for energy storage applications.

## 5. Conclusions and recommendations

In summary, this study investigated the use of a phase change material (Dm) to modify bitumen grade (35/50) in order to store



**Fig. 6.** (a) Warming temperature, (b) cooling temperature, and (c) cooling temperature difference curves of modified and pure bitumen samples.

thermal energy and slow down the cooling rate of the bitumen. The impact of the presence of Dm was observed only on the physical modification of the bitumen, without any chemical interaction, as demonstrated by ATR-FTIR. Results from physical characterizations and DSC analysis show that Dm can be used as a solid-liquid medium PCM for thermal energy storage in latent form without affecting the performance of the bitumen. The melting point of the modified bitumen was observed to be around that of pure Dm ( $T_m = 167^\circ\text{C}$ ), and the Dm crystallization during cooling indicates that it can store or release heat in latent form. It was also observed that the heating and cooling rate of the samples was influenced by the PCM content. The cooling temperature difference reached  $9^\circ\text{C}$  for the PCM-modified bitumen with 8 wt%. This result is significant for medium-temperature storage of bitumen applications in paving. The use of higher concentrations of PCM and encapsulation to prevent leakage is recommended for future studies.

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## CRedit authorship contribution statement

**Loubna Najemi:** Writing – original draft, Supervision, Investigation, Writing – original draft, Supervision, Investigation. **Imane Belyamani:** Writing – original draft. **Mohsine Bouya:** Supervision, Resources.

## Declaration of Competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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