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Characterization of different sugar alcohols as phase change materials for thermal energy storage applications

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ABSTRACT

Sugar alcohols (SA) are attractive phase change materials (PCM) for thermal energy storage applications at low-to-medium temperatures (70–180 °C). Five pure sugar alcohols (xylitol, adonitol, L-arabitol, erythritol, D-mannitol) and three eutectic blends (eythritol/xylitol, L-arabitol/erythritol, L-arabitol/xylitol) are investigated in this paper. Experimental characterization of such materials as PCMs is provided. This encompasses the measurement of their melting point and latent heat of fusion, as well as the experimental determination of all key physical properties (specific heat, thermal conductivity, thermal diffusivity, density, viscosity) as a function of the temperature. The performances of the studied materials are compared to those of most currently used PCMs (paraffin waxes, salt hydrates etc.) in the field of thermal energy storage. The most significant applications, including solar seasonal energy storage, are also discussed.

1. Introduction

Sugar alcohols (SA), also called hydrogenated carbohydrates and polyols, belong to the family of low molecular weight carbohydrates. More than 900 SA are listed in the dictionary of carbohydrates edited by Collins [1]. However, only few of them are commonly used and produced at a large scale. The most commonly used SA are sorbitol, mannitol, xylitol, lactitol, maltitol, erythritol and isomalt. Some SA are found naturally in various fruits and vegetables. However, most of them are produced by chemical reduction of carbohydrates. The production of polyurethane is the largest and the oldest industrial application of polyols, with a market which is nowadays mature. SA are also widely used in the food industry, mainly as sugar replacers, and in the pharmaceutical sector. Detailed information about SA (production, applications and market) can be found on the web-site of the European Association of Polyol Producers (www.polyols-eu.com) (Tables 1 and 2).

The use of SA as PCMs for thermal energy storage applications was described for the first time in the patents of Guex et al. [2] and Hormansdorfer [3]. They noted that some of the SA have volumetric latent heat as much as twice that of commonly used PCMs (i.e. paraffin waxes). Besides, SA are of natural origin, they are non-flammable, non-toxic and non-corrosive, and they have affordable cost. Since then, different SA have been considered and studied for storage applications at medium temperatures (100–200 °C). Among them, erythritol has received the most attention so far and has been used in various applications such as waste-heat transportation [4,5], solar cookers [6,7], absorption chillers [8], and as an automotive coolant waste heat storage system [9]. It is characterized by a melting temperature around 118 °C and relatively large latent heat of 340 J/g (see Table 3). For applications at higher temperatures, other SA such as D-mannitol, dulcitol, mio-inositol and their mixtures have also been investigated [10–15] and envisaged for thermal energy storage in industrial applications.

More recently, SA have been considered as candidates for latent heat storage applications at temperatures below 100 °C (e.g.: solar heating and DHW, district heating). New SA-based mixtures with a melting point in the temperature range from 75 °C to 100 °C and relatively high latent heat (170–260 J/g) have been proposed by Hidaka et al. [16], Nakada et al. [17] and Diarce et al. [18]. In the recent European project SAM.SSA (FP7 2012–2015; www.samssa.eu/) sugar alcohol-based materials for solar thermal seasonal storage

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Table 1
General information about the studied sugar alcohols.

	Acronym	CAS number	Formula	Provider	Purity (%)
Xylitol	Xyl	87-99-0	$C_5H_{12}O_5$	Roquette	98.43
Adonitol	Ado	488-81-3	$C_5H_{12}O_5$	Sigma Aldrich	≥99
L-arabitol	Ara	7643-75-6	$C_5H_{12}O_5$	Standford Chem.	98
Erythritol	Ery	149-32-6	$C_4H_{10}O_4$	Cargill	99.5
D-mannitol	Man	69-65-8	$C_6H_{14}O_6$	Roquette	99

Table 2
Sugar alcohol based eutectic blends [20].

Binary system	Acronym	Eutectic composition molar (%)	Eutectic composition gravimetric (%)
Erythritol/ Xylitol	Ery/Xyl	36:64	30:70
L-arabitol/ Erythritol	Ara/Ery	60:40	65:35
L-arabitol/ Xylitol	Ara/Xyl	44:56	44:56

Table 3
Melting point and latent heat of fusion of the studied materials.

	This work		Previous works: [20] and refs. therein	
	Melting point (K)	Latent heat (J/g)	Melting point (K)	Latent heat (J/g)
Xyl	368.1	267	365–369	219–263
Ado	373.0	250	368–377	220–250
Ara	376.0	280	274–276	227–284
Ery	391.2	340	389–393	310–334
Man	440.0	316	439–442	306–326
Ery/Xyl	355.0	270	355–357	248–270
Ara/Ery	359.0	225	359	225
Ara/Xyl	350.0	243	350	243

applications have also been developed and studied in-depth [19]. Contrary to short-term applications, long term storage requires PCMs undergoing stable and severe undercooling like most of SA. This allows reducing thermal losses by storing the energy at temperatures far below the melting point while minimizing the risk of spontaneous crystallization during the long period of storage. SAM.SSA developments include new SA-based eutectic mixtures lowering the original high temperatures of the single materials while keeping a high volumetric energy density [20], low-cost tailor-made carbon porous structures and corresponding carbon/SA composites with enhanced thermal conductivity [21–24], and SA micro-encapsulation [25]. A new appealing solution to easily activate the SA crystallization was also established [26].

This paper provides a rather complete characterization of the most promising SA and SA-blends among those studied in SAM.SSA project. This is five single sugar alcohols (xylitol, adonitol, L-arabitol, erythritol, D-mannitol) and three eutectic blends (eythritol/xylitol, L-arabitol/erythritol, L-arabitol/xylitol). Compared to previous publications dealing with sugar alcohols as PCMs (see [20] and references therein), this paper not only presents measurements of the melting points and latent heats of fusion, which is insufficient for modeling or for proper material comparisons, but it also provides measurements of key physical properties such as specific heat, thermal conductivity, thermal diffusivity, density and viscosity as a function of the temperature. As far as we know, the variation of the specific heat with temperature has been studied for erythritol [32,33], xylitol [30] and adonitol [31] only. The measurements were performed in solid and liquid state, but supercooled liquids were not considered. The temperature dependence of the thermal conductivity was analyzed only for erythritol [33] and, again, the supercooled liquids were not studied. Moreover, to the best of our knowledge, there are no available data for the density-temperature dependence, nor for viscosity. Besides providing full characterization of the studied materials, this paper compared their performances to those of most currently used PCMs (paraffin waxes, salt hydrates etc.) and discusses their most significant applications, including solar seasonal energy storage. The crystallization kinetics of the studied materials, which completes their characterization as PCMs for heat storage applications, will be presented and discussed in a forthcoming paper.

2. Materials and methods

2.1. Materials

Xylitol, L-arabitol, adonitol, erythritol and D-mannitol at high purity grade were purchased from different providers as indicated in Table 1. This table also includes the CAS number and the stoichiometric formula of the studied materials. The acronyms used hereinafter to refer to the single compounds are given in the second column of the table.

The eutectics formed in the binary systems erythritol/xylitol, L-arabitol/erythritol and L-arabitol/xylitol were also studied. The corresponding compositions, recently established by Palomo et al. [20], are given in Table 2.

2.2. Methods

A differential scanning calorimeter (DSC 1 Mettler Toledo) was used to determine the melting point, the specific heat and the latent heat of fusion of the studied materials. The DSC was calibrated using indium, lead and tin. Samples of about 10 mg were weighed by a Meltter XP6U ultra-microbalance and placed into aluminium crucibles with perforated lid. A flow of dry N_2 (30 ml min^{-1}) was used to purge the measuring cell. A heating rate of 10 K/min was employed for the measurements of melting point and the latent heat of fusion, with the temperature dynamic range varied from the room temperature up to 10 K above the corresponding melting point. The sapphire method was applied according to DIN 51007 to determine the specific heat of the studied materials both in solid and liquid states. In solid state, the measurements were performed every 10 K within the temperature interval from 293 K to 334 K. In liquid, the scanned temperature range is different for each sugar alcohol. The upper limit is chosen to be around 30–40 K above the melting point. The lower limit is often determined by the temperature at which the sugar alcohol crystallization occurs. Therefore, the specific heat of undercooled liquids is also investigated (Figs. 1 and 2).

Most of the materials considered in this work undergo severe undercooling, with very long nucleation induction times. Some of them do not crystallize on cooling but tend to vitrify. Besides, sugar alcohols can exist in two or more crystalline states. Therefore, all DSC tests were carried out by heating fresh crystalline powders. For SA-blends, the corresponding single compounds were ground by mortar and pestle. Then, the batches needed for DSC tests were prepared by accurate weighing (accuracy ± 0.03 mg) of the components powders and gentle mixing.

The transient HotDisk method [27] was used to determined the thermal conductivity and the thermal diffusivity of the studied materials in solid state (crystalline form). The measurements were carried out on parallelepiped-shaped samples with 50 mm×50 mm side and 12 mm thickness. A melt blending method was used to prepare the samples. A TPS 2500 instrument equipped with a Kapton insulated sensor of 2 mm in diameter was employed for measurements. The power applied to the rear face of the sample was 25 mW during 4 s. The measurements were performed at room temperature (300 ± 0.2 K).

For the measurements of the thermal conductivity and the thermal

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