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Investigating the Energy Intake/Discharge Behavior of Xylitol-Based PCM with Polymer as Thermal Storage Material

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ABSTRACT

Improving xylitol (XYL) phase change behavior is essential to maximize the energy rating in positive-coefficient thermal energy storage (PC-TES). The XYL requires activation to release the stored latent heat during solidification, which necessitates some modifications to the TES unit. This work solves the issue by introducing binder material from polymer to improve the operability aspect of XYL as TES material. Various high-density (HD) polyethylene ratios (5%, 10%, 15% and 20%) are prepared using direct mixture method to form XYL-HD mixture (XYH). Thermal properties assessment indicates that the HD content has a substantial effect on the melt behavior, particularly for ratios of 15% and 20%, resulting in a two-step melt process between 99.8/110.1℃ and 98.3/113.9℃. The microscope profile indicates that the XYL and HD are physically mixed. It confirms the FT-IR profile for the mixture, which has similar chemical identity as the base material. All synthesized XYH systems exhibit a peak during the cooling process, indicating that the system can solidify within the range of 104.7-116.9℃. It contributes significantly to the improvement of energy intake/discharge behavior. The XYH system able to achieve maximum charge and discharge efficiency around 84.6% and 74.1%, which is higher than XYL (53.9% and 42.6%). Thus, using HD contributes positively to initiating crystallization and eventually improves the operability level of XYL for PC-TES system.

1. INTRODUCTION

Technical challenge due to hyper population growth requires solution to sustain human life and protect the environment. Transition from conventional to renewable technology brings positive impact on global energy mix. It proves the effort through series of research allow high increment and production from renewable energy, such wind technology [1-3]. Further optimization utilizes hybridization and integration to combine one or more different energy technologies, such as waste recycling, for energy systems [4]. One crucial barrier for renewable systems is intermittency, which presents a serious drawback, necessitating sophisticated and advanced storage to accommodate the fluctuating supply. Various option for storing energy is offered, while the most proven technology by now is electrical and thermal energy storage (TES). In terms of storage price, TES offers greater cost-effectiveness and high flexibility in operation, which is adjusted according to the system's coefficient of temperature. For example, negative coefficient (NC) temperature is commonly applied to provide cooling effect and mobile cold storage [5-7]. Positive coefficient (PC) TES is focused on high thermal load system, such thermal plants [8], room and air heating [9]. PC-TES offers a direct contribution to support the energy transition, which is possible when combined with various heat sources through hybridization.

PC-TES works by heating the material inside the container. The type of material depends on temperature operation of the system. The usability of organic material like fatty-acid [10] and wax [11] typically applied for low load system (operates < 90℃), while higher load systems (> 200℃), like thermal power plant, use molten salt [12]. TES operates through heat exchange, making the conductivity of the material a crucial factor. PC-TES material typically has low conductivity. It is solved by several approaches, like adding volcanic particles to increase the heat intake/discharge by 25%/17.8% [13] or employing NiCo-system to enhance the conductivity up to 12.873 W/m·K [14]. Also, arranging multi-tube containers brings higher charger power about 79.7% under a suitable flow rate [15]. Recently, incorporating direct heat intake with renewable electricity has been considered as a feasible option for design simplification [16-18]. However, specific constraints remain challenging, such as the inability to recrystallize and poor solidification, which are commonly found in polyols PC-TES (PPC-TES).

PPC-TES is generally more advantageous in term of operability and economic as it has high thermal capacity, meaning compact storage can be achieved as PPC-TES has density above 1.4 kg/l [19]. Unfortunately, inability to perfectly crystallize causes serious problem, especially for xylitol (XYL) based PPC-TES. The molecular motion prone slowly during heat discharge, resulting the XYL remains in liquid state while the temperature is already below its melting point [20]. It causes severe impact on the energy release process of PPC-TES since the stored energy is unable to be discharged perfectly, resulting in a performance decrement of the system. The maximum discharge of the stored latent heat occurs when the material transforms into the solid phase, demanding excellent crystallization ability to provide excellent heat release and improve the operability of the TES system [21]. Thus, the performance increment of PPC-TES correlated significantly with the solidification ability of the material in order to achieve higher energy release and excellent discharge output. Additionally, it is related to the melting fraction of the TES material as discharge occurs, indicating the highest power output achieved along with the phase transition until the melting fraction of the material reaches zero or fully solidified [22]. Therefore, the challenge of poor discharge behavior of XYL as PPC-TES requires specific approach to improve its operability.

Literature review indicates several approaches were taken to solve the solidification issue for XYL-based PPC-TES. For example, Coccia et al. [23] employed a triggering technique (agitation) for XYL in solar cooker. The study demonstrated the triggered XYL system has a better cooling time around 80% compared to the non-triggered system. The manual triggering initiates the solidification of supercooled XYL, causing the releasement of latent heat as it solidifies. Different activation techniques were performed using bubbling, seeding, and combined bubbling-seeding for large PPC-TES. As discussed here [24], the non-triggered system caused gradual decrement of the working fluid from 110°C to 50°C while the XYL remains in a liquid state at the end of the discharge stage. It results in lower rating output with only 20 kWh_{th}, while system with activation technique achieved higher rating close to the designed target (42 kWh). Triggering is also possible for the eutectic PPC-TES using XYL and erythritol as presented here [25]. The outcome also reveals certain limitation for system with mechanical agitation only due to extreme subcooling of the mixture, resulting in longer agitation (43.3 minutes) which can be reduced with the help of seeding technique and allow for shorter agitation duration (1.8 minutes).

Activation to trigger solidification for XYL brings positive outcome to release the stored energy more effective. However, it demands additional equipment which leads to additional auxiliary component for the system. Also, another work [26] compared different triggering techniques (shearing and bubbling) for PPC-TES, implying that crystallization occurred unspontaneously with a certain induction time (around 20 minutes). Therefore, a different technique is required to overcome the technical limitation for the mechanical activation of XYL in PPC-TES. For example, embedding TES material with polymer is proven as a reliable method to improve the discharge behavior and decrease the supercooling degree. Rahman et al. [27] evaluated fatty acid (FA) with polymer for TES application, showing the mixture has a shorter liquid-solid transition (9.5°C) than FA (21.3°C). The shorter transition temperature during solidification indicates excellent discharge behavior, which maximizes the average rating output. Another study [28] evaluated wax-based PC-TES with high-density (HD) polyethylene, showing the mixture of wax/HD (85:15 wt%) has supercooling degree of 3.73°C which considerably lower than wax (8.01°C). The finding emphasizes that HD improves the material's ability to solidify during the discharge cycle, resulting in a positive outcome for the released energy.

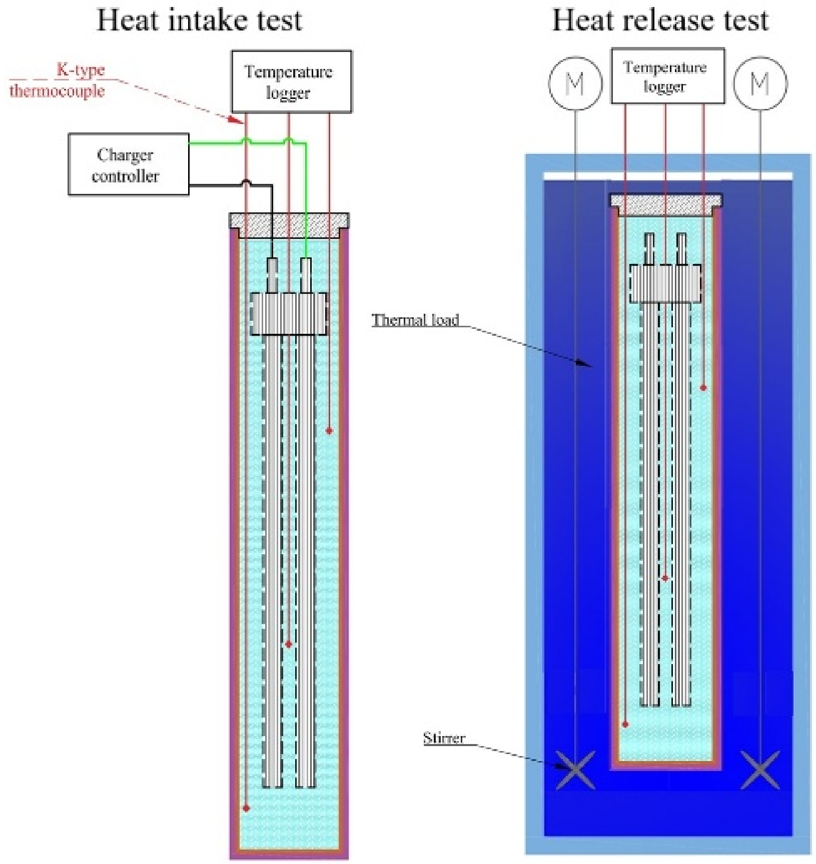
Embedding XYL with polymer can be considered as feasible option to improve its solidification behavior and bring positive influence on the operability of TES system. However, there is limited research that addresses the applicability of XYL-polymer system for TES system. Thus, the present study aims to investigate the effect of polymer addition on the XYL performance as PPC-TES material. It is the key novelty of this work, exploring a new chance to enhance the energy-interaction of XYL by embedding it with a polymer. The uniqueness comes from the solution to merge the PC-TES (XYL-HD) rather than introducing an additional system to trigger solidification. The goal is expected to improve XYL operability at specific suitable HD ratios. As a preliminary study, various HD content for XYL is examined and evaluated to understand the impact of HD content on the operability of XYL as PPC-TES material.

2. MATERIALS AND METHOD

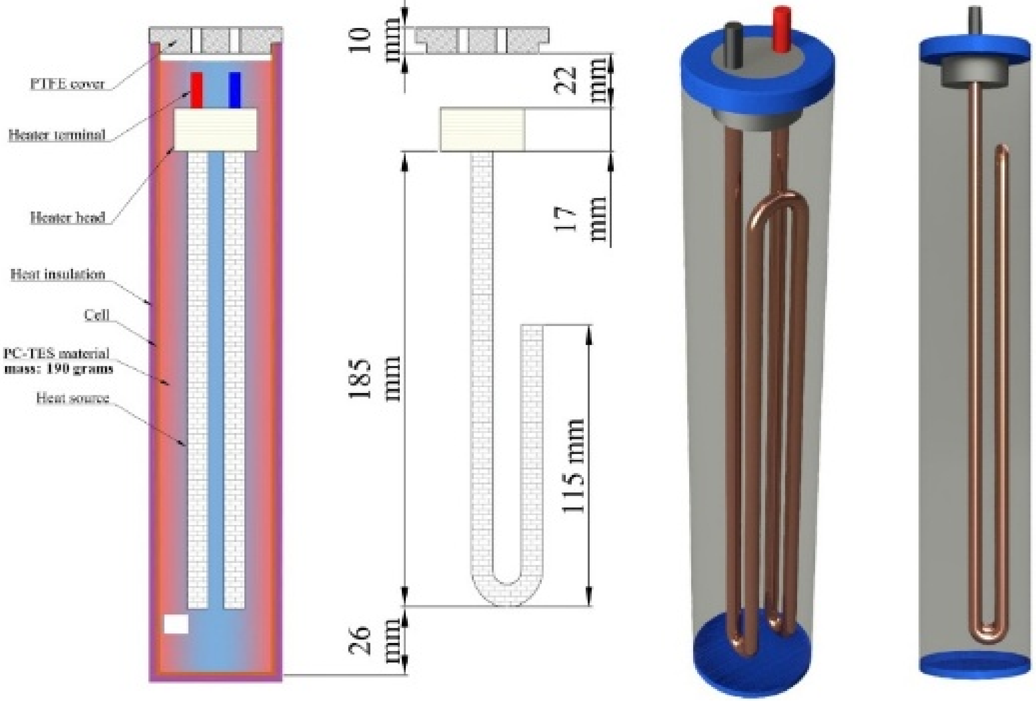
The study assessed commercial XYL ($C_5H_{12}O_5$) and HD polyethylene. The synthesizing process was conducted at 130°C and 180°C ± 10°C for XYL and HD. The molten materials were mixed using a direct blending approach. The mixture was stirred continuously at temperature slightly higher than melting point of HD. Once the two components were mixed adequately, cooling process was conducted until reaching room temperature. Then, the process was repeated five times to ensure homogeneity. In addition, the cooling process allowed the XYL content to penetrate the HD structure, ensuring an effective binding mechanism between the two components. HD content was adjusted carefully at a specific ratio: 5% (XYH_1), 10% (XYH_2), 15% (XYH_3), and 20% (XYH_4).

Several material characterizations were performed to obtain the necessary information for understanding the effect of HD content on the XYL. Structural phase was analyzed with X-ray diffraction (XRD, PANalytical) and chemical structure was evaluated using infrared spectrometer (FTIR, PerkinElmer). Thermal properties were assessed using scanning calorimetry/thermogravimetric (DSC/TG, NETZSCH). It used 20.2 mg of each sample and placed in an alumina crucible (Al_2O_3, 85 μl). The heating rate for DSC was set at 5°C/min based on the recommendation of this study [29] with a measuring range 25-150°C. For the cooling process, nitrogen flow was set at 250 ml/min. The determination of heating/cooling rate was also considered as per per ISO 11357-3. The TGA was performed at a temperature range of 30-550°C with an identical heating rate. In addition, surface characteristic observation was performed using a scanning electron microscope/SEM (Prisma E, Thermofisher) operating at 10 kV for all specimens, including XYL in both powder form and after resolidified.

Experimental evaluation of intake/discharge behavior was conducted to assess the impact of material modification on performance and operability aspects of the PC-TES material, focusing on the ratio of thermal energy intake/discharge between the material and system. The evaluated sample was placed in a cylindrical cell, where a heater is located to heat the material. Three measuring points were directly in contact with the material and recorded its temperature during evaluation (Figure 1(a)). The energy rate for heating the material was 80 J/s with a temperature operation between 30-140°C. The energy input to the heater was recorded using digital wattmeter and used as a base point to define the energy input to the system [30]. After completing the charge cycle, the cylindrical cell was placed in liquid tank for discharge. The stored heat discharged to the load (water) at a starting temperature 10°C. The changes in water temperature are then taken as an indicator to determine the total energy output from the material. Detailed assembly of the apparatus is shown in Figure 1(b) (including the dimensions).



(a) Heat intake/release evaluation scenario



(b) Dimension and model of the cell assembly

Figure 1. Experimental evaluation for assessing the energy transfer for the materials

3. RESULTS AND DISCUSSION

Evaluation of the structural phase from the XRD pattern provides essential information on the impact of mixing XYL with HD (Figure 2). Multiple peaks appear from XYL at 2θ: 17.6°, 22.1°, 29.3°, 38.1°, 46° and 54.6°, indicating enantiomorphic structure from typical XYL. It causes the XYH structure to transition into an amorphous pattern, as the synthesis process requires melting XYL initially. Moreover, the transition to amorphous signifies the high solubility of XYL with water. On the other hand, HD demonstrates sharp peaks at 21.7° and 24.1°, showing high crystallinity nature of the polymer. The mixture reveals the peaks HD shift as a result of structural change of XYL.

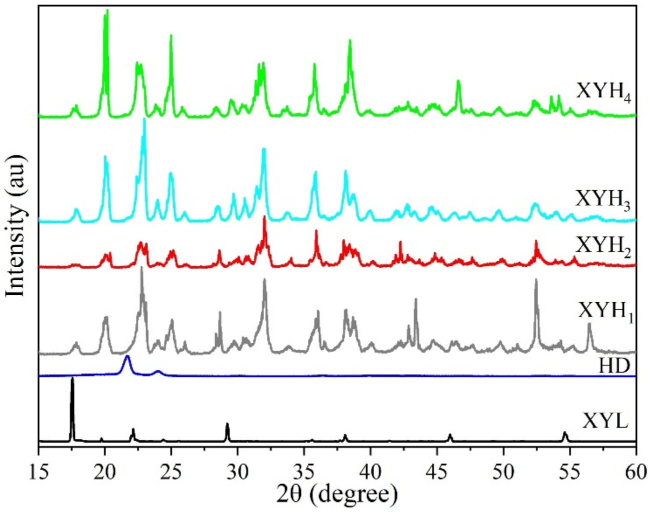


Figure 2. Evaluation of structural phase for XYL, HD and XYHs

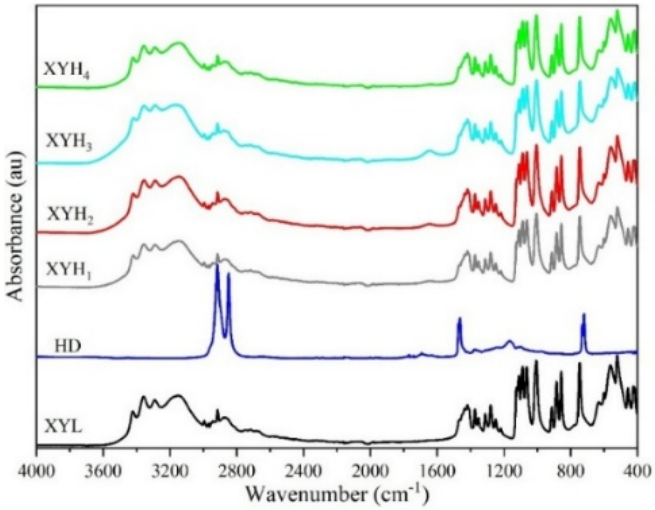
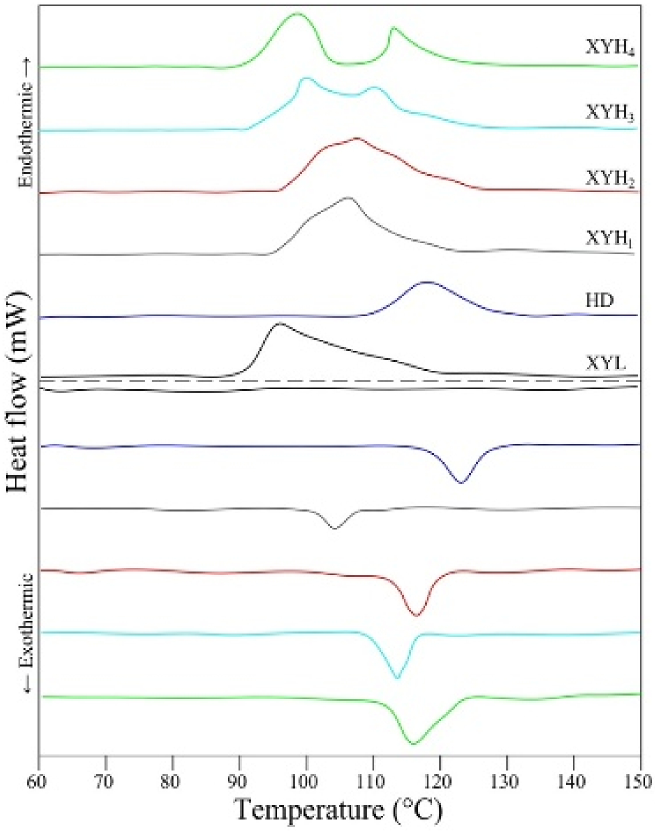


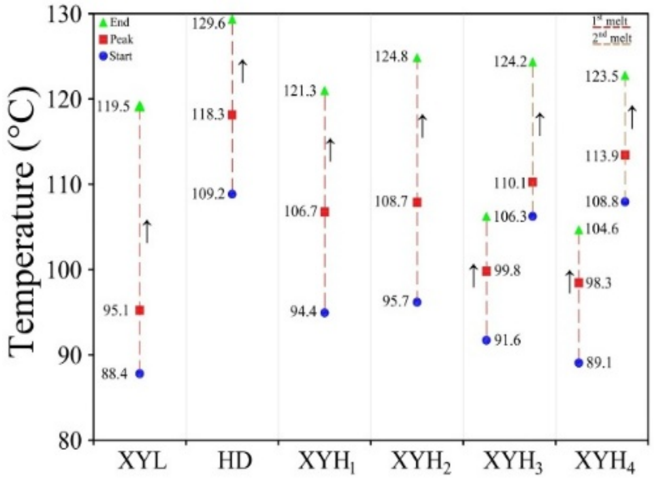
Figure 3. FT-IR spectra for XYL, HD, and XYHs

Chemical profiles from XYL and HD, including the mixtures (XYH), were obtained based on FT-IR spectra (Figure 3). Broadband at ranges 3460-3014 cm^{-1} indicates stretching vibration −OH, which is the specific characteristic of the hydroxyl group, including the possibility of water adsorption of the prepared XYL. It confirms the transition of XYL into an amorphous state (Figure 2), making additional modification extremely essential for XYL-based PPC-TES to prevent high water contamination from moist air. It is also the reason to perform material preparation without pretreatment since a typical TES system operates in an open environment without using inert gas. The peaks are also observed at 1311 cm^{-1} and 1005 cm^{-1}, defined as stretching vibration C−O. The fingerprint area reveals the key identity of rocking vibrations of CH_2 which occurs at HD, while XYL shows vibration O−H functional group at 745 cm^{-1}. Peak HD is obtained at 2900-2800 cm^{-1}, demonstrating stretching vibrations of $-CH_2$, followed by functional groups of $-CH_2$ and $-CH_3$ at 1463-1472 cm^{-1}.

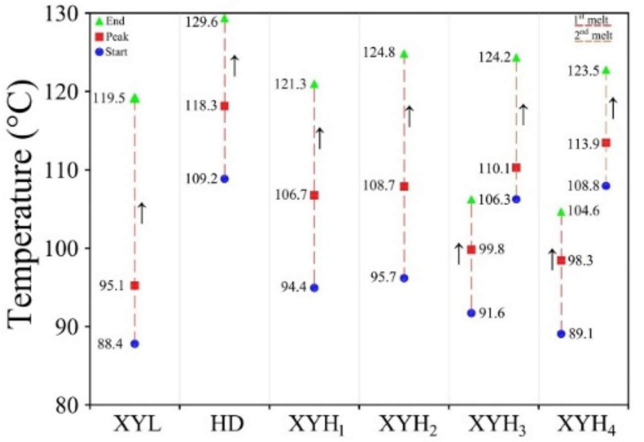
Principal peaks HD (stretching vibrations of $-CH_2$) remain shown for XYH, while the key identity of XYL also observed distinctively, particularly for hydroxyl group, stretching vibration C−O, and bending vibration O−H in the corresponding spectrum. The profile for XYH shows the mixture maintains each pattern of the base (XYL and HD), implying no chemical reaction occurs between the two components within the mixture.



(a) DSC profile



(b) Detailed transitional temperature solid → liquid



(c) Detailed transitional temperature liquid → solid

Figure 4. Thermal evaluation PC-TES

The operability of XYL as PC-TES is related to the melting/freezing behavior. It is shown according to the DSC curve (Figure 4(a)). The curve provides an important parameter to understand the phase transition behavior during energy intake/discharge. For example, peak temperature for XYL is observed at 95.1°C with thermal capacity (latent heat) of 235.8 kJ/kg. The properties are considered as the essential value which makes XYL is thermodynamically favorable as PC-TES material, making it ideal for various thermal system which demands relatively higher temperature than typical wax/fatty acid PC-TES. HD-polymer experiences melting peak at 118.3°C with total thermal capacity of 121.5 kJ/kg. It is the reason to limit the HD ratio in order to maintain the effective thermal capacity of the mixture and ensure the total storage capacity of PC-TES remains applicable. The shifts in melting pattern for XYH_1 and XYH_2 demonstrate a single curve, which occurs between the peak temperature of XYL and HD. The total thermal capacity for both mixtures drops to 207.6 kJ/kg and 214.7 kJ/kg. Also, adding a higher HD ratio causes substantial changes in the melting behavior, which is shown by two consecutive peaks.

The presence of HD improves the melt behavior of XYL based on the melting range (Figure 4(b)). The total range for XYL is 31.1°C, while HD is only 20.4°C. As a result, all XYH samples exhibit a lower total melting range than XYL, with XYH_1 and XYH_2 showing reductions of 13.5% and 6.4%, respectively. The viscoelastic nature of HD affects the rheological aspect of the mixture, contributing to a decrease in the melting duration of XYL. However, higher HD content (XYH_3 and XYH_4) causes multiple transitions, which is unideal for the practical consideration of PC-TES. The key issue of XYL is apparent clearly in the cooling process, as it shows no peak. It indicates that the XYL cannot resolidify under the designed process, which corresponds to the supercooling state.

Indication of recrystallization ability of XYL-HD system is shown based on the presence of peak upon cooling process (Figure 4(c)). Transformation micro-phase of HD allows the freezing range to achieve at lower point (11.9°C). Also, smaller HD ratio makes the range even shorter to 7.7°C which appears in XYH_1. The transition becomes longer as the HD content elevates to 10%, demonstrating the total transition to 10.4°C (XYH_2). However, the point shows a lower value for XYH_3 (9.4°C), while 20% of HD causes a longer freezing range to 13°C (XYH_4). The variation is considered as substantial evidence to justify complex solidification mechanism between XYL-HD system. Thus, adjustment on suitable HD content becomes essential as an interplay aspect on solidification behavior, including factors to maintain the freezing range at the desired rate.

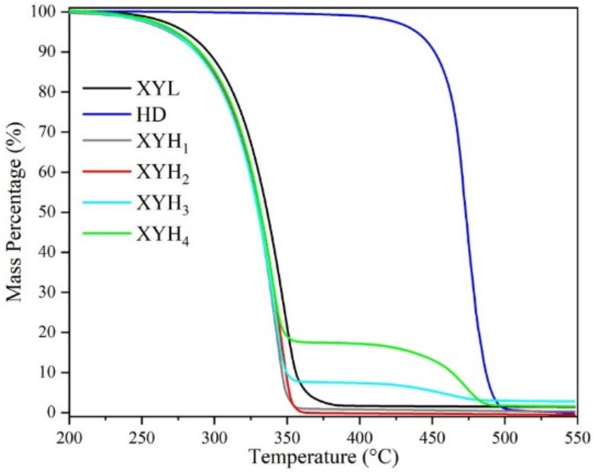
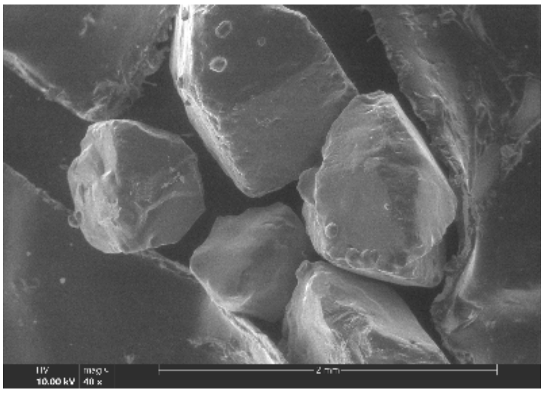
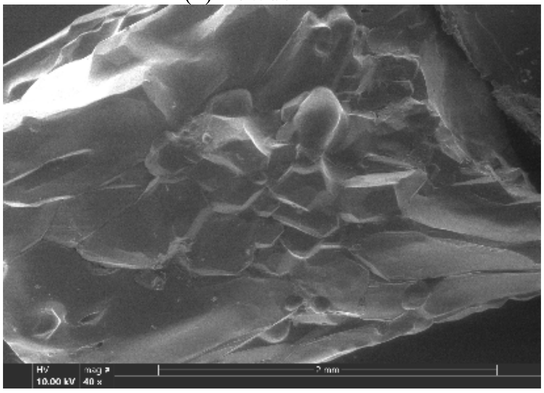


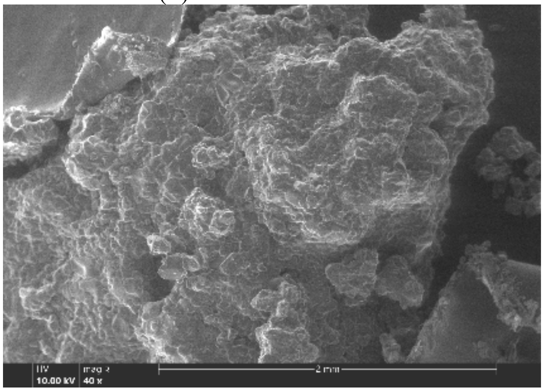
Figure 5. Decomposition behavior for tested PC-TES



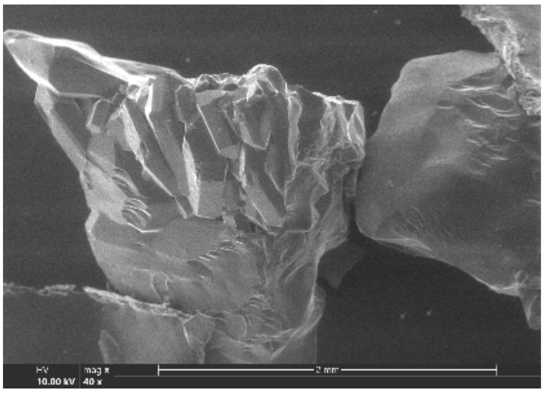
(a) Powder XYL



(b) Resolidified XYL

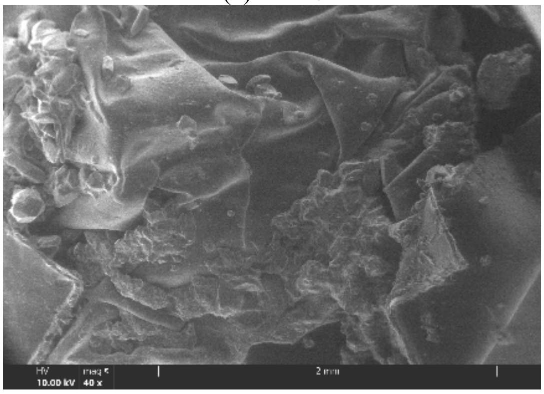


(c) XYH_1



(d) XYH_2

(e) XYH_3

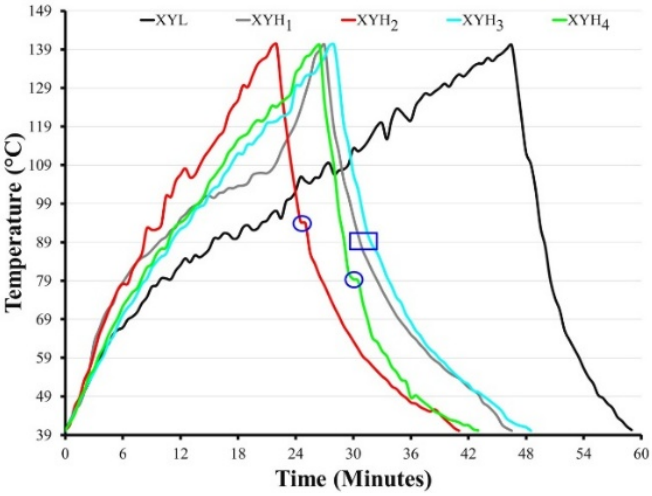


(f) XYH_4

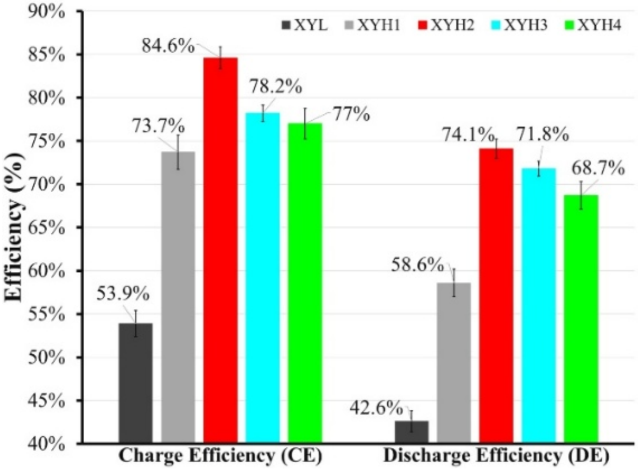
Figure 6. Microscope profiles for the evaluated PPC-TES

Figure 5 plots the decomposition characteristic for all evaluated samples. From the plot, thermal stability of XYL is insufficient for high operating temperature system as direct decomposition occurs > 235°C. The thermoplastic properties of HD cause decomposition to start at a higher temperature (above 408°C). XYH_1 and XYH_2 show the identical decomposition range with XYL, where the peak point occurs at 352°C. However, increasing the HD content above 10% results in multiple decomposition stages. It shows the possibility of phase segregation between XYL-HD. It emphasizes the endothermic pattern (Figure 4(a)) for both samples, which shows two distinctive peaks at different temperatures.

The first impression reveals a notable effect of the mixing process on the changes in the surface profile of XYL, as observed in the microscope image. The powdered XYL shows multiple independent crystal chunks (Figure 6(a)). However, melting the powder and resolidifying it causes the chunks to merge together into a solid block with minor crystal formation on the surface (Figure 6(b)). It may relate to the main issue of XYL which experiences severe supercooling as not all crystal is perfectly reformed after heating, resulting in semi-solidified mixture that demonstrates imperfect solidification. Introducing HD as binder (5%) enhances the reformation ability of XYL, as seen in multiple blocks that form together (Figure 6(c)). Interestingly, adding more binder ratio (10%, Figure 6(d)) provides excellent encapsulation, which causes a clearer boundary region between the XYL/binder. It becomes distributed appropriately as the binder increases (15%, Figure 6(e)), producing well-attached region of XYL/HD system. However, employing 20% of HD (Figure 6(f)) dominates as an encapsulated system, indicated by a clear HD-plastic profile. Despite the variation, XYHs demonstrate suitable solidified profile, indicating the HD content promotes a better solidification mechanism for XYL.



(a) Charging/discharging pattern



(b) Comparison of charging/discharging efficiency

Figure 7. Operability behavior based on heat intake/release

The effect of HD content is observed notably according to the temperature evolution, which relates to the heat intake/release behavior (Figure 7(a)). XYL exhibits a longer temperature evolution, resulting in a longer charging duration to achieve the targeted upper temperature. The presence of HD changes the heat intake pattern, resulting in a faster charge duration, which is essential to prevent potential losses during the process. Specifically, the optimum charge duration is achieved by XYH_2, which able to reduce the charge duration around 52.7%. It indicates positive impact for using specific ratio of HD content which promotes excellent energy input process. The discharge pattern for XYL occurs directly without indicating any graduation, demonstrating the effect of supercooling which causes temperature decrease rapidly without involving latent heat releasement. In contrast, XYH exhibits temperature fluctuation, as indicated by a slower decrement that occurs around 89°C for XYH_1 and XYH_3 (blue rectangular). Interestingly, using HD content 10 wt% (XYH_2) causes the transformation to become clearer, which reveals the initiation of solidification at 92.2°C, while a higher HD content of 20 wt% (XYH_4) leads to initiation occurring at a lower temperature around 80.4°C (blue circle). Despite that, the temperature fluctuation during discharge indicates the changes of phase transformation behavior for XYH, confirming the DSC obtained profile (Figure 4(a)).

Indicator aspect operability can be obtained from charging efficiency (CE) and discharging efficiency (DE). As presented in Figure 7(b), XYL has the lowest CE value around 53.9%, indicating that the input energy cannot be utilized effectively to achieve maximum temperature operation (fully charged). It emphasizes the profile temperature of XYL (Figure 7(a)), which indicates the longest charge duration, leading to substantial heat losses through the process. A shorter charge cycle enables all XYH to achieve a higher CE value, with the highest percentage in XYH_2 (84.6%). Furthermore, XYL exhibits poor discharge behavior, resulting in the lowest DE of 42.6%. It signifies the stored latent heat unable to release effectively as the discharge process occurred. It is solved by introducing HD, where all XYH demonstrate a higher DE value ranging between 58.6-74.1%.

Introducing HD for XYL based PC-TES results in two benefits, both in the intake and discharge process, making the operability of XYL as PC-TES system more reliable. However, the oxygen content within the XYL structure may result in oxidative degradation of the HD. It is also considered by the operation condition of PC-TES system which is directly exposed to the environment without using inert gas. The degradation potentially causes phase segregation and affects the operability aspect of the system after certain cycles. As this work focuses on fundamental discovery for improving the operability aspect of XYL based PC-TES, future work can focus on long term stability for the system XYL-HD by assessing the key parameters of the system after specific thermal cycling treatment.

4. CONCLUSIONS

An experimental assessment to address the poor operational behavior of XYL is conducted using polymer. The mixture (XYL/HD) exhibits a superior aspect compared to pure XYL, particularly in terms of the intake/released energy ratio, which increases to 30.7%/31.5% when 10% HD is used. The study shows a clear indication for the potential of using HD on XYL system to improve the phase-change behavior, allowing the system to achieve a higher ratio energy intake/discharge. The mixture indicates no substantial chemical transformation, making the produced XYL-HD system only mixed physically, confirming the morphology and phase structure analysis based on SEM and XRD patterns. However, adjustments to the specific HD content become crucial, as the findings indicate that the most effective energy ratio is obtained using 10% HD. Higher HD content may lead to a significant decrease in storage capacity and energy intake/release for operation.

This work reveals a new chance to reutilize HD for energy system as a binder material for XYL in PC-TES technology. Despite that, the current work assesses a fundamental aspect, which encourages further evaluation to develop an advanced PPC-TES system. Future work is recommended to optimize HD content for the system, including methods to prevent potential degradation and ensure cyclic stability. Furthermore, modifications to the operability aspect, such as the heat exchanger and cell assembly, can be performed to produce an integrated PC-TES technology. Comprehensive work in this study provides essential information to maximize the usability of XYL as a ssadvanced storage material to support the global energy transition, especially for thermal-based system.

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