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Experimental Investigation of Natural Convection Heat Transfer of Ionic Liquid in a Rectangular Enclosure Heated from Below

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ABSTRACT

This paper presents an experimental study of natural convection heat transfer for an Ionic Liquid. The experiments were performed for 1-butyl-2, 3-dimethylimidazolium bis(trifluoromethylsulfonyl)imide, ([C_4mmim][NTf_2]) at a Raleigh number range of $1.26x10^7$ to $8.3x10^7$. In addition to determining the convective heat transfer coefficients, this study also included experimental determination of thermophysical properties of [C_4mmim][NTf_2] such as, density, viscosity, heat capacity, and thermal conductivity. The results show that the density of [C_4mmim][NTf_2] varies from 1.437-1.396 g/cm^3 within the temperature range of 10-50°C, the thermal conductivity varies from 0.105- 0.116 W/m.K between a temperature of 10 to 60°C, the heat capacity varies from 1.015 J/g.K - 1.760 J/g.K within temperature range of 25-340°C and the viscosity varies from 18cp – 243cp within temperature range 10-75°C. The results for density, thermal conductivity, heat capacity, and viscosity were in close agreement with the values in the literature.. Measured dimensionless Nusselt number was observed to be higher for the ionic liquid than that of DI water. This is expected as Nusselt number is the ratio of heat transfer by convection to conduction and the ionic liquid has lower thermal conductivity (approximately 18%) than DI water.

Keywords: Ionic Liquid; Density; Viscosity; Heat Capacity; Thermal Conductivity; Convective Heat Transfer Coefficient.

1. INTRODUCTION

Environmental concern [1] and quick depletion [2] and soaring price [3] of conventional carbon based fuel pushes energy researchers to find reliable and economically viable alternate source of energy. Solar energy has already been proven to be a reliable and economically viable alternative source of energy [4]. Solar energy can be harvested either by direct conversion of solar energy into electric energy by photo voltaic solar cell [5] or it can be collected and transferred by means of a fluid known as solar collector. Solar cell faces the problem of lower efficiency [6] and cost effective ratio is very high [7] whereas solar collector posses superior performance than the solar cell [8]. In solar collector heat transfer fluid plays a very important role. High temperature stability and high heat storage capability are the critical factors for those heat transfer liquids. Currently used heat storage liquid has the low decomposition temperature and high melting point which results in high operating cost [9]. To meet the above requirements ionic liquids has great potential for replacement of the heat storage medium [10-11] of the solar collector.

Ionic liquids (IL) are the group of salts which are liquid at ambient temperature (less than 100°C) [12] and consists of ionic species. Typically IL contains large organic cations, such as imidazolium, pyrazolium, triazolium, thiazolium, oxazdium, pyridinium, pyridazinium, pyrimidinium, pyrazinium cations, and halogen, fluorinated or organic anions. These Room Temperature Ionic Liquids (RTILs) has the excellent physical and chemical properties including high thermal stability, exposure to air and moisture stability, low melting point, negligible vapor pressure [13-15]. For those excellent properties, ionic liquids become very useful for material processing [16], as a catalyst for synthesis of inorganic nano-materials [17], as lubricants [18], and in solar cell [10-11].

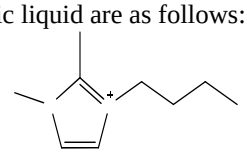
A bulk of studies already have been done for synthesis and basic physical properties such as density, viscosity, heat capacity, thermal conductivity, and surface tension study of ionic Liquids [19-21]. Most of the previous studies were focused on determining thermo-physical properties [22] and no study has been so far reported for natural convection which is the very fundamental heat transfer study of any liquid. This motivates the authors to study natural convection heat transfer of ionic liquids.

In the present work natural convection heat transfer experiments have been carried out for 1-butyl-2, 3-dimethylimidazolium bis(trifluoromethylsulfonyl)imide, ([C_4mmim][NTf_2]) ionic liquid and the study was performed in a rectangular enclosure heated form below and cooled from top. By measuring the top and bottom wall temperature and the applied power, the heat transfer coefficient was calculated. For greater fidelity of the reported results, thermal properties such as density, viscosity, heat capacity, and thermal conductivity of the Ionic Liquids were also measured and reported.

2. EXPERIMENTAL PROCEDURE

2.1 Ionic Liquid

99% pure 1-butyl-2,3-dimethylimidazolium bis(trifluoromethylsulfonyl)imide, ([C_4mmim][NTf_2]) Ionic Liquid was purchased from IoLiTec Company (Germany) . Molecular weight of [C_4mmim][NTf_2] is 433.39 g/mol. The chemical structure of the anion and cation and the molecular formula of the ionic liquid are as follows:



Cation

$(CF_2SO_2)_2N^-$
Anion

Molecular formula: $C_{11}H_{17}F_6N_3O_4S_2$

2.2 Measurement of viscosity

The viscosity of the ionic liquid was measured by using a cone and plate type rotary viscometer (LVDV-II+ProCP, from Brookfield Engineering Co.). The sample size of the cone and plate arrangement is 1mL. The cone and plate arrangement has a thermal jacket to maintain a constant sample temperature and it has the temperature accuracy within ±0.1^{o}C. For temperature control a thermal bath (Thermo NESLAB) was used with temperature accuracy within ±0.01K. The viscometer was calibrated by using company standard liquid. T each temperature three measurements were taken and the measurement uncertainty was calculated to be 1.6%.

2.3 Measurement of heat capacity

The heat capacity of ionic liquid was measured by using Differential Scanning Calorimetry (DSC Q200 from TA instruments Inc.). The ionic liquid sample was placed in a standard Tzero hermetic pan and the average sample size was 32.45 mg. The initial temperature was 25°C and ramp up a rate of 20°C/min up to 350°C. The ionic liquid was run four times and uncertainty has been calculated to be ±1.5%.

2.4 Measurement of thermal conductivity

Thermal conductivity was measured by using the KD2 Pro thermal property analyzer (Decagon Device, USA). The measurements principle is based on the transient hot wire method. The meter has a probe with 60 mm length and 1.3 mm diameter with a heating element and a thermoresistor which is inserted vertically into the test sample. The probe is connected with a microcontroller for controlling and conducting the measurements. Before using for ionic liquid the meter was calibrated with distilled water and company supplied standard glycerin. A thermal bath (Thermo NESLAB) was used to maintain a constant temperature of the measuring sample. The temperature accuracy of the bath is within ±0.01 K. For each measurement at least five readings were taken for each temperature and the uncertainty of the measurement has been calculated as ±3%.

2.5 Measurement of density and volume expansion coefficient

The density of ionic liquid has been measured using a 1 mL Pycnometer from Thomas Scientific. The pycnometer and the samples were placed in a thermal bath (Thermo NESLAB) to maintain a uniform temperature. The weight of the sample was measured by using METTLER TOLEDO balance which has a precision of 0.01 mg. Before using for Ionic Liquid the pycnometer was calibrated with water and was found to be accurate to within 0.5%. At each temperature the density measurements were repeated at least five times and the uncertainty of the measurement was ±2%. The volume expansion coefficient was calculated by using equation

$$\beta = -\frac{1}{\rho}\left(\frac{\partial \rho}{\partial T}\right)_P$$

2.6 Measurement of heat transfer coefficient

2.6.1. Experimental System

Figure 1(a) shows the rectangular cavity used as experimental test section, Figure 1(b) is the schematic of experimental setup. The experimental test section is a rectangular enclosure, made with clear polycarbonate Lexan sheet. It's length x width x height hasdimensions are 50mm x 50mm x 75 mm and 50mm x 50mm x 50mm at two different aspect ratios of the test section..

Figure 1(a): Experimental enclosure

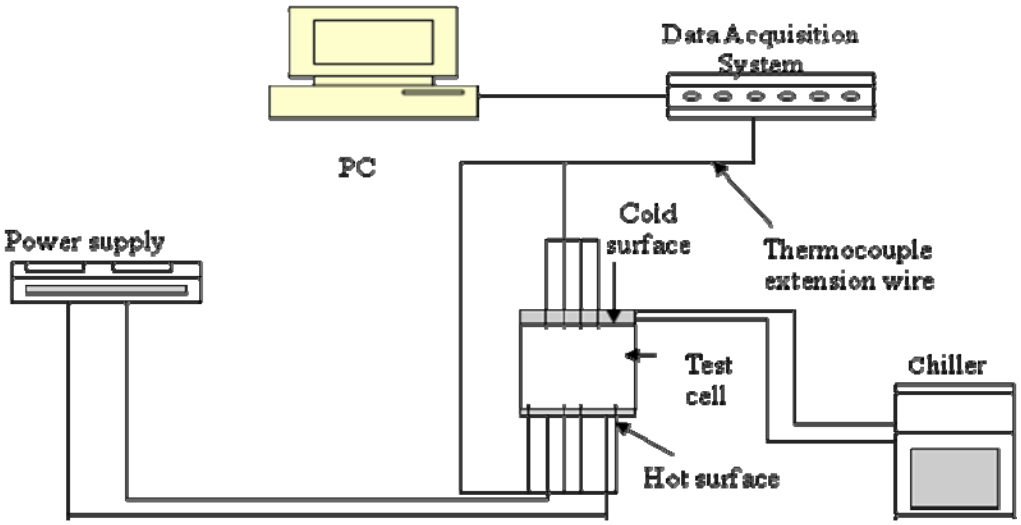


Figure 1(b): Schematic of experimental setup

The two ends of the test section enclosure are made with conductive copper sheets of thickness 3 mm, which are made to perform as hot and cold surfaces. There are two openings in the top copper sheet these are used as liquid filling ports. One hole is used for filling and the other is used for remove air bubbles from the enclosure. The top copper sheet is maintained at a uniform temperature by flowing cold water through a secondary enclosure of 25 mm height situated on top of the copper sheet. A flexible silicone rubber fiberglass insulated heater (20W, from OMEGA) is closely attached to the lower copper surface. The heating power is supplied from a DC power supply (120W, from MPJA Inc., 9312 PS). The heating and cooling surface temperatures are measured by using K-type thermocouples of 0.13 mm diameter (4 at hot and 4 at cold surface). There are two other thermocouples which are connected to the cold water inlet and outlet lines to measure the inlet and outlet temperatures of the cold water. All of the thermocouples are connected to a National Instrument (NI) data acquisition system cDAQ 9178 via a temperature cards NI 9211 which was interfaced with a computer and, Labview software was used for collecting and recording the data. The input voltage and current were measured from the display of the power supply. The whole system was insulated with the fiber glass to reduce the heat loss to the environment.

2.6.2. Data reduction

During the experiment the hot and cold surface temperatures were monitored and recorded until a steady state was reached. Fig. 2 shows the typical hot and cold surface temperature profile. In the experiment different Rayleigh number has been achieve by changing the heat flux.

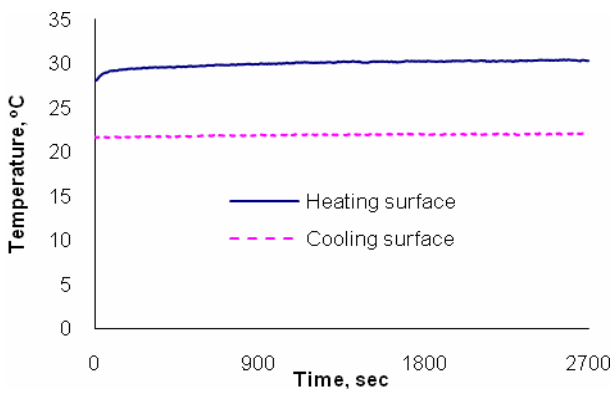


Figure 2: Initial transient hot and cold surface temperature

Heat flux, q'' was calculated from the input power of the heater divided by the surface area of the copper plate.

$$q'' = \frac{VI}{A}$$

Where, V is the input voltage, I is the input current, A is the surface area of the heater, which is same as the hot surface area. The inside of the hot surface T_{hf} and the cold surface T_{cf} temperature have been calculated from the thermocouple readings using 1D steady state heat conduction equation

$$T_{hf} = T_h - (\frac{q''}{k_c} x) \qquad T_{cf} = T_c + (\frac{q''}{k_c} x)$$

where T_h and T_c are the hot and cold surface temperature respectively, x is the thickness of copper plate, and k_c is the thermal conductivity of copper. Finally the heat transfer coefficient, h, was calculated by

$$h = \frac{q''}{(T_{hf} - T_{cf})}$$

2.7. Uncertainty and error analysis

The measurements uncertainty of thermocouples, voltage, and current are ±0.2°C, ±0.1V, and ±0.001A. A systematic uncertainty analysis was performed using standard Kline and McClintock method [23].

$$W_p = \sqrt{\sum_{i=1}^{n} \left(\frac{\partial p}{\partial a_i} w_i \right)^2}$$

where W_p is the total uncertainty of calculated parameter, P, and a_i variables of functional dependence, and w_i *is* the uncertainty of the independent variables. Input power uncertainty was calculated ±1.65%, heat transfer coefficient has an uncertainty of ±1.85%. Since the dimensionless numbers are the functions of numerous measured quantities and physical properties, therefore the uncertainty will propagate. The uncertainties associated with the Nusselt number and Rayleigh number were deermined to be ±2.29% and ±9.78% respectively.

3. RESULTS AND DISCUSSION

3.1. Viscosity of Ionic Liquid

Fig.3 shows the shear rate as a function of shear stress of [C_4mmim][NTf_2] ionic liquid at 25°C. The linear behavior of shear stress and shear rate proves the Newtonian behavior of ionic liquid.

The Fig.4 shows the shear viscosity as a function of temperature which indicates the strong temperature effect on the ionic liquid and the viscosity result can be represented by the following equation:

$$\mu = \exp\left[-8.517 + 3.964 \times \frac{1000}{T} \right]$$

The temperature dependent viscosity results correlates well with the literature [23], which measured the viscosity to be 127.18cP at 19.5°C.

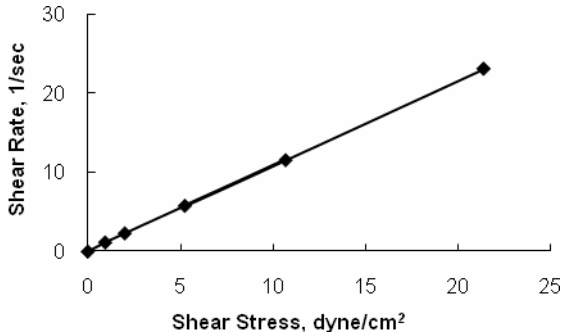


Figure 3: Shear rate as function of shear stress of $[C_4mmim][NTf_2]$ at 25°C

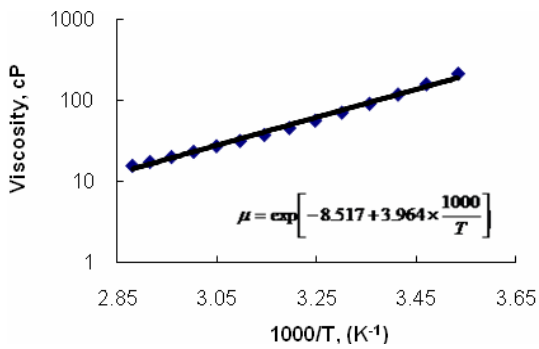


Figure 4: Viscosity of $[C_4mmim][NTf_2]$ as a function of temperature

3.2. **Heat capacity of Ionic Liquid**

Fig. 5 shows the heat capacity of $[C_4mmim][NTf_2]$ as a function of temperature. The heat capacity increases with temperature. Initially there is a sharp increment and after that the increments are linear. There is no clear explanation of the sharp rise in heat capacity. The linear temperature relation well correlates with the literature [19].

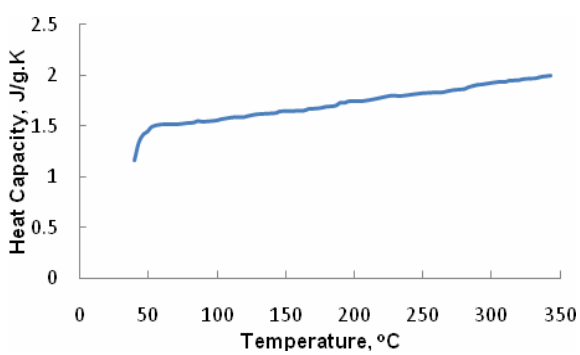


Figure 5: Heat capacity of $[C_4mmim]$ $[NTf_2]$ as a function of temperature

3.3. Thermal conductivity of Ionic Liquid

The Fig. 6 shows the thermal conductivity of $[C_4mmim]$ $[NTf_2]$ as a function of temperature within the temperature range of 10-60°C. Within this temperature limit the thermal conductivity varied from 0.105- 0.116 W/m.K which indicates that $[C_4mmim]$ $[NTf_2]$ has a relatively lower thermal conductivity and has thermal conductivity of approximately 18% of that of DI water at room temperature. From Fig. 6 it it is apparent that that within the temperature limit studied the thermal conductivity of $[C_4mmim]$ $[NTf_2]$ is not a strong function of temperature. It was noticed that Haisheng Chen et al., Rile Ge et al., and C. A. Nieto de Castro et al. [22, 24, and 25] used the same method and same device for different Imidazolium and Pyrrolidinium based Ionic Liquids. Haisheng Chen et al., [22] reported measured thermal conductivity for 1-butyl-3-dimethylimidazolium bis (trifluoromethylsulfonyl)imide, ($[C_4mim][NTf_2]$) and found its thermal conductivity to be 0.13 W/m.K up to a temperature of 40°C which matches well with the present value.

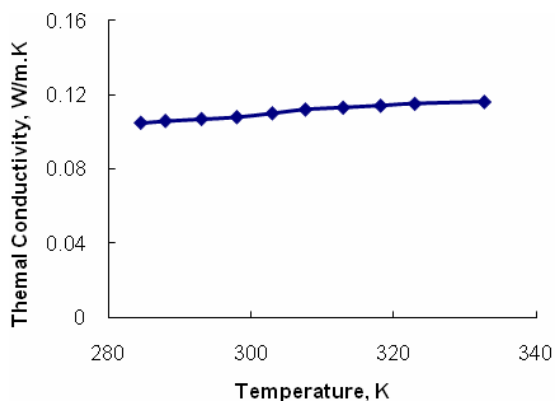


Figure 6: Thermal conductivity of $[C_4mmim][NTf_2]$

3.4. Density and volume expansion coefficient of Ionic Liquid

The fig. 7 and fig. 8 show the density and volume expansion coefficient of $[C_4mmim]$ $[NTf_2]$ as a function of temperature. The density decreases and volume expansion coefficient increases slightly with temperature increases. Here the density was measured within 10-50 °C and the density correlation with temperature was $\rho = 0.000809T + 1.6561$, where temperature is in Kelvin and density is in g/cm^3. I comparison Shoichi Katsuta et al. [23] reported the results for $[C_4mmim]$ $[NTf_2]$ Ionic Liquid with their correlation

of $\rho = 0.000812\,T + 1.658 \pm 0.002$, clearly the experimental result correlates well the literature.

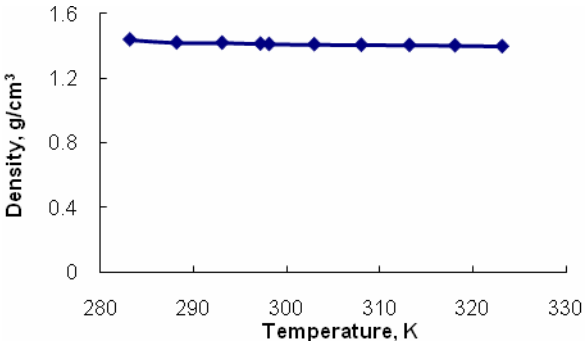


Figure 7: Temperature dependent density of $[C_4mmim][NTf_2]$

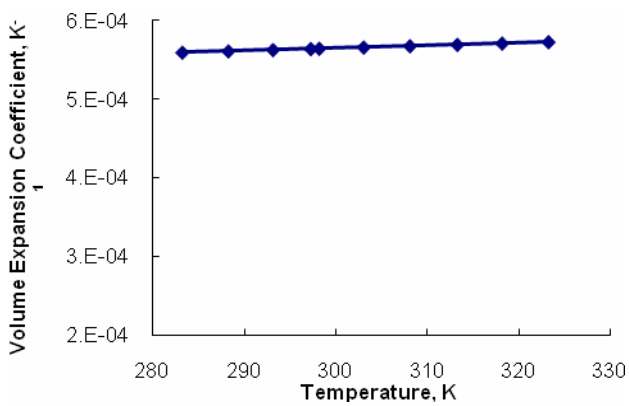


Figure 8: Temperature dependent volume expansion coefficient of $[C_4mmim]\ [NTf_2]$

3.5. Heat transfer behavior of Ionic Liquid

Before performing any experiment the test enclosure was rinsed thoroughly with DI water and dried. The liquid is poured into the test enclosure with care to avoid entrapment of any air bubbles into the enclosure. At first the experiment has been carried out for DI water and the results have been compared with the other published results [27-29] to ensure the credibility of our experimental setup and procedure. Nusselt number (Nu) as a function of Rayleigh number (Ra) for DI water is plotted and compared with that of the published result in Fig.9. The Nusselt and the Rayleigh number are computed with the following equations:

$$Nu = \frac{hL}{k_f} \qquad \Pr = \frac{\nu_f}{\alpha} \qquad Gr = \frac{g\beta\Delta TL^3}{\nu_f^2}$$

$$Ra = Gr.\Pr$$

Where, L is the height of the enclosure, k_f is the thermal conductivity, $\Pr$ is the Prandtl number, ν_f is the kinematic viscosity, α $(=\frac{k_f}{\rho C_p})$ is the thermal diffusivity, Gr is the Grashof number, β is the volume expansion coefficient, ρ is the density, C_p is the heat capacity of fluid, ΔT is the temperature difference between hot and cold surface, g is the gravitational acceleration, All the fluid properties were evaluated at the average of the hot and the cold surface temperature. The natural convection correlation can be represent as, $Nu = cRa^n$ where c and n are the empirical constants. The fig. 9 shows that the experimental result and reference result has the same trend, there appears to be difference in the value of empirical constants. Those constants depend on the geometry of the enclosure, and heating condition.

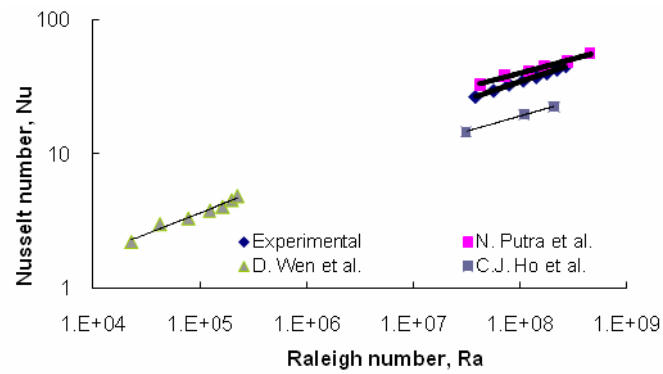


Figure 9: Comparison of experimental and published result for natural convection of water

The heat transfer coefficient of ionic liquid as a function of input power is presented in fig. 10. The fig. 10 shows that the heat transfer coefficient of ionic liquid is lower (approximately 16%) than DI water and heat transfer coefficient increases with increases input power. The lower heat transfer coefficient of ionic liquid indicates that the ionic liquid molecules moves slowly with density variation due to temperature difference, these may happened because of the higher viscosity of ionic liquid. It may also happen due to lower thermal conductivity of ionic liquid which influences on the thermal diffusivity of ionic liquid. At same temperature ionic liquid has 2.5 times lower thermal diffusivity than DI water. It is also clear at higher power the temperature of the hot surface become high and the ionic liquid viscosity goes down, which gives the higher heat transfer coefficient.

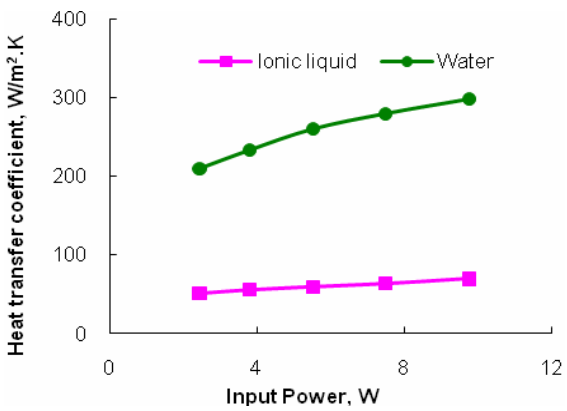


Figure 10: Heat transfer coefficient as a function of input power

The Nusselt number as a function of input power is presented in fig. 11 and it can be noticed that the Nusselt number of ionic liquid is higher than DI water, which is expected because thermal conductivity ratio of water to ionic liquid is approximately 5.68 (0.613/0.108) and the heat transfer coefficient ratio of water to ionic liquid is approximately 4. That means by the Nusselt number of the ionic liquid should be approximately 1.42 (5.68/4) times higher than DI water.

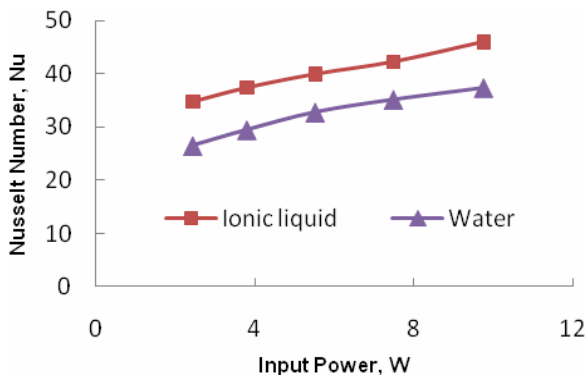


Figure 11: Nusselt number as a function of input power

Fig. 12 shows the Nusselt number as a function of Raleigh number, it can be noticed that at the same Raleigh number ionic liquid has the higher Nusselt number. The higher Nusselt number indicates that the convection to conduction heat transfer ratio is higher. From fig. 10 it is clear that ionic liquid has lower convective heat transfer coefficient which implies that the higher Nusselt number was obtained from the much lower thermal conductivity of ionic liquid.

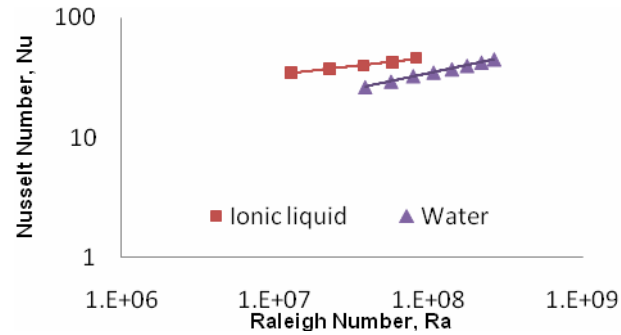


Figure 12: Nusselt number as a function of Raleigh number

4. CONCLUSIONS

Natural convection study of [C_4mmim][NTf_2] ionic liquid has been performed for the first time and at the same time the properties of [C_4mmim][NTf_2] was also measured. The following conclusions can be drawn:

- The thermal conductivity of [C_4mmim][NTf_2] ionic liquid varies from 0.105- 0.116 W/m.K over the temperature (10-60°C) range studied, which is approximately 18% thermal conductivity of DI water.
- Heat capacity of [C_4mmim][NTf_2] ionic liquid varies from 1.015 J/g.K - 1.760 J/g.K within temperature range of 25-340°C.
- The ionic liquid [C_4mmim][NTf_2] shows a Newtonian fluid behavior and high temperature dependency where viscosity decreases with increasing temperature.
- The convective heat transfer coefficient of ionic liquid is lower than DI water at same heat input.
- The dimensionless Nusselt number and Raleigh number relation has the same trends for water and ionic liquid.
- The dimensionless Nusselt number of ionic liquid is higher than DI water at same experimental condition.

Acknowledgement

The financial support for this research came from Department of Energy (DOE) through Savannah River National Laboratory (SRNL); contact # . The author would like to thank Dr. Guiren Wang to give opportunity to use the lab facilities to measure density, and Shakena Daniel from Dr. John Lavigne group for helping to measure heat capacity. The DOE-EERE Solar Energy Technology Program is gratefully acknowledged for funding of this work. Savannah River National Laboratory is managed by Savannah River Nuclear Solutions. This work was prepared under Federal Contract DE-AC09-08SR22470. The United States Government retains, and by accepting the article for publication the publisher

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