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Review-Paper: Sugar alcohol based phase change materials: progress and challenges in medium to high temperature heat storage

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***Abstract* -** The present study analyses the potential of sugar alcohols as latent heat storage materials for applications in the medium to high temperature range (60-200 °C). In view of the increasing share of volatile renewable energies, thermal energy storage, especially on the basis of phase change materials (PCM), represents a promising solution. Sugar alcohols, including erythritol, xylitol, D-sorbitol and D-mannitol, are distinguished by their high melting enthalpies, optimal melting temperatures and advantageous polymorphic properties. The study systematically compares the thermophysical properties of the materials under varying heating rates and analyses eutectic mixtures to optimise melting behaviour and undercooling. Furthermore, the crystallisation triggering mechanisms (e.g. air injection, ultrasound, additives) and the rheological properties in the supercooled state are investigated. The findings demonstrate that the utilisation of appropriate mixtures facilitates stable supercooling and high cycle stability over an extended period, which is a pivotal consideration for seasonal heat storage. The combination of targeted sample preparation, mixing strategy and rheological control opens up new perspectives for sustainable thermal energy storage systems.

***Keywords*:** Sugar alcohols, Phase Change Materials (PCM), Subcooling, Rheological properties.

1. Introduction

The global energy systems are to be reorganised through the adoption of renewable energy sources, with a particular emphasis on solar and wind power. However, it should be noted that these sources are inherently volatile. Solar energy is contingent upon the diurnal cycle, and wind power is subject to fluctuations in weather conditions [1]. In order to guarantee a stable and continuous energy supply, robust short- and long-term energy storage solutions are essential, especially in view of the expectation that renewables will constitute up to 50% of the global energy mix by 2030 [3].

It is evident that the storage of thermal energy – particularly that of the latent heat – is of paramount importance if this challenge is to be overcome. In contradistinction to sensible heat storage, which is contingent on temperature fluctuations in a storage medium, latent heat storage employs phase transitions, thereby facilitating more efficient, compact, and stable energy retention [4, 5]. Sugar alcohols are regarded as being amongst the most promising materials for such systems, due to their favourable melting points (up to 200°C) and their effectiveness in waste heat recovery [8, 9].

It is estimated that approximately 80% of the world's waste heat is produced in the low to medium temperature range (below 300°C), with 63% of this being below 100°C. However, the process of converting heat to electricity is rendered inefficient by thermodynamic limitations (Carnot's theorem) [2]. A more efficient approach would be to directly store and reuse this thermal energy in applications such as residential heating, industrial processes and hot water supply.

The heat demand at low to medium temperatures in Germany alone exceeds 800 TWh [6], while more than 171 TWh of waste heat from large industrial consumers remains unused [7]. The utilisation of renewable heat has remained static in recent years, and there remains a continued reliance on fossil fuels in high-temperature applications. Consequently, the effective reuse of heat and the development of advanced thermal storage represent an immediate path towards greater sustainability.

2.Theoretical fundamentals

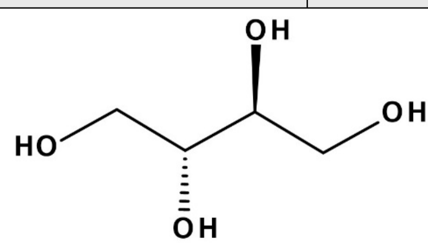
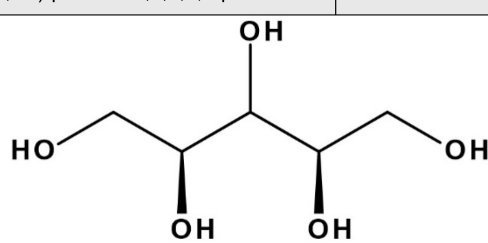
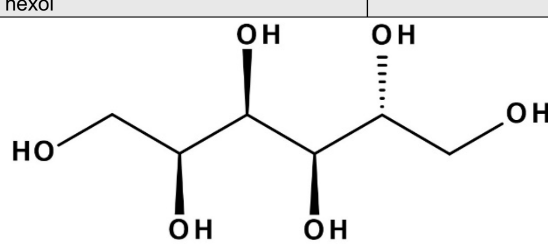
2.1. Sugar alcohols

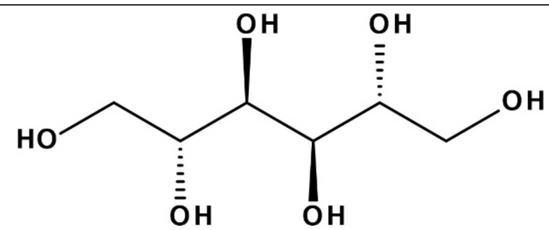
Sugar alcohols, otherwise known as polyols, are defined as organic carbon compounds. These compounds belong to the class of hydrogenated carbohydrates. Distinctions between them can be made primarily based on position and spatial orientation of the hydroxyl groups, in addition to the length of the respective carbon chains. These factors greatly influence the molecular geometry and physical properties of the compounds, such as their melting temperature and enthalpy. In contrast to non-polar alkanes, high melting and enthalpy temperatures are not directly proportional to carbon chain length [10, 11].

Erythritol ($C_4H_{10}O_4$), despite having a lower number of carbon atoms, has a higher melting point and enthalpy than D-sorbitol ($C_6H_{14}O_6$) (Table 1), due to its symmetrical molecular structure, which enhances crystal stability. It is evident that both xylitol ($C_5H_{12}O_5$) and D-sorbitol are known to vitrify upon cooling, whereas erythritol and D-mannitol ($C_6H_{14}O_6$) crystallise, although the latter shows lower stability [10-14].

These sugar alcohols are valued for their phase change behaviours, which renders them as potential candidates for thermal energy storage. From a commercial standpoint, erythritol is produced through a fermentation process [23], whereas xylitol is derived from biomass hydrolysis [25]. Additionally, both D-sorbitol and D-mannitol are synthesised via a catalytic hydrogenation of glucose process [26, 28]. The range of applications extends from sweeteners to pharmaceutical excipients and energy materials, with their thermal and polymorphic properties being of critical importance for industrial utilisation.

Table 1: Comparison of the sugar alcohols erythritol, xylitol, D-sorbitol and D-mannitol.

Sugar alcohol	T_m (°C)	ΔH_m (kJ/kg)	IUPAC	Chemical Formula	Molar Mass (g/mol)	Reference	Heating Rate (K/min)
Erythritol			(2R,3S)-Butane-1,2,3,4-tetrol	$C_4H_{10}O_4$	122,12		
	118,8	325,4				[15]	0,5
	117,8	322,6				[13]	1
	118,3	327,3				[15]	1
	118,7	333,7				[12]	1
	118,1	318,5				[16]	2
	118	315				[14]	5
	118,9	332,3				[15]	5
	119,4	338,7				[17]	10
Xylitol			(2S,4R)-pentane-1,2,3,4,5-pentol	$C_5H_{12}O_5$	152,15		
	92,7	240,1				[12]	1
	93,3	231,4				[15]	5
	93,4	237,5				[15]	5
	93	263,03				[18]	10
	95,1	251,4				[17]	10
D-Sorbitol			(2S,3R,4R,5R)-Hexane-1,2,3,4,5,6-hexol	$C_6H_{14}O_6$	182,17		
	93,2	153				[12]	1
	93,4	165,8				[13]	1
	97	170,7				[19]	2
	101,7	173,4				[20]	2
	96,8	217				[21]	3,5
	97,4	164				[15]	5
	99,4	184,4				[15]	5
	99,2	168,3				[22]	10
D-Mannitol			(2R,3R,4R,5R)-Hexane-1,2,3,4,5,6-hexol	$C_6H_{14}O_6$	182,17		

	166,2	278,6			[15]	0,5
	166	308			[13]	1
	166,3	278,7			[15]	1
	164,4	273,4			[19]	2
	169	325,5			[20]	2
	165	338			[21]	3,5
	166,9	296,1			[17]	10

2.2. Melting temperatures and melting enthalpies

The melting behavior of sugar alcohols varies with heating rates and structural properties (Figures 1-4). While higher heating rates typically raise melting temperatures and enthalpies, this trend is not uniform, especially for polymorphic alcohols like D-sorbitol and D-mannitol [24].

In contrast, erythritol and xylitol do not exhibit polymorphism, which is why other factors such as the purity of the starting materials and the moisture content play a crucial role. The influence of bound water on the enthalpies of fusion is a primary concern, as it necessitates the allocation of additional energy for the process of evaporation [29].

Supercooling poses challenges for their use as phase change materials (PCMs) in thermal storage, as high viscosity at low temperatures hinders crystallization. However, combining sugar alcohols with varying crystallization behaviours—like stabilizing spontaneous crystallizers (e.g., erythritol) with vitrifying ones (e.g., xylitol)—may help maintain metastable melts while enabling targeted crystallization. Crystallization can be initiated thermally, electrically, or mechanically [27, 30].

It is evident that the sugar alcohols deemed to be appropriate for utilisation as phase-change materials (PCMs). However, it is imperative to consider parameters such as humidity, heating, and cooling rates during the implementation stage.

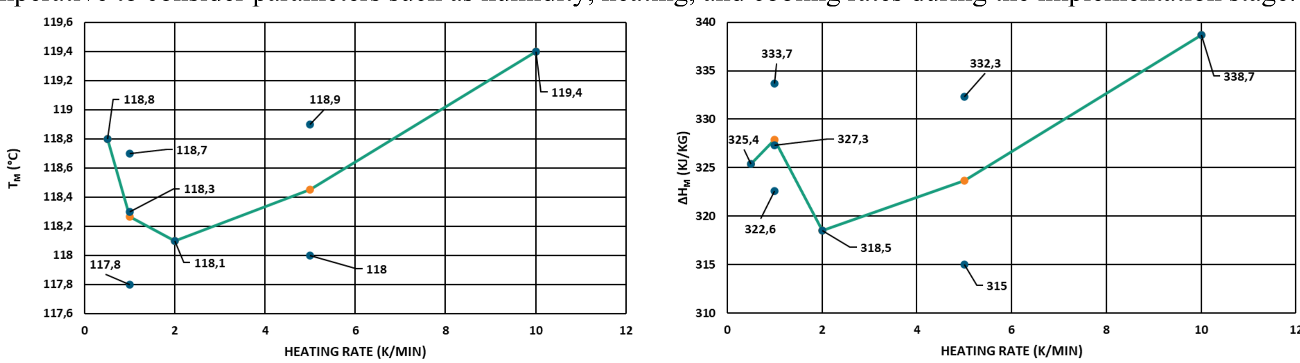


Figure 1: T_m (°C) and ΔH_m (kJ/kg) per Heating Rate (K/min) Erythritol [12-17].

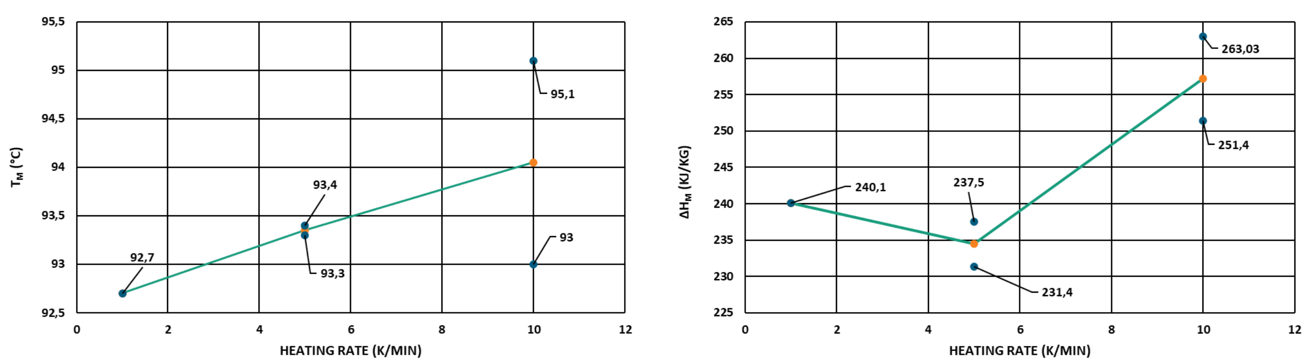


Figure 2: T_m (°C) and ΔH_m (kJ/kg) per Heating Rate (K/min) Xylitol [12-17].

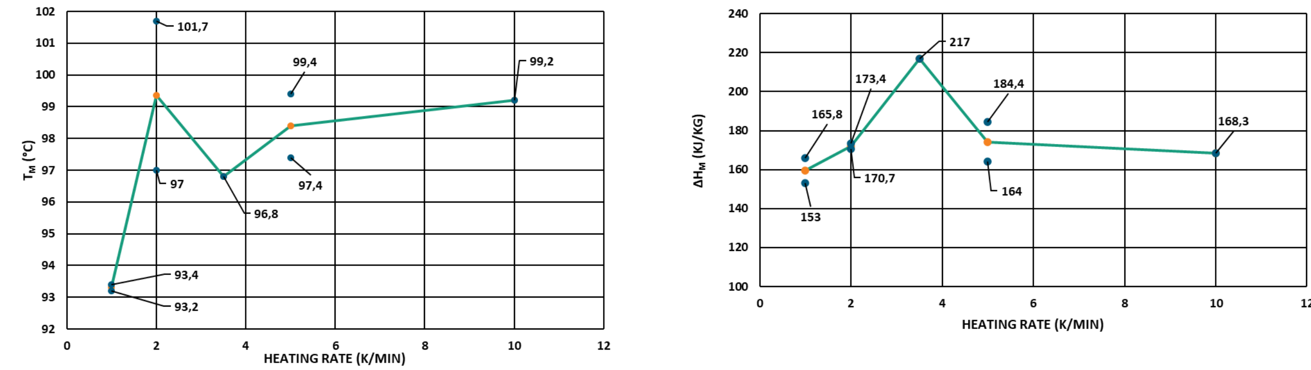


Figure 3: Tm (°C) and ΔH_m (kJ/kg) per Heating Rate (K/min) D-Sorbitol [12, 13, 15, 19–22]

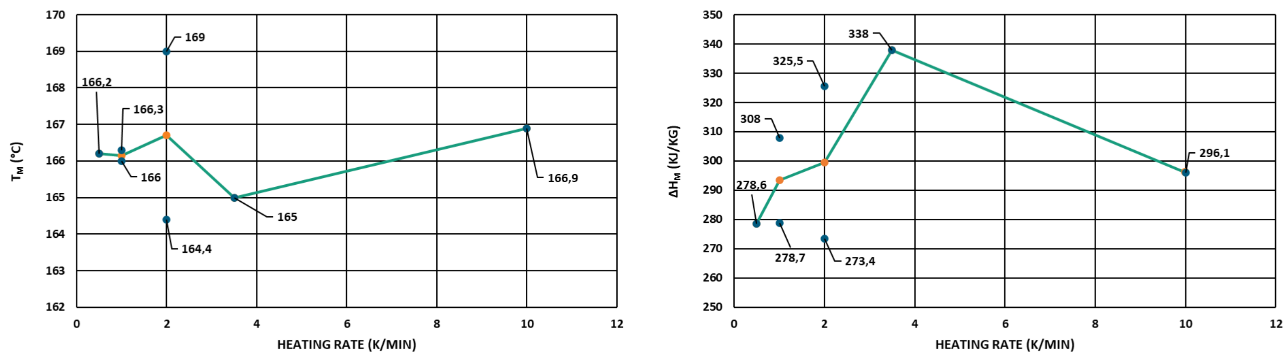


Figure 4: T_m (°C) and ΔH_m (kJ/kg) per Heating Rate (K/min) D-Mannitol [13, 15, 17, 19–21]

3. Experimental

3.1. Sample Preparation and Composition Strategies for Sugar Alcohol Mixtures

The meticulous preparation of samples constitutes a pivotal stage in the examination of sugar alcohols as phase change materials (PCM), particularly when these substances are present in crystalline or powder form. The grinding process, which is typically conducted using a ball mill, is of paramount importance for the homogenisation and reduction of the particle size [31, 32]. This results in an augmented specific surface area, thereby significantly enhancing heat transfer during thermal analysis, for instance by differential scanning calorimetry (DSC) [33]. In the course of the study, the drying process (for example, 3 hours at 60°C) was integrated prior to grinding with the objective of removing moisture and enhancing the stability of the samples [34].

Sugar alcohols are typically selected and combined in the form of mono, binary or tertiary mixtures, with each stage offering specific advantages in terms of melting behaviour and energy efficiency. Monomixtures, such as xylitol, are distinguished by their high latent heat storage capacity and are well-suited for seasonal heat storage, due to their stable melting temperatures. However, in the case of many monocomponents, the targeted release of the stored heat through spontaneous crystallisation is more difficult [33, 35].

Consequently, eutectic binary mixtures are gaining prominence due to their ability to significantly reduce the melting temperature without substantially compromising the storage capacity. A notable example is the combination of xylitol and erythritol, which at approximately 83 °C falls within the optimal range for building heating systems. These mixtures are distinguished by their capacity to enable a homogeneous phase transition temperature, in addition to a substantial latent heat storage capacity and stable subcooling [31-33].

Tertiary mixtures extend these concepts by offering even greater flexibility in thermal adaptation. Systems composed of xylitol, sorbitol and mannitol (refer to figure 5) have been shown to exhibit a melting range of between 60°C and 90°C, a property that renders them particularly well-suited for application-specific storage solutions. It is evident that, owing to their composition, individual thermal properties such as melting enthalpy, undercooling behaviour and crystallisation dynamics can be meticulously calibrated. The effective combination of suitable mixing ratios with careful sample preparation through drying, grinding and homogenisation thus forms the basis for the development of high-performance, customisable PCM systems for seasonal or short-term thermal energy storage [34].

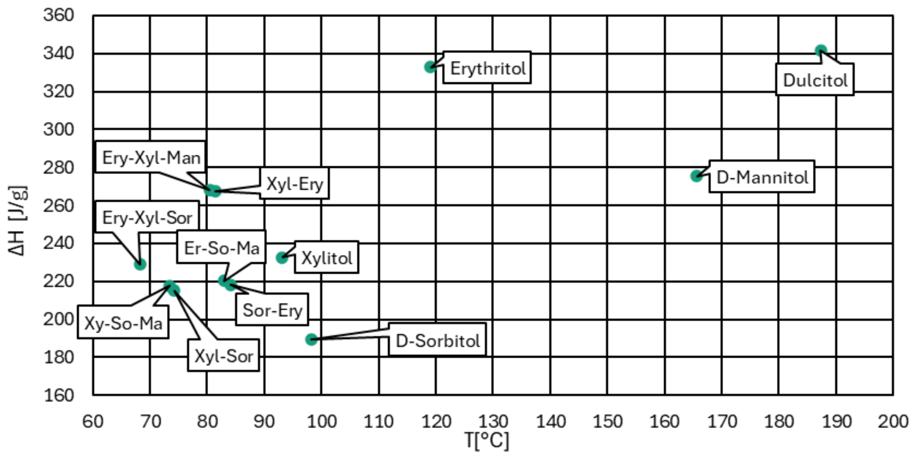


Figure 5: The melting temperature and melting enthalpy of the individual materials and eutectic mixtures.

3.2. Triggering mechanisms

The controlled triggering of crystallisation is a central aspect in the application of supercooled sugar alcohols as PCM. Mechanical stirring is a widely utilised technique for stimulating nucleation by forced convection [34, 39]. However, this process frequently results in localised crystallisation [35]. A more efficient alternative is air injection, which achieves more uniform crystallisation through the homogeneous distribution of air bubbles [35]. The combination of seeding and stirring has been demonstrated to be particularly efficacious [31, 40]: This process involves the strategic introduction of crystal nuclei, which facilitate expeditious and replicable crystallisation. Other methods include the addition of additives [18, 41] (e.g. ethanol, methanol) to promote nucleation and ultrasonic treatment [31], which generates additional crystallisation centres through high-frequency vibrations.

Each technique has specific advantages and disadvantages in terms of homogeneity, energy efficiency and technical feasibility. In industrial contexts, the selection of methodology is contingent upon process control, scalability and economic imperatives. It is noteworthy that the integration of seeding with shear forces has emerged as a particularly salient avenue for the targeted regulation of crystallisation processes.

3.3. Cycle and long-Term Stability of Supercooled Sugar Alcohol Melts

A pivotal consideration in the development of efficient thermal energy storage (TES) systems pertains to the cycle and long-term stability of supercooled phase change materials (PCMs). Research conducted on D-mannitol and related polyols has demonstrated that recurrent cycles of melting and crystallisation can result in degradation, particularly when exposed to oxygen. This phenomenon is characterised by a substantial decline in the melt enthalpy. The utilisation of inert gases, such as nitrogen, has been demonstrated to reduce oxidative decomposition and enhance stability, despite the occurrence of specific degradation processes, including the formation of 2,5-anhydro-D-mannitol [42-45].

Concurrent studies on the long-term stability of eutectic mixtures, such as xylitol/erythritol, have demonstrated that these mixtures can remain in a supercooled state for a minimum of ten days without undergoing crystallisation. The enhanced stability exhibited by the material can be attributed to elevated dynamic viscosity, which curtails molecular movement and consequently impedes spontaneous nucleation. It has been demonstrated that other sugar alcohol-based systems, including D-sorbitol and maltitol, exhibit elevated levels of transparency and structural integrity over extended periods, with no evidence of crystallisation [31, 33].

The capacity of these materials to withstand both repeated thermal cycling and prolonged supercooling is pivotal to their integration into seasonal heat storage applications with stringent requirements for long-term and operational safety.

3.4. Rheological properties

The The rheological properties of sugar alcohols are a pivotal factor in their application as phase change materials (PCM), given that they have a substantial influence on heat transfer, crystallisation kinetics and the control of storage processes.

It has been demonstrated by a considerable number of studies that the viscosity of sugar alcohols is a property contingent on temperature, exerting a considerable influence on the crystallisation kinetics [32, 36, 37]. As the temperature is reduced, the viscosity rises considerably, thereby restricting the mobility of the molecules and consequently slowing down or preventing nucleation and crystal growth [8]. The behaviour of sugar alcohols at varying temperatures can be described by the Arrhenius model, which demonstrates that the viscosity increases exponentially with decreasing temperature [36, 37].

$$\eta(T)_\infty = \eta_0 \exp\left(\frac{E_a}{RT}\right) \tag{1}$$

This results in a substantial impediment to the crystallisation process, as the molecules are required to surmount a greater activation energy in order to rearrange themselves and form a crystal structure [31].

With regard to rheological properties, it was found that sugar alcohols exhibit non-Newtonian flow behaviour under certain conditions, which varies depending on the shear rate. In circumstances where the rate of shear is minimal, sugar alcohols such as xylitol and erythritol exhibit a phenomenon known as shear thinning behaviour. This phenomenon can be attributed to a decline in viscosity that occurs in response to an elevated shear rate. This behaviour, as described by the Ostwald-de Waele model, demonstrates the adaptability of sugar alcohols under various flow conditions and provides insights into the dynamic interactions between the molecules [36].

$$\eta\left(T\right) = \frac{\sigma\left(T\right)}{\gamma} = K(T)\gamma^{n-1} \tag{2}$$

However, at higher shear rates, the viscosity remains constant, indicating Newtonian flow behaviour. These rheological phenomena are of particular importance for the application of sugar alcohols as PCMs, as they influence the flow properties under varying thermal and mechanical conditions [32, 36].

The extant research demonstrates that the rheological properties of sugar alcohols are contingent on their molecular structure. Linear sugar alcohols, for instance mannitol and dulcitol, are characterised by the Herschel-Bulkley model. This model dictates that a yield point, representing the material's liquidity threshold, must be exceeded before flow occurs [36].

$$\sigma = \sigma_y + K\gamma^n \tag{3}$$

These specific rheological properties are crucial for controlling crystallisation processes, as they influence the movement of molecules and the formation of crystal nuclei.

The Cross model provides a comprehensive description of the viscosity curves of sugar alcohols, such as D-dulcitol and inositol, in the supercooled state. It elucidates the transition from Newtonian to non-Newtonian behaviour [36].

$$\eta\left(T\right) = \eta(T)_\infty + \frac{\eta(T)_0 - \eta(T)_\infty}{1 + \left(\lambda\gamma\right)^m} \tag{4}$$

The combination of shear forces and crystallisation has demonstrated that shear forces can significantly accelerate the crystallisation rate. The application of shear forces leads to the erosion and distribution of minute crystal fragments, which function as additional nuclei and thereby stimulate the crystallisation process. Consequently, the efficiency of crystallisation can be specifically controlled by the level and intensity of the shear stress. Furthermore, an elevated proportion of small crystal fragments has been shown to increase the viscosity of the melt, which can subsequently be utilised as a metric for the progression of crystallisation [38]. Viscosity is a crucial indicator of crystallisation, and its influence extends to thermal conductivity and natural convection within the material. Consequently, it is a pivotal factor in determining the performance of sugar alcohols as PCMs.

3. Conclusion

This review has systematically summarised and evaluated the current state of research on sugar alcohols as phase change materials (PCMs) for thermal energy storage in the medium to high temperature range (60-200 °C). Materials such as erythritol, xylitol, D-sorbitol and D-mannitol possess high melting enthalpies and suitable melting points, and are characterised by polymorphic properties that are of central importance for latent heat storage applications. The extant literature demonstrates unequivocally that eutectic mixtures of these substances can be utilised to reduce the melting temperature, regulate the undercooling behaviour and enhance the stability during repeated storage cycles.

Moreover, a plethora of studies have examined the impact of crystallisation triggering mechanisms, including air injection, ultrasound and additives, which facilitate the precise regulation of solidification processes. These strategies offer a promising approach to the controlled release of heat in supercooled systems.

The rheological properties of the sugar alcohols were the primary focus of the study, with particular emphasis placed on the temperature-dependent viscosity. This has been demonstrated to exert a substantial influence on the mobility of the molecules in the supercooled state, consequently affecting the crystallisation kinetics. Despite the pivotal function of these properties in ensuring the practical applicability of the materials, the extant literature contains incomplete data. A paucity of research has been conducted on the rheological parameters of sugar alcohols, which hinders the targeted selection and combination of these substances in binary or tertiary mixtures.

In conclusion, sugar alcohols demonstrate considerable promise in the domain of sustainable heat storage solutions, particularly within the broader context of a progressively renewable energy system. Concomitantly, there is a necessity for additional research, particularly with regard to rheological characterisation, long-term behaviour under practical conditions and technical implementation in scalable storage systems.

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