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Performance evaluation of solar cooker using some [N^+_{4444}] based ionic liquids as thermal energy storage materials

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ABSTRACT

Seven ionic liquids (ILs) based on tetrabutylammonium cation [N^+_{4444}] and inorganic anions were tested for their applications in a solar cooker. Thermophysical characteristics like melting point, density, heat of fusion, heat of solid - solid transitions, specific heat capacity and thermal conductivity were used to determine the efficiency of ILs used as phase change materials (PCMs) in solar thermal applications. A delay time of two hours was observed on an average in achieving the highest temperatures. A majority of the ILs extended the time required for achieving ambient temperature to a tune of around four hours. The ILs containing bromide, iodate and nitrate proved to be more efficient than the thiocyanate, bromate and hexafluorophosphate moieties. Copyright © 2013 VBRI press.

Keywords: Ionic liquid; thermal energy storage; phase change materials; solar cooker.

Introduction

The utilization of solar energy in all the viable thermal applications can reduce the emission of greenhouse gases and can help in the conservation of fuel **[1]**. The said task can be carried out more effectively in the parts of the world where the sunshine is ample. However, the discrete nature of this energy has led a surge in the search of materials with the capacity to store and then release the energy in a sustained manner. These materials are known as thermal energy storage devices (TESDs) **[2-5]**. Such materials have their melting points within the usable temperature range and are hence known as phase changing materials (PCMs). Several thermophysical, chemical and economic assets are desired from an ideal PCM to be used as TESD. It is expected that PCMs should have high value of density, heat of fusion, heat capacity and thermal conductivity **[3]**.

The classical PCMs include organic as well as inorganic materials. Organic PCMs like paraffin, fatty acids and polyethylene glycol have been used for thermal energy storage **[1]**. However, high volatility and flammability have restricted the popularity of organic phase changing materials in thermal applications. Inorganic materials like hydrated salts, potassium hydroxide and magnesium chloride also have the properties, which make them suitable PCMs. Major disadvantages of the inorganic phase changing materials are corrosiveness, low thermal stability and high undercooling **[6]**. Eutectic materials like 66.6 % $CaCl_2 \cdot 6H_2O$ + 33.33 % $MgCl_2 \cdot 6H_2O$ have recently emerged as promising PCMs. However, they have no chemical combination between different materials. Water also allows thermal applications. The main drawbacks of water are corrosiveness and volume expansion at a high temperature **[7]**.

Ionic Liquids (ILs) are organic salts having a low melting point. Low vapor pressure and recycle ability are major green features of ILs. High Density, high thermal conductivity along with high thermal stability and non-flammability are some important characteristics of ILs **[8, 9]**. ILs of organic cations like substituted imidazole, pyrrole, pyridine and ammonium with anions like halides, tetrafluoroborate, hexafluorophosphate and bis (trifluoromethylsulfonyl) imide have been mainly investigated for their roles in catalysts, solvent, Li - Ion Battery and extracting agent. Recently, they have been studied for their uses in the fields of thermal energy storage, solar cell, solar power plant and heat transfer fluids **[10-17]**.

Solar cookers provide a cost-effective and green solution for reducing energy consumption **[18-20]**. Different materials with thermal energy storage capacity are being used in such cookers to increase the achievable temperatures and efficiency of these systems **[21-23]**.

In continuation of our earlier work **[24, 25]**, the present paper aims to investigate; thermal energy storage efficiency of some tetrabutyl ammonium based ILs. The investigations were carried out in standard solar cookers during April 24, 2012 to May 18, 2012 at 22° 33' N, 72° 55' E. The presence of ILs imposed a significant delay of about 2 hours in achieving the cooking temperature. Due to the sustained release of energy by ILs, the time required for attaining the ambient temperature was also delayed in each case by

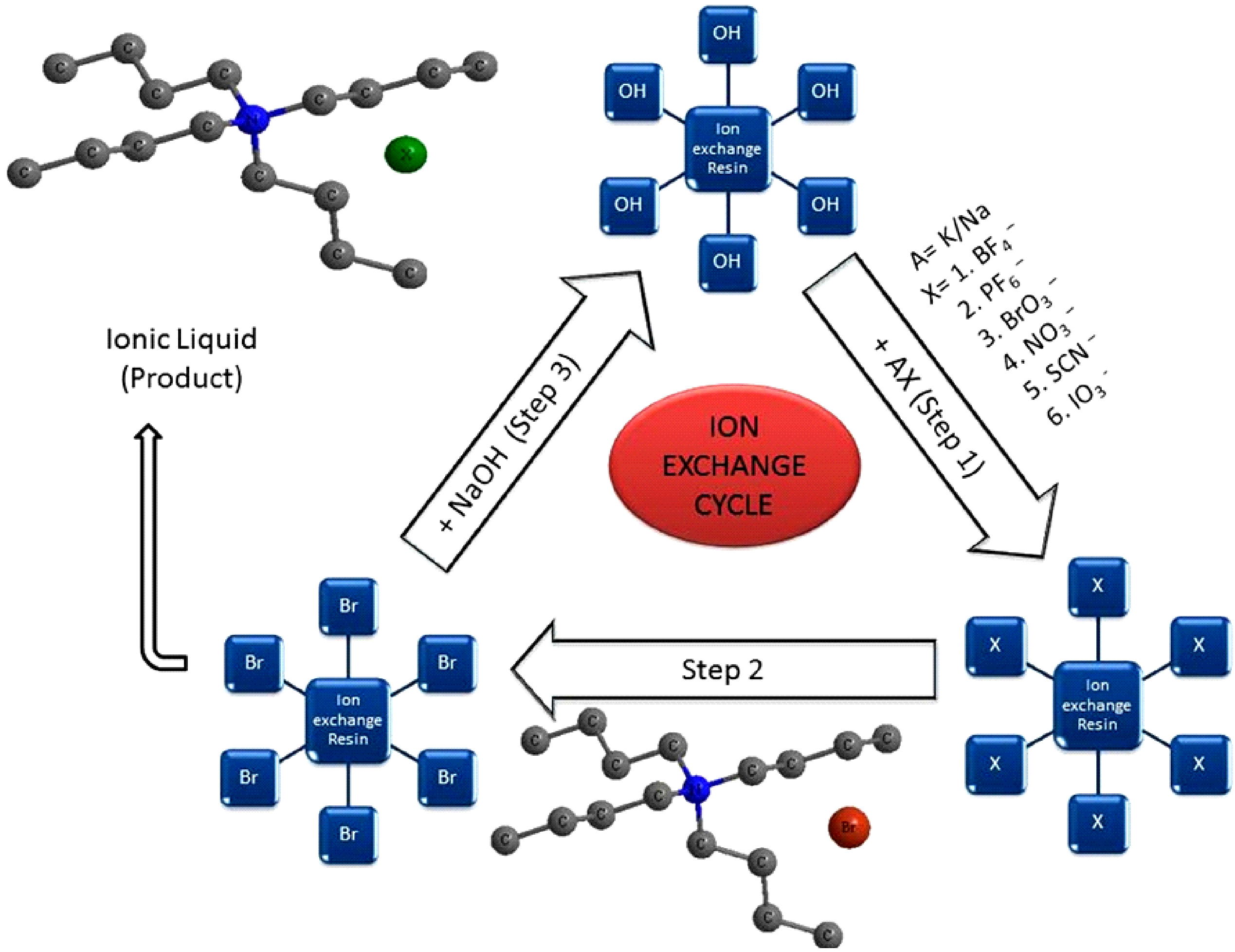

Fig. 1. Ion-exchange approach in IL synthesis.

around three to four hours. The efficiency of the ILs used as TESD was calculated using a known method.

Experimental

Materials

Tetrabutylammonium bromide (≥99.0%) was procured from Sigma-Aldrich. Ion exchanger Amberlite IRA-67 (weakly basic anion exchanger hydroxide form) resin was obtained from Merck. Potassium bicarbonate, sodium tetrafluoroborate, potassium hexafluorophosphate, potassium bromate, potassium nitrate, potassium nitrite, potassium thiocyanate and potassium iodate (≥99. 5%) obtained from Sigma-Aldrich were used directly. Tetrabutylammonium based ILs have been synthesized by the ion-exchange method with slight modification in the procedure **[26]**.

Synthesis

A column with 0.2 m length and 0.02 m diameter was filled up with 20 g of activated resin. The column was then washed with deionized distilled water. The column was then treated with 100 ml of 1N salt solution containing the desired anion. The column was once again washed with distilled water until the eluant did not show any traces of the anion. 5g quantity of tetrabutylammonium bromide dissolved in 100 ml of water was then introduced in the column. Bromide was found to be absent in the eluant. Water from the eluant was then removed under vacuum (15mm/Hg) at 60^{o}C to 70^{o}C temperature followed by drying the samples in vacuum desiccators over calcium chloride for 72 hours. **Fig. 1** shows the ion-exchange approach in IL synthesis.

Measurements

Melting points were measured using capillary tube apparatus, and were quoted as the visual observation onset of the melt. The thermometers used in the entire study had an uncertainty of ±0.5 ^{o}C. Differential Scanning Calorimeter (DSC) was carried out on Mettler STAR SW-8.10 in the temperature range of -50 ^{o}C to 200 ^{o}C in a nitrogenated atmosphere with the scanning rate of 10 ^{o}C min^{-1}.

The solar energy storage efficiency of ILs was investigated in a solar cooker with standard specifications as shown in **Table 1**. Horizontal insolation (I_s (hori.)) was recorded by pyranometer with ±2 % accuracy. Thermal sensor for ambient temperature (T_{amb}), pyranometer and calibrated J-type thermocouples made of copperconstantine were used to measure temperatures at different state points attached with data logger DT600 Series 3.

Table 1. Solar Cooker Specification.

Part	Length (cm)	Width (cm)	Height (cm)
Outside	30	30	12
Inside	22	22	08
Tin-box (Wt. =162.0 g)	20 (diameter)	-	06

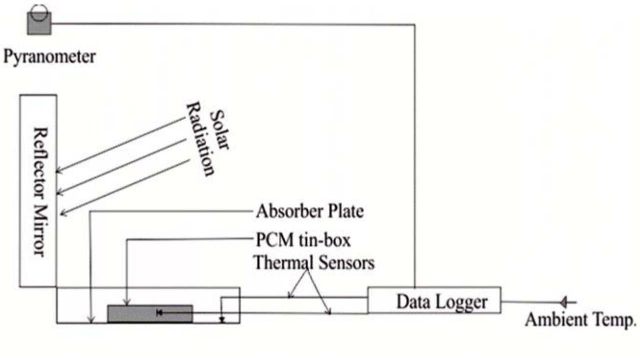

Fig. 2. Experimental Setup.

Table 2. Thermophysical Properties of ILs

Sample No.	ILs	T_m (^{o}C)	ρ (kg m^{-3})	ΔH_m (kJ kg^{-1})	Cp (kJkg^{-1} ^{o}C^{-1})	k_s & (k_l) (W m^{-1} ^{o}C^{-1})
1	$[N^{+}_{4444}][BF_4^{-}]$ Tetrabutylammonium tetrafluoroborate	71	1291	253.75	1.00	0.108 (0.383)
2	$[N^{+}_{4444}][PF_6^{-}]$ Tetrabutylammonium hexafluorophosphate	71	1234	155.00	0.40	0.062 (0.381)
3	$[N^{+}_{4444}][BrO_3^{-}]$ Tetrabutylammonium bromate	86	1050	153.71	0.30	0.048 (0.659)
4	$[N^{+}_{4444}][NO_3^{-}]$ Tetrabutylammonium nitrate	98	1195	144.00	1.50	0.062 (0.954)
5	$[N^{+}_{4444}][SCN^{-}]$ Tetrabutylammonium thiocyanate	110	1143	253.00	0.20	0.054 (0.936)
6	$[N^{+}_{4444}][Br^{-}]$ Tetrabutylammonium bromide	102	1039	193.00	2.00	0.034 (0.402)
7	$[N^{+}_{4444}][IO_3^{-}]$ Tetrabutylammonium iodate	44	1279	113.00	2.70	0.166 (0.902)

Fig. 2 shows a schematic diagram of the experimental setup. First of all, the measurements were done in an empty tin - box during the period of 10:00 am to 4:00 pm. The insolation data and ambient temperatures were constantly recorded during this period. The temperatures of the air inside the empty tin - box ($T_{w/o\ pcm}$) and absorber plate (T_{plate}) were recorded at an interval of 10 minutes. After 4:00 pm, the reflector mirror of the cooker was closed with continued measurement of the temperatures. The readings were recorded till the air temperature inside the tin - box equalled the ambient temperature. In the subsequent experiments, 500 g quantity of ILs was loaded in the tin – box and the temperatures were measured in the same manner. In these experiments, the temperatures of ILs were measured instead of air temperature inside the tin-box.

Result and discussion

Thermophysical properties

Table 2 shows the melting points (T_m) of all ILs measured by the direct melting point method. A wide range of melting points (44 ^{o}C to 110 ^{o}C) was observed across the series. Density (ρ) of the materials under present investigation was measured using a specific gravity bottle method at their melting temperatures. The uncertainty of measurement for the bottles used was 0.00001 g/ml. A close proximity in the density values was observed in the samples under present investigation as shown in **Table 2.**

Melting endotherms in the DSC thermograms (**Fig. 2.**) were used to calculate the heats of fusion (ΔH_m) for the samples as shown in **Table 2.**

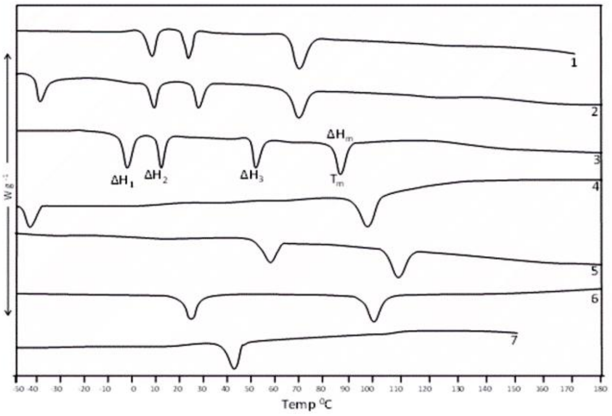

Fig. 3. DSC thermograms of ILs.

Thermograms of most of the ILs exhibited one or more additional endotherms prior to melting point (T_m). Each of these peaks represents a solid-solid transition occurring in the samples before melting **[26]**.

Specific heat capacities (C_p) of all the ILs reported were calculated from the DSC thermograms **[27]**. These values varied in the range of 0.20 to 2.70 kJ kg^{-1} ^{o}C^{-1}. The thiocyanate moiety registered the minimum value of heat capacity while nitrite and iodate containing ILs showed highest values. The remaining samples exhibited intermediate values for heat capacity as shown in **Table 2.** The thermal conductivity (k_s and k_l) of ILs in solid as well as liquid state was calculated using a known method **[28]**.

In order to remove the visual uncertainties, the experiments were repeated with fresh samples until constant values of time were recorded. All the samples registered several fold high values of thermal conductivity in the liquid state as compared to that of the solid state. These values showed a considerable variation both in state to state basis and sample to sample basis. Thermal conductivities of the samples are shown in **Table 2.**

The materials prepared by the method shown ahead exhibited interesting thermal properties. It was worthwhile to examine the efficiency of the materials under investigation in real thermal applications. Performance improvement of a solar cooker was therefore tested using the ILs as PCM in it. **Fig. 4(a)** shows the variations in horizontal insolation (I_s (hori.)), ambient temperature (T_{amb}), air temperature inside the tin-box ($T_{w/o\ pcm}$) and absorber plate temperatures (T $_{plate}$). The highest insolation was registered at 881 W / m^2 at 12:20 pm. The highest values of ambient temperature, air temperature inside the pot and absorber plate temperatures were 42.7 °C at 3:00 pm, 115 °C at 1:40 pm and 118 °C at 1:40 pm respectively on the 24th of April 2012.

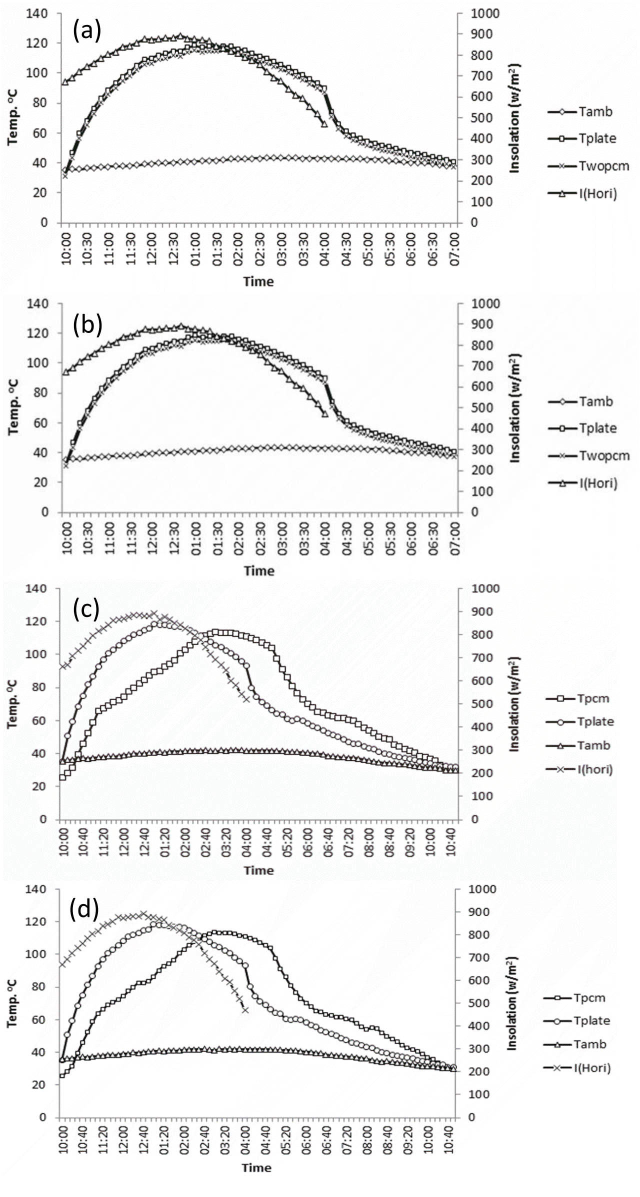

Fig. 4. Time versus I$_{s\ (hori)}$, T$_{amb}$, T$_{plate}$ and T$_{w/o\ pcm}$ for (a) without PCM, (b) [N$_{4444}^{+}$] [BF$_4^-$], (c) [N$_{4444}^{+}$] [PF$_6^-$] (d) [N$_{4444}^{+}$] [BrO$_3^-$].

Fig. 4(b-f) represent the variations in horizontal insolation (I$_s$ (hori.)), ambient temperature (T$_{amb}$), absorber plate temperature (T $_{plate}$) and PCM temperature (T$_{pcm}$). Only minor changes were observed in insolation and ambient temperature on a day to day basis during the entire period of study. The curve of T$_{pcm}$ was fairly similar to that of plate temperature. However, the initial rise in temperature of the plate was much faster than that of the PCM. The plate temperature generally exhibited its maxima after around 30 minutes of the maximum insolation. The plate temperature exhibited a plateau shape during the time period of 12:30 to 2:00 pm in the temperature range of 112-117 °C. The PCM initially absorbed the heat which was released later on. The PCM absorbed heat gradually with an increase in temperature until 70 °C. Near the melting point (71 °C), the PCM strongly absorbed the heat maintaining isothermal situation for a short but observable time. The PCM temperature exhibited a 2 hour delay in achieving the temperature of 100 °C which is required for cooking. It was also observed that the highest temperature achieved by PCM was around 5 °C lower than the highest temperature value of the plate. After melting, the PCM temperature kept on rising until it came near to the plate temperature and was maintained for about 2 hours. From the figures, it is seen that after the intersection with the plate temperature curve, the PCM temperature curve remained significantly higher than plate temperature. This provided a two hour delayed cooking time. Significant time parameters of ILs like the time required to achieve 100°C t $_{req.}$ (100), a time in which temperatures above 100 °C maintained (t $_{maint.}$ (100)) and time extended in achieving ambient temperature t $_{ext.}$ (amb.) are shown in **Table 3.**

Table 2. Significant time parameters for different ILs used in the solar cooker.

System	T $_{req.}$ (100) (Minute)	T $_{maint.}$ (100) (Minute)	t $_{ext.}$ (amb.) (Minute)
Without PCM	100	200	n. a.
[N$^+_{4444}$] [BF$_4^-$]	220	190	260
[N$^+_{4444}$] [PF$_6^-$]	220	190	230
[N$^+_{4444}$] [BrO$_3^-$]	220	190	230
[N$^+_{4444}$] [NO$_3^-$]	210	180	300
[N$^+_{4444}$] [SCN$^-$]	220	190	260
[N$^+_{4444}$] [Br$^-$]	220	190	290
[N$^+_{4444}$] [IO$_3^-$]	180	200	210

Fig. 4(b) represents the thermal energy storage and release characteristics of the ILs containing tetrafluoroborate anion used as a PCM. In this PCM temperature curve, the solid - solid transitions occurring before the melt were not reflected. This observation was as expected because the said transitions occur below the ambient temperature values. The solid-liquid phase change transition occurring around 71 °C prevented the rise in the PCM for a short time as shown in the graph. During the temperature decrease in the evening time, the solidification of the PCM occur around 70 °C. The release of lattice energy increased the PCM temperature to a tune of about 5 °C during the solidification. This increased temperature further extended the time required for gaining the ambient temperature for about 4 hours and 20 minutes. Thermal energy storage and release characteristics of IL containing hexafluorophosphate anion showed in **Fig. 4(c)** are practically indistinguishable from the former one. The DSC thermograms of both these PCMs showed only a minor difference in terms of an additional low temperature solid-solid transition observed in case of the fluorophosphates moiety. This difference is not expected to create any significant change in the thermal application under the studied temperature range. However, due to the relatively low heat of fusion, the fluorophosphates anion containing PCMs delayed the achievement of ambient temperature only up to 3.5 hours.

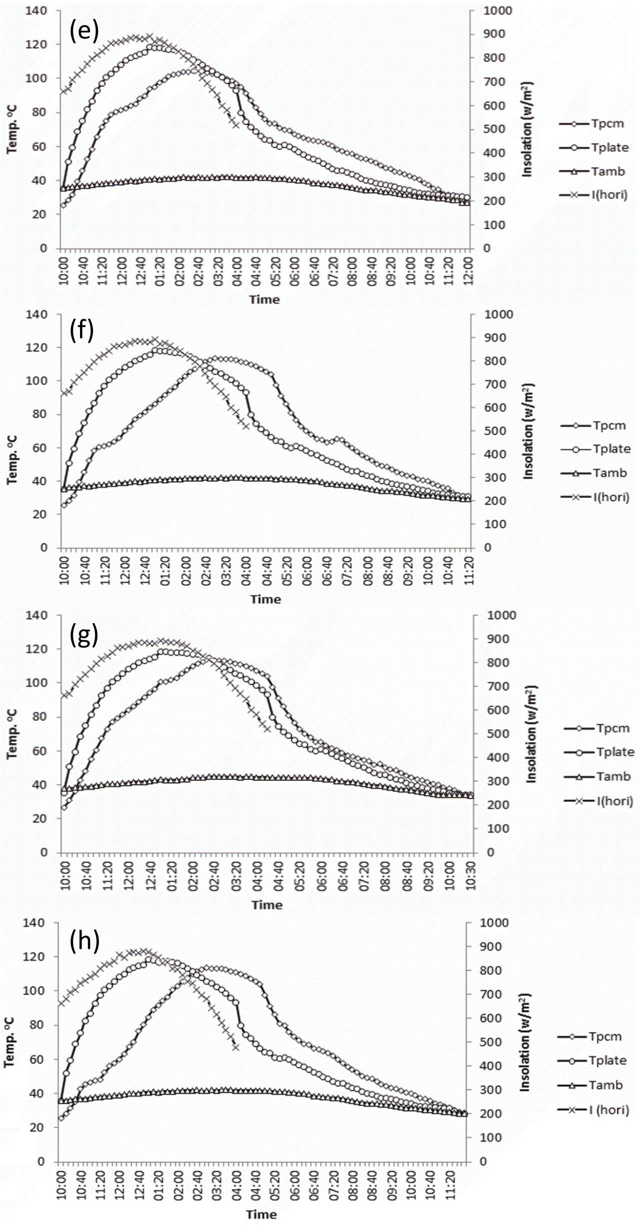

Fig. 4. Time versus $I_{s\ (hori)}$, T_{amb}, T_{plate} and $T_{w/o\ pcm}$ for (e) [N_{4444}^+] [NO_3^-], (f) [N_{4444}^+] [SCN^-], (g) [N_{4444}^+] [Br^-] and (h) [N_{4444}^+] [IO_3^-].

Fig. 4(d) represents the thermal energy storage and release characteristics of the IL containing bromate anion used as a PCM. In this T_{pcm} curve, one of the three solid - solid transitions occurring before the melt are reflected. This transition (53 ^oC) coupled with melting transition (86 ^oC) resulted in a dramatic decrease in the slope of the PCM temperature curve. Due to this, while using bromate moiety as a PCM, the cooking temperature (100 ^oC) was achieved with a delay of 2.5 h, which is maximum across the series. However, a relatively smaller value of heat of fusion did not extend the time required for achieving the ambient temperature for more than 3 hours. Thermal properties of the ILs containing nitrate, thiocyanate and bromide anions are shown in the **Fig. 4(e), (f)** and **(g)** respectively. All these PCMs have melting temperatures in the vicinity of 100 ^oC. The nitrate, melting at 98 ^oC, resisted the PCM temperatures above 100 ^oC resulting in reduction of the highest achievable temperature. The highest temperature achieved in this case was only around 100 ^oC. A handsome heat retention characteristic was exhibited by this PCM to an extent of around 4 h and 20 min.

A solid-solid phase transition observed at 60 ^oC was the characteristic feature of the thiocyanate T_{pcm} curve. During solidification, a significant amount of energy was released which in turn helped in maintaining about 5 ^oC higher temperature and extension in time required for achieving ambient temperature (3.5 h). While, the bromide based PCM recorded a smooth temperature rise and extended the time required for getting ambient temperature for 3 hours. Thermal characteristics of the iodate moiety shown in **Fig. 4(h)** indicated an isotherm in the range of 44 - 45 ^oC which was not clearly seen during solidification. The delay time recorded for obtaining the ambient temperature in this case was 4 h and 20 min.

Efficiency analysis

Energy analysis of the systems is necessary for determining the performance of the system **[5]**. Energy incident on the collector is given by:

$$Q_c = AI_s \tag{1}$$

where, A is the area of the reflector mirror.

The useful energy transmitted into the solar cooker is absorbed by the PCM and can be calculated by equ. (2) when the PCM is in the same phase and the equ. (3) is for PCM after the phase change **[29]**.

$$Q_f = m \cdot C_p \cdot \Delta T \tag{2}$$

$$Q_f = m \cdot C_p \cdot \Delta T + m \cdot \Delta H_m \tag{3}$$

Where 'm' is the mass of the PCM, ΔT is the temperature difference recorded in a time interval of 10 minutes and ΔH_m is the heat of fusion.

The efficiency of the system is defined as the ratio of the energy incident on the collector and energy absorbed by the PCM.

$$\eta = \frac{Q_f}{Q_c} \tag{4}$$

From the **eq. (4)**, it is clear that the heat capacity and the temperature difference observed during a fixed period of time have the greatest impact on the efficiency of the PCM. Time versus efficiency graphs of ILs are shown in **Fig. 5(a)** and **(b)**. In general, it was observed that the efficiency initially decreased with the passage of time and exhibited minima when the insolation was at its peak. This type of trend was observed as the reflector mirror was constantly maintained at an angle perpendicular to the base of the cooker. In the afternoon hours, the efficiency showed a gradual increase for some time and a sharp increase was registered around 3 pm. This sharp increase continued till a maximum was reached around 4 pm in all the cases.

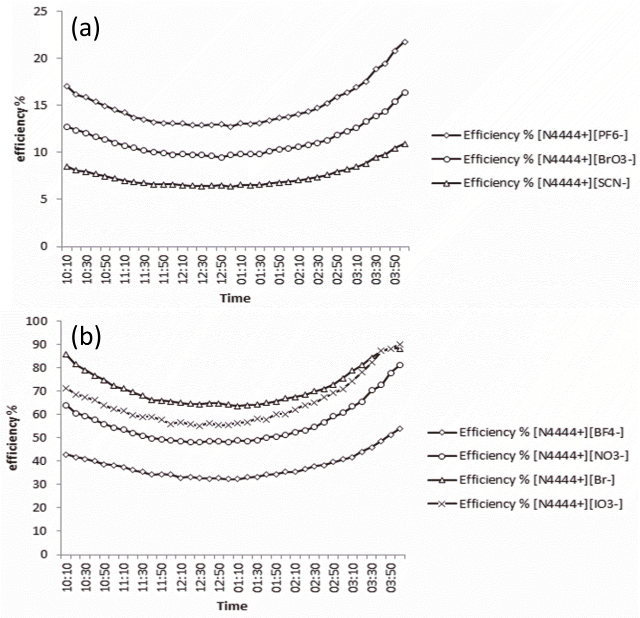

Fig. 5. Time versus Efficiency for (a) PF_6^-, BrO_3^- and SCN^- (b) BF_4^-, NO_3, Br^- and IO_3^- based ILs.

The iodate containing IL possesses the highest value of heat capacity; it registered a higher efficiency of about 83% and could successfully maintain the low end close to 55%. The ILs containing nitrate and bromide did not register efficiency less than 48% and were successful in achieving the efficiency levels much nearer to 90%. As expected, tetrafluoroborate moiety was the next most efficient PCM registering the minima above 33% and maxima touching to the value of 54%. The remaining ILs registered much poorer efficiency value. The fluorophosphates, bromate and thiocyanate containing ILs exhibited a minimum efficiency of 13%, 10% and 6% respectively. While, the maximum efficiency observed around 4 pm in these cases were 22%, 17% and 11% respectively.

Conclusion

The $[N^+_{4444}]$ based ILs when used as the PCM in solar cooker extended the time required for achieving cooking temperature by an average period of about two hours. They also exerted a delay in the occurrence of the ambient temperature in the cooker by 3-4 hours. This time gap can be utilized for late - hour - cooking as well as maintaining the food hot for a longer period.

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