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Possible New Heat Transfer Fluid: The IoNanofluid of 1-Ethyl-3-methylimidazolium Dicyanamide + Nano-Titanium Oxide—Studying Its Thermal Conductivity and Viscosity

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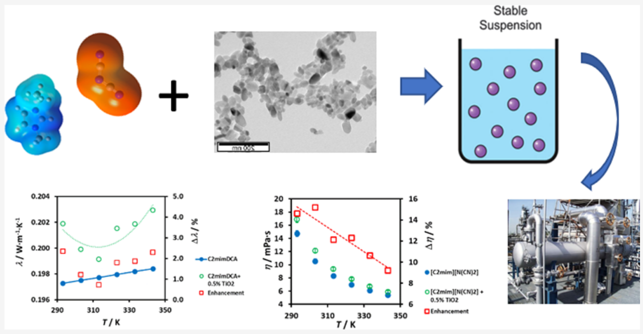

ABSTRACT: Ionic liquids with the dicyanamide anion, namely, with 1-alkyl-imidazolium cations, have been receiving attention recently due to their potential applications. The utilization of these liquids as heat transfer fluids, specifically in small heat exchangers and microchannels for microprocessor cooling, is presently deemed highly feasible, as it can be both more efficient and environmentally acceptable. The design of a heat transfer equipment that makes use of fluids requires knowledge of their thermophysical properties. In this regard, dispersions of nanoparticles have been extensively studied in recent years to improve thermal conductivity or obtain desirable optical properties. IoNanofluids is what we have taken to name the result of such dispersions in ionic liquids. In this paper, we report measurements of the thermal conductivity and viscosity of the IoNanofluid of 1-ethyl-3-methylimidazolium dicyanamide, $[C_2mim][N(CN)_2]$, with 0.5% mass fraction of TiO_2 nanoparticles (diameter 20 nm) in the temperature range ($293 < T/K < 343$), at P = 0.1 MPa. Reasonable enhancements were found for thermal conductivity and viscosity, which were temperature-dependent. The IoNanofluid was found to behave as a non-Newtonian fluid in most of the temperature range studied. A discussion about the possible use of this IoNanofluid as a heat transfer fluid shows that it has very promising properties to be used in heat transfer applications.

1. INTRODUCTION

Ionic liquids have emerged as compelling components within the realm of alternative engineering fluids, boasting distinctive properties that render them enticing candidates for replacing solvents, heat transfer fluids, and homogeneous catalysts. The imperative need for environmentally acceptable, nontoxic solutions with enhanced energy efficiency underscores the significance of exploring their diverse applications. Rooted in their chemical structure, characterized by ionic melts at low to medium temperatures, these liquids embody a synergistic interplay of Coulombic forces and polarizable electron clouds in both cations and anions.

Their potential utility in novel chemical processes and cooled microsystems employed by the semiconductor industry, and advanced battery technologies accentuate the critical importance of comprehending their thermophysical properties. Typically investigated at low pressures and over extensive temperature ranges, particularly around room temperature, the exploration of ionic liquids becomes pivotal.

In the context of optimizing heat transfer applications, the focus on thermal conductivity and viscosity assumes prominence in determining heat transfer coefficient enhancement.[1] This quest for efficiency has spurred innovative approaches such as the utilization of nanoparticles in IoNanofluids.[2−6] However, the incorporation of nanomaterials in IoNanofluids and nanofluids at large necessitates careful consideration of various constraints, encompassing potential impacts on human health and concerns related to disposal, reusability, and recyclability. The judicious selection of nanomaterials becomes paramount, extending beyond technical efficacy and availability.[7] A case in point is titanium oxide, the chosen material in this study, which prompts health concerns, especially in food products (namely, sweets, baked goods, or chewing gum). As E171, its use has been banned in the EU due to potential damage to intestinal flora and carcinogenic risks. Consequently, for laboratory or industrial

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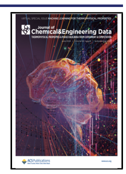

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Table 1. Materials Specifications: In Mass Percentage, When Applicable

	identity (NMR, IR)	assay (NMR)	cation (IC)	anion (IC)	halides (IC)	water (KF)/ppm	appearance	CAS N#
$[C_2mim][N(CN)_2]$ batch: M00450.1.2-IL-0003	pass	>98%	98.7%	98.9%	<2%	1850	yellow liquid	370865−89−7
			purity	phase	particle diameter/nm			
titanium oxide (TiO_2) batch: HNO046007			99.9%	rutile	10−30 (average 20)		white powder	04−03−2011

applications, careful handling and preventive measures against skin contact and powder inhalation are imperative when employing such materials.

Our examination of the most promising ionic liquids for use in heat transfer fluids, either in their pure form or as IoNanofluids, led us to focus on nanosystems involving the ionic liquid dicyanamide $[N(CN)_2]^-$ in conjunction with multiwalled carbon nanotubes (MWCNT). The viscosity values of ionic liquids from this family were identified as particularly noteworthy, ranging between 0.02, and 0.07 Pa·s. These values fall within a desirable range, avoiding the criticism associated with liquids exhibiting excessively high viscosity around room temperature.[8] To explore the impact of nanoparticles on the properties of the ionic liquid in a stable dispersion and identify potential enhancements, we conducted measurements on the thermal conductivity and viscosity of the IoNanofluid with 0.055 mass fraction of TiO_2 nanoparticles (20 nm diameter) within the same temperature range, at P = 0.1 MPa.

2. EXPERIMENTAL SECTION

2.1. Materials. 1-Ethyl-3-methylimidazolium dicyanamide, $[C_2mim][N(CN)_2]$, was purchased from IoLiTec (Product N# IL-0003-HP), Ionic Liquids Technologies, Germany, and the product specifications of the certificate analysis are described in Table 1.

Titanium oxide for the preparation of the TiO_2 NFs (rutile, 10−30 nm, 99.9%, from IoLiTec; NO-0046-HP; LOT: HNO046007). A complete analysis by energy-dispersive system (EDS), scanning electron microscopy (SEM), and transmission electron microscopy (TEM), performed at the MICROLAB−Electron Microscopy Laboratory, ULisboa (https://microlab.ist.utl.pt/), with FEG/EDS/EBSD JEOL 7001F/Oxford 250/HKL INCA Energy and with TEM/EDS Hitachi H8100/ThermoNoran System SIX confirmed the manufacturer's purity. They also showed that the nanoparticles have an average diameter of 20 nm for 11 determinations, confirming the manufacturer's characteristics. Nevertheless, the images also reveal evidence of particle aggregation, a phenomenon that could be mitigated through judicious sonication methods,[7] as elucidated in the following section.

2.2. Sample Preparation. The ionic liquid was dried under vacuum for 48 h and sealed in a flask for further measurements of its properties. The water content was determined by coulometric Karl Fischer titration (Metrohm 831) (Table 2). IoNanofluids were prepared by a two-step method,[9] adding the nanoparticles of TiO_2 using a sonicator (Hielscher, UP200HT), with a probe S26d40, optimized for an amplitude of 70%, a pulse cycle of 70% (max. Eleven W), and a frequency of 25 ± 1 Hz for 2 + 1.5 min. No surfactant was added to provide additional stabilization. To avoid a temperature rise of the IoNanofluid due to the sonication, the sample container was cooled with ice. The IoNanofluids

Table 2. Water Content in $[C_2mim][N(CN)_2]$ Samples, Used for IoNanofluids Preparation

sample	w_w/ppm[a]	w_w/ppm[b]
viscosity	625 ± 82	5863 ± 208
thermal conductivity	485 ± 75	10,468 ± 256

[a]Before measurements, dried. [b]After measurements.

were stable during the whole measurement period (4 months) and found stable in the laboratory flask after 2 years.

2.3. Methods of Measurement. *2.3.1. Thermal Conductivity.* Thermal conductivity, λ, measurements were performed at temperatures 295.06 < T/K < 345.55, using commercial equipment from Hukseflux Thermal Sensors TPSYS02 and a non-steady-state probe (NSSP) TP08, SN# 283, with a temperature standard uncertainty of $u(T)$ = 0.02 K, as reported earlier.[10] Measurements with water permitted the validation of the probe operation, as described in a recent publication,[11] with data obtained between 300 and 350 K not deviating from the standard reference data value (IUPAC SRD correlation) by more than 0.5%, as obtained before. Data for different power inputs showed no indication of convection in the cell, proving that the theory of the transient hot-wire technique[12] could be used to obtain the thermal conductivity of the ionic liquid and its mixtures with water. The authors are called to references[10−12,20,46] to find details on the validation of measurements in ionic liquids and water with the present equipment. These studies justify an estimated expanded relative uncertainty U_r (λ) = 0.02, at a 95% confidence level (k = 2) for the data herein reported.

2.3.2. Viscosity. An AR 1500EX rheometer from TA Instruments was used to measure the viscosity of the IoNanofluids, η, for 293.15 < T/K < 343.15. This model is a controlled stress, strain, and rate rheometer with a wide torque range [0.0001−150 mN·m], ultrahigh resolution [0.04 μ rad], and high angular velocity [300 rad/s], with a standard 0.5°, 40 mm cone geometry, and a gap of 1150 μm. A Peltier plate allows testing to be done from 263 to 473 K with a standard uncertainty u (T) = 0.1 K. Newtonian behavior was observed in this ionic liquid, as the viscosity has a constant value with varying shear rate in the interval of temperatures studied and in the shear rate range 0.01−562 s^{-1}. The expanded relative uncertainty of the measurements at a 95% confidence level (k = 2) was found to be U_r (η) = 0.03, except for the lowest temperature (293 K), where a value of 0.06 translates better the deviations found with data existing in the literature (please refer to Section 3).

3. RESULTS AND DISCUSSION

The dispersion of nanomaterials in ionic liquids has the effect of changing the values of the thermal conductivity, as we have shown for several ionic liquids recently[6] and in several papers in the past, as mentioned before.[2,4,5,8,13] This effect is

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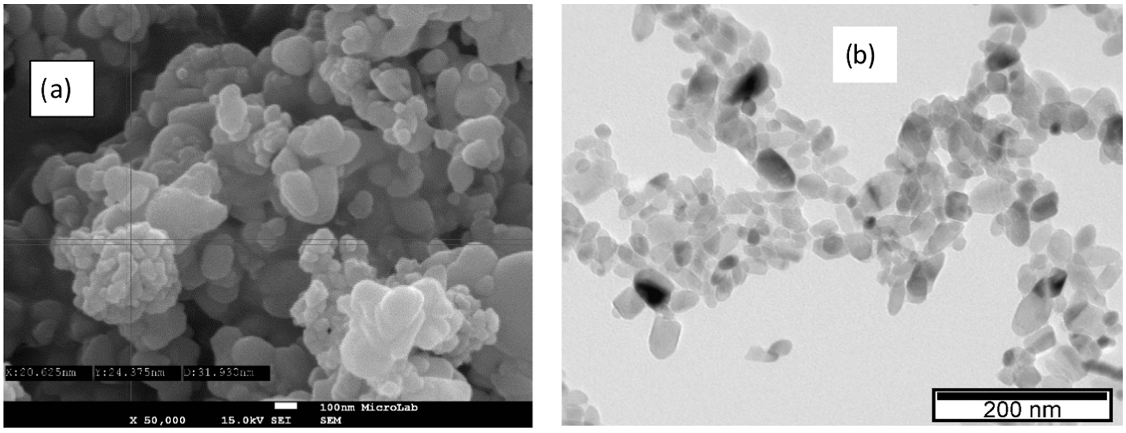

Figure 1. Morphological characterization of the nano-TiO_2. (a) SEM image of the nano powder before producing the IoNanofluid; (b) TEM image of the nanoparticles after producing the IoNanofluid. Dimensions can be seen on the SEM image: X = 20.625 nm; Y = 24.375 nm; D = 31.930 nm.

Table 3. Thermal Conductivity[a], λ, for $C_2mim[N(CN)_2]$ + TiO_2 IoNanofluid from T = (295.06 to 345.55) K, at P = 0.1 MPa[a]: Mass Fraction of TiO_2 Nanoparticles w_{np} = 0.005

T/K	λ/W·m^{-1}·K^{-1}	T_{nom}/K	λ_{INF}/W·m^{-1}·K^{-1}	λ_{pure}[b]/W·m^{-1}·K^{-1}	$\Delta\lambda$/W·m^{-1}·K^{-1}	$\Delta\lambda$/%
295.06	0.2016	293.15	0.2019	0.1973	0.0046	2.3
304.32	0.1989	303.15	0.1999	0.1975	0.0024	1.2
314.43	0.1988	313.15	0.1991	0.1977	0.0014	0.7
324.38	0.2007	323.15	0.2015	0.1980	0.0036	1.8
334.46	0.2021	333.15	0.2019	0.1982	0.0037	1.9
345.55	0.2037	343.15	0.2029	0.1984	0.0045	2.3

[a]Standard uncertainty $u(T)$ = 0.02 K, $u(P)$ = 1 kPa; expanded relative uncertainty U_r (λ) = 0.02, at a 95% confidence level (k = 2). [b]Calculated at the nominal temperatures from eq 1.

translated in thermal conductivity enhancement concerning the original base fluid, which varied from around 2 to 20% for multiwalled carbon nanotubes, MWCNTs, for NPs mass concentrations of 0.5 to 3%.

The morphology of the nanomaterials used, namely, for CNTs has been studied recently by Jóźwiak et al.[14] and Dzida et al.[15] who concluded that "the morphology, physical chemistry of nanoparticles and the IL-nanostructure interactions on the mechanism of heat transfer and rheological properties of IoNanofluids (INFs)···" strongly affects the thermal conductivity enhancement of IoNanofluids (INFs). The longer (and better defined) the carbon nanotube, the higher the enhancement (44% for 770 μm), possibly caused by the existence of three-dimensional (3D) networks with thermal bridges in the whole INF volume, the "cobra-like macromolecular architectures."

It must be stated that the measured thermal conductivity of an IoNanofluid (and also of any nanofluid) is an effective (or apparent) thermal conductivity and not a true thermal conductivity, as these nanosystems are at least biphasic and therefore heterogeneous. However, these authors identified what seemed to be one of the most sensitive phenomena in heat transfer in the last century: the enhancement of thermal conductivity of the media concerning the original base fluid by small amounts (concentration) of dispersed nanoparticles. The behavior of these systems was also analyzed theoretically by using models to estimate the thermal conductivity of the base ionic liquids and that of the IoNanofluids,[16] although the results proved our limitations in the understanding of the molecular mechanisms of heat transfer, both in ionic liquids and IoNanofluids.[16,17] Recently, a new method was reported for the production of ultralong carbon nanotubes with ultrahigh yields by Jiang et al.[18] with lengths over 1 cm, capable of future mass production. It is envisaged to prepare INFs with this type of carbon nanotubes to produce very high thermal conductivity enhancements, and we hope to report on these new systems soon.

However, and as mentioned in a recent publication,[12] the measurement of the thermal conductivity of ionic melts is difficult, mostly because the liquids are electrically conducting, and done many times with high uncertainty, as we did in the past in our laboratory (between 6 and 11%). In recent measurements for $[C_2mim][CH_3SO_3]$[10,19] and for $[C_6mim][(CF_3SO_2)_2N]$[20] using equipment based on the transient hot-wire technique, with an expanded uncertainty of 2%, as described in Section 2.3.2, it was possible to obtain data with better uncertainty.

Measurements of the viscosity of nanofluids/IoNanofluids are fewer than those with thermal conductivity, and quantitatively, we know that the nanomaterials dispersed can increase the viscosity to an extent that it can limit the possible technical applications of these systems.[3] Recently, Murshed and Estellé[21] Shakeel et al.[22] and Jóźwiak and Boncel[23] reviewed the viscosity behavior of nanofluids and IoNanofluids, calling to attention the fact that several INFs are non-Newtonian, as we mentioned before[3] for $[C_4mpyrr][N(CN)_2]$, with a shear thinning behavior. Although in many cases, the dispersion of nanomaterials increases the viscosity, some cases were reported where MWCNTs and graphene decreased the viscosity, as reported by Liu et al.[24] and Wang et al.[25]

Several studies have been performed with other nanoparticles dispersed in ionic liquids. Not intending to be exhaustive, some examples for metal oxides like TiO_2,[26−28]

ZnO,[29] Al_2O_3,[30−32] SiO_2,[33−36] MgO,[31] metal particles, like Ag,[37] Ru[38] and others,[39] and for graphene,[24,40] can be referred.

The structure of these systems is complex, and a general overview can be found in the papers by Shakeel et al.[41] and He and Alexandridis.[42] In this last paper, and also according to our previous discussion on the thermal conductivity of ionic liquids and IoNanofluids of MWCNTs, it is important to understand the type of forces between the ionic liquid cation and anion, between the ions and the nanoparticles, and their influence in the organization of the dispersion. However, as this discussion on the structure needs more information about the dependence of the enhancements on the mass fraction and on the interphase $nTiO_2$−IL, which is not available in this work, we report here only our experimental results and compare with some similar systems data available in the literature.

The specifications of nano-TiO_2 are displayed in Table 1. In addition to the manufacturer's information, SEM and TEM images were obtained before and after producing the IoNanofluid. Figure 1 displays both images. These images support the fact that no significant morphology variation of the nanoparticles is found with the suspension sonication procedure and that the dimensions of the particle are between 10 and 30 nm, with an average diameter of 20 nm, as per the manufacturer's statement.

The results obtained for the thermal conductivity of the $[C_2mim][N(CN)_2]$ + TiO_2 nanofluid are shown in Table 3, for the experimental (or reference) temperatures and the nominal temperatures, corrected accordingly to the eq 1, obtained with an extensive investigation of the thermal conductivity of $[C_2mim][N(CN)_2]$, and its mixtures with water:[43]

$$\lambda(T) = a_0 + a_1(T/\mathrm{K}) \tag{1}$$

with the coefficients $a_0 = (0.19059 \pm 0.00517)$ $W\cdot m^{-1}\cdot K^{-1}$ and $a_1 = (2.278 \pm 1.605) \times 10^{-5}$ $W\cdot m^{-1}\cdot K^{-2}$. The root-mean-square variation of the fit is 0.0012 $W\cdot m^{-1}\cdot K^{-1}$, and no point deviates from the fit by more than 0.0020 $W\cdot m^{-1}\cdot K^{-1}$. The values at nominal temperatures were calculated assuming that the temperature coefficient of the thermal conductivity was the same as found for the pure ionic liquid. The enhancement in the thermal conductivity is calculated as the difference between the thermal conductivity of the IoNanofluid, λ_{INF}, and that of the pure ionic liquid, λ_{pure}, obtained from eq 1. The enhancement varies between 0.7, and 2.3%, increasing slightly with temperature, as plotted in Figure 2. This result, obtained for a low mass concentration of the nanoparticles, is of the same order of magnitude as that obtained with the MWCNTs IoNanofluid (same mass fraction),[8] spherical nanospheres (organized heat transfer paths) competing with a random array of heat transfer paths in carbon nanotubes. If we increase the mass fraction of the nanomaterials, the enhancement will be in principle greater.

As explained in Section 2.3.1, the measurements of viscosity were performed with a rheometer.[6,18] Therefore, it is possible to measure the sheer stress as a function of the shear rate, as a function of temperature. This allowed us to verify the viscosity enhancement in the IoNanofluid, as well as the fluid behavior. The general relation between the shear stress and the shear rate, $\dot{\gamma}$, has the form, eq 2

$$\tau = \tau_0 + k\dot{\gamma}^n \tag{2}$$

where τ_0 is a residual shear stress (at very low shear rates) necessary to apply for the fluid to flow, k is a proportionality

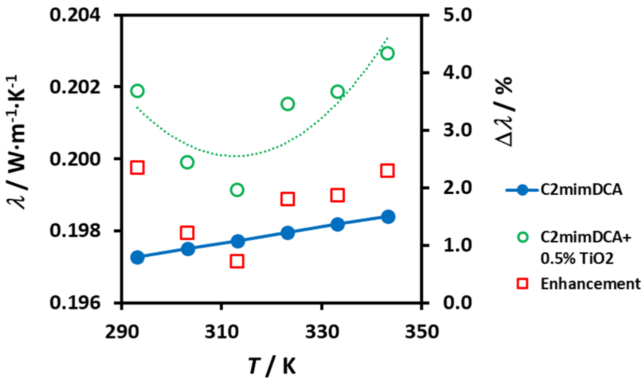

Figure 2. Thermal conductivity for $[C_2mim][N(CN)_2]$ + TiO_2 IoNanofluid as a function of temperature. Also shown for comparison is the thermal conductivity of the pure ionic liquid, obtained from eq 1, and the thermal conductivity enhancement for the TiO_2 0.5% IoNanofluid.

factor, and n is the exponent of the variation law. This equation is in principle valid for all of the stress rate spectrum, from 0 to the maximum stress that the material can support before it collapses. For $\tau_0 > 0$, as found for $[C_2mim][CH_3SO_3]$,[28] the fluid is said to have a Bingham plastic behavior, while for $\tau_0 = 0$, the fluid has a non-Newtonian shear thinning behavior, with shear viscosity decreasing with increasing shear rate and is called a pseudoplastic, the phenomena being called viscoelasticity. In the case where $n = 1$, shear stress is proportional to shear rate, the proportionality factor k being equal to the dynamic viscosity, η.

Figure 3 displays the viscosity (as the ratio between shear stress and shear rate) as a function of the shear rate for

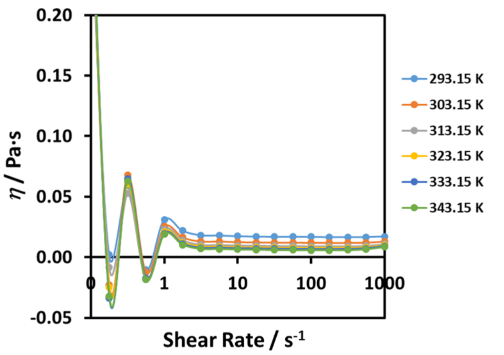

Figure 3. Viscosity (as the ratio between shear stress and shear rate) as a function of the shear rate for $[C_2mim][N(CN)_2]$ + TiO_2 IoNanofluid, for the different temperatures studied.

$[C_2mim][N(CN)_2]$ + TiO_2 IoNanofluid, for the different temperatures studied. It is clear that there is a stress relaxation response for very low shear rates (<1 s^{-1}) typical of the laboratory realization of the measurement[44] and that, above 4 s^{-1}, an approximately constant value of the viscosity is obtained for all temperatures, permitting the calculation of the dynamic viscosity, and it can be assumed that our IoNanofluid (and the pure ionic liquid) are Newtonian fluids, for, $\dot{\gamma} > 4$ s^{-1}. However, above 200 s^{-1}, the apparent viscosity starts to be nonconstant, increasing with the shear rate.

A clearer analysis can be shown in Figure 4, whereby the shear rate is plotted as a function of the shear stress for all of the studied shear rate ranges (up to 1000 s^{-1}). Only for the

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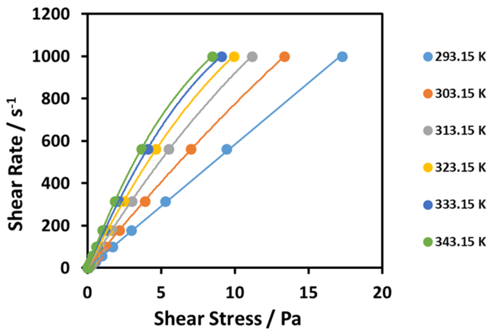

Figure 4. Shear rate as a function of the shear stress for the different temperatures studied. The higher the temperature, the smaller the temperature range where the IoNanofluid can be considered Newtonian.

lower temperature, 293.15 K, there is a proportionality between the two physical quantities (and therefore a Newtonian fluid behavior), and $n = 1$. Increasing the temperature, the value of n starts to be smaller than one, and therefore, the system is a pseudoplastic. Therefore, the value of the viscosity was calculated for $10 < \dot{\gamma} < 200\ s^{-1}$, with an average standard deviation of 0.17 mPa·s. Values for each temperature are displayed in Table 4.

The results obtained for the viscosity of the $[C_2mim][N(CN)_2]$ + TiO_2 IoNanofluid are shown in Table 4, together with the values obtained for the pure fluid, for the temperatures 293.15 to 343.15 K. The values obtained for the pure fluid can be compared with previous data available in the ILTHERMO database from NIST.[45] There are 15 reported works, using several measuring methods, like falling or rolling sphere viscometry, used by Zheng et al.[46] Fatima et al.[47] Lepre et al.[48] Vatascin and Dohnal[49] and Larriba et al.[50] cone and plate rheometry, used by Hothar et al.[32] Pamies et al.[51] and Stoppa et al.[52] concentric cylinders viscometry used by Neves et al.[53] and Freire et al.[54] capillary viscometry, used by Larriba et al.[55] Quijada-Maldonado et al.[56] and Schreiner et al.[57] and by using two methods with higher uncertainty, like the pressure drop in a flow channel, used by Huang et al.[58] and the moving piston method, used by Mei et al.[59] Figure 5 shows all of the data reported, as a function of temperature. It is clear that there is a high dispersion of the data and that our data agrees within the mutual uncertainty with other data, except for the lower temperature (293 K), whereby our claimed relative uncertainty seems to be around 0.06, instead of 0.03 for $T > 293$ K, probably due to a deficient equilibration of the rheometer cone and plate. Other authors claimed uncertainties oscillate between 3 and 10%, and some of them do not report a

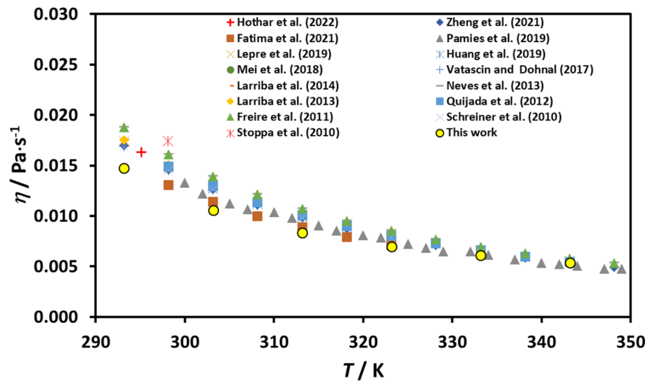

Figure 5. Comparison with reported data for the viscosity of $[C_2mim][N(CN)_2]$, with previously published data.[45−60]

detailed characterization of the samples used, namely, the water content, which might explain the deviations found.

Analyzing the results obtained for the IoNanofluid, the dispersion of TiO_2 in $[C_2mim][N(CN)_2]$ increases the viscosity of the system. These values are displayed in Figure 6. The enhancement of the viscosity of the IoNanofluid

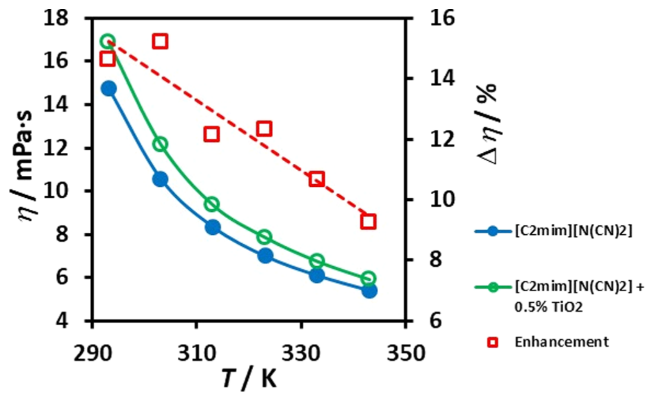

Figure 6. Viscosity of $[C_2mim][N(CN)_2]$ + TiO_2 IoNanofluid as a function of temperature. Also shown for comparison are the viscosity of the pure ionic liquid and the viscosity enhancement for the TiO_2 0.5% IoNanofluid. The dashed line was only drawn for eyesight view.

decreases significantly with the increase in temperature, being of the order of 15−9%. This result is quite interesting because, in conjunction with the increase in the thermal conductivity enhancement with temperature, it might contribute to an increase of the heat transfer coefficients (inner and outer) of a shell and tube type heat exchanger as a function of temperature, as illustrated by França et al.[1] an excellent

Table 4. Viscosity[a], η, for $C_2mim[N(CN)_2]$ + TiO_2 IoNanofluid from T = (293.15 to 343.15) K, at P = 0.1 MPa[a]: Mass Fraction of TiO_2 Nanoparticles w_{np} = 0.005; σ, the Standard Deviation of the Viscosity Determination, is Also Shown

T/K	η_{INF}/mPa·s	σ/mPa·s	η_{pure}/mPa·s	σ/mPa·s	$\Delta\eta$/mPa·s	$\Delta\eta$/%
293.15	16.89	0.17	14.74	0.12	2.15	14.6
303.15	12.15	0.14	10.55	0.05	1.60	15.2
313.15	9.34	0.17	8.33	0.03	1.01	12.1
323.15	7.84	0.19	6.98	0.05	0.86	12.3
333.15	6.73	0.18	6.09	0.06	0.65	10.6
343.15	5.89	0.17	5.40	0.07	0.50	9.2

[a]Standard uncertainty $u(T)$ = 0.02 K, $u(P)$ = 1 kPa; expanded relative uncertainty U_r (η) = 0.03, at a 95% confidence level (k = 2), except for 293 K, where the reported data point has an estimated expanded relative uncertainty U_r (η) = 0.06.

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J. Chem. Eng. Data 2024, 69, 2227−2235

promising result as a heat transfer fluid, as it will not increase the fluid pumping requirements.

As previously reported,[20] for base liquids other than ionic liquids, with TiO_2 nanoparticles, the temperature has a mixed effect (increase, decrease, and a combination of both) on the viscosity of these nanofluids. Unfortunately, there is no discussion for IoNanofluids for the effect of temperature on the viscosity enhancement of TiO_2 INFs, although, for alumina nanoparticles inf [C_2mim][CH_3SO_3], it has been reported that a Vogel−Fulcher−Tammann (VFT) equation, identical to that used for the base ionic liquid, could be used (viscosity decreasing with temperature).[30] No data values were published that permit the calculation of the variation of the enhancement with temperature. Witmar and co-workers[27,28] have shown that for a hydrophobic ionic liquid, like [C_2mim] [$(CF_3SO_2)_2N$] with 0.5% w/w TiO_2, the dispersion of the nanoparticles was not very good, as they aggregated, while for [$C_n$mim][BF_4] (n = 2, 4, and 6), hydrophilic ionic liquids, a better dispersibility was obtained, the longer the alkyl chain the better, leading to a narrowing of the particle size distribution and a decrease of the agglomerate size in dispersion. From the rheological point of view, it was found that the ionic liquid [C_6mim][BF_4] was least affected by nanoparticle addition, and [C_4mim][BF_4] was the most affected by it. Several types of preparation methods were used for the nanoparticles, obtaining different nanoparticle dimensions, and it was found that small nanoparticles tend to agglomerate, especially when introduced in higher amounts, indicated by the higher viscosities at low shear rates. Similar behavior was found for ZnO IoNanofluids. Although it is not possible to compare these results for other INFs, it can be said that our ionic liquid is also hydrophilic and that the nominal size of our nanoparticles is 20 μm, a value that falls within the dimensions used by Witmar and collaborators' work,[27] 5.7 and 8.7 μm for samples prepared by chemical vapor synthesis (CVS), a modified chemical vapor deposition (CVD) process,[60] and 26.4 nm for a commercial sample (Sigma-Aldrich). Recently, Hothar et al.[32] reported data on the rheological behavior of [C_2mim][$N(CN)_2$] and [C_2mim][$C(CN)_3$] + Al_2O_3 and MgO IoNanofluids, the only work that can be used to compare with our system with TiO_2. They also reported a general Newtonian behavior of IoNanofluids based on [C_2mim][$N(CN)_2$] with 5, and 10% Al_2O_3 load for high shear rates, as reported here, namely, for lower temperatures. They quote "At sufficiently low shear rates, the IoNanofluids containing aluminum oxide displayed an unusual and irreversible increase in viscosity when heated to a sufficiently high temperature." The concentrations used are much higher than ours (0.5%), but we have noticed the same non-Newtonian behavior at high sheer stresses and high temperatures.

It is interesting to know that results by Nieto de Castro et al.[61] with the IoNanofluid [C_{12}mim][$(CF_3SO_2)_2N$] + graphene, w_{np} = 0.00053, in about the same temperature range, showed that this IoNanofluid did not produce significant enhancements for viscosity, which is a positive fact for its utilization as an alternative heat transfer fluid. However, it produced a negative or almost zero thermal conductivity enhancement, which invalidates its use for the same application. Although the mass fraction of the graphene nanoplatelets is about ten times smaller, these results demonstrate that thermal conductivity reductions in nanofluids are possible, as already found for heat capacity.[62]

Therefore, all of these results can be said to agree qualitatively, and only further studies can prove if our results are general or strictly related to the IoNanofluid currently studied.

4. CONCLUSIONS

The study of the thermal conductivity and the viscosity of the IoNanofluids of [C_2mim][$N(CN)_2$] with nano-TiO_2 illustrates reasonable enhancements of the thermal conductivity, especially when temperature rises, and controllable increases in the viscosity. Values obtained with TiO_2 are not substantially different than those obtained previously with MWCNTs, a very interesting fact due to the morphological difference between the nanoparticles, which should, in principle, originate different structured heat transfer paths and, consequently, different thermal conductivity enhancements, which is not the case.

It was also shown that the higher the temperature, the smaller the temperature range, where the IoNanofluid can be considered Newtonian, although the ionic liquid is Newtonian.

These findings align qualitatively with the observations of previous researchers working with other base fluids, substantiating the potential for diverse applications of these systems in heat transfer applications, namely, as replacement heat transfer fluids.

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Author Contributions

The experimental measurements were performed by A.L. and X.P. characterization of the Titania nanoparticles was performed by M.J.V.L. M.J.V.L. and F.S. supervised the experimental work and, with X.P., participated with TAG in the final interpretations and data discussion under the supervision of C.A.N.C. The final manuscript was written by C.A.N.C. and edited by the remaining co-workers.

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Notes

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■ REFERENCES

(1) França, J. M. P.; de Castro, C. A. N.; Lopes, M.L.; Nunes, V. M. Influence of Thermophysical Properties of Ionic Liquids in Chemical Process Design. *J. Chem. Eng. Data* **2009**, *54*, 2569−2575, DOI: 10.1021/je900107t.

(2) de Castro, C. A. N.; Lourenço, M. J. V.; Ribeiro, A. P. C.; Langa, E.; Vieira, S. I. C.; et al. Thermal Properties of Ionic Liquids and IoNanofluids of Imidazolium and Pyrrolidinium Liquids. *J. Chem. Eng. Data* **2010**, *55* (2), 653−661, DOI: 10.1021/je900648p.

(3) Nieto de Castro, C. N.; Paredes, X.; Vieira, S.; Murshed, S.; Lourenço, M. J.; Santos, F. IoNanofluids: Innovative Agents for Sustainable Development. In *Nanotechnology for Energy Sustainability*; Raj, B.; Van de Voorde, M.; Mahajan, Y., Eds.; WILEY-VCH Verlag GmbH & Co. KGaA: Weinheim, 2017.

(4) França, J. M. P.; Vieira, S. I. C.; Lourenço, M. J. V.; Murshed, S. M. S.; de Castro, C. A. N. Thermal Conductivity of [C4mim]-[(CF3SO2)2N] and [C2mim][EtSO4] and their IoNanofluids with Carbon Nanotubes: Experiment and Theory. *J. Chem. Eng. Data* **2013**, *58* (2), 467−476, DOI: 10.1021/je301183r.

(5) França, J. M. P.; Lourenço, M. J. V.; Murshed, S. M. S.; Pádua, A. A. H.; de Castro, C. N. A. Thermal Conductivity of Ionic Liquids and IoNanofluids and their Feasibility as Heat Transfer Fluids. *Ind. Eng. Chem. Res.* **2018**, *57* (18), 6516−6529.

(6) de Castro, C. N. A.; da Costa Ribeiro, A. P.; Figueiras, A. O.; Langa, E.; Vieira, S. I. C.; Lourenço, M. J. V.; dos Santos, A. F. S.; dos Santos, F. J. V.; Lampreia, I. M. S.; Goodrich, P.; Hardacre, C. Thermophysical Properties of 1-butyl-3-methylimidazolium tris-(pentafluoroethyl) trifluorophosphate, [C4mim][(C2F5)3PF3] and of its IoNanofluid with multi-walled carbon nanotubes. *J. Chem. Eng. Data* **2021**, *66*, 1717−1729.

(7) Lourenço, M. J.; Alexandre, J.; Huisman, C.; Paredes, X.; de Castro, C. N. The balance between energy, environmental security, and technical performance: the regulatory challenge of nanofluids. Special Issue on Colloids and Nanofluids for Energy Management. *Nanomaterials* **2021**, *11* (8), No. 1871, DOI: 10.3390/nano11081871.

(8) França, J.; Reis, F.; Vieira, S. I. C.; Lourenço, M. J. V.; Santos, F. J. V.; de Castro, C. A. N.; Pádua, A. H. Thermophysical Properties of Ionic Liquid Dicyanamide (DCA) Nanosystems. *J. Chem. Thermodyn.* **2014**, *79*, 248−257, DOI: 10.1016/j.jct.2014.05.008.

(9) Lourenço, M. J.; Nanofluids, S. I. V. Preparation Methodology. In *Nanofluids: Synthesis, Properties and Applications*; Murshed, S. M. S.; de Castro, C. A. N., Eds.; NOVA Science Publishers, Inc.: New York, NY, USA, 2014; Chapter 1.

(10) Bioucas, F. E. B.; Queirós, C. S. G. P.; Lozano-Martín, D.; Ferreira, M. S.; Paredes, X.; Santos, A. F.; Santos, F. J. V.; Lopes, M. L. M.; Lampreia, I. M. S.; Lourenço, M. J. V.; de Castro, C. A. N.; Massonne, K. [C2mim][CH3SO3] − A Suitable New Heat Transfer Fluid? Part 2. Thermophysical Properties of Its Mixtures with Water. *Ind. Eng. Chem. Res.* **2022**, *61* (5), 2280−2305.

(11) Lozano-Martín, D.; Vieira, S. I. C.; Paredes, X.; Lourenço, M. J. V.; de Castro, C. A. N.; Sengers, J. V.; Massone, K. Thermal Conductivity of a Metastable Ionic Liquid. *Molecules* **2020**, *25*, No. 4290, DOI: 10.3390/molecules25184290.

(12) de Castro, C. A. N.; Lourenço, M. J. V. Towards the Correct Measurement of Thermal Conductivity of Ionic Melts and Nanofluids. *Energies* **2020**, *13* (1), No. 99, DOI: 10.3390/en13010099.

(13) Ribeiro, A. P. C.; Vieira, S. I. C.; Goodrich, P.; Hardacre, C.; Lourenço, M. J. V.; de Castro, C. A. N. Thermal conductivity of [Cnmim][(CF3SO2)2N] and [C4mim][BF4] IoNanofluids with carbon nanotubes − Measurement, theory and structural characterization. *J. Nanofluids* **2013**, *2*, 55−62.

(14) Jóźwiak, B.; Greer, H. F.; Dzido, G.; Kolanowska, A.; Jędrysiak, R.; Dziadosz, J.; Dzida, M.; Boncel, S. Effect of ultrasonication time on microstructure, thermal conductivity, and viscosity of ionanofluids with originally ultra-long multi-walled carbon nanotubes. *Ultrason. Sonochem.* **2021**, *77*, No. 105681.

(15) Dzida, M.; Boncel, S.; Jóźwiak, B.; Greer, H. F.; Dulski, M.; Dulski, M.; Scheller, Ł.; Golba, A.; Flamholc, R.; Dzido, G.; Dziadosz, J.; Kolanowska, A.; Jędrysiak, R.; Blacha, A.; Cwynar, K.; Zorębski, E.; Bernardes, C. E. S.; Lourenço, M. J. V.; de Castro, C. A. N. High-performance ionanofluids from subzipped carbon nanotube networks. *ACS Appl. Mater. Interfaces* **2022**, *14* (45), 50836−50848, DOI: 10.1021/acsami.2c14057.

(16) Paredes, X.; Lourenço, M. J. V.; de Castro, C. N.; Wakeham, W. A. Thermal Conductivity of Ionic Liquids and IoNanofluids. Can Molecular Theory Help? *Fluids* **2021**, *6*, No. 116, DOI: 10.3390/fluids6030116.

(17) Almeida, R. M.; Lourenço, M. J. V.; de Castro, C. N. The Thermal Conductivity of Ionic Liquids. Experiment and Molecular Interpretation. *J. Mol. Liq.* **2024**, *397*, No. 124123.

(18) Jiang, Q.; Wang, F.; Li, R.; Li, B.; Wei, N.; Gao, N.; Xu, H.; Zhao, S.; Huang, Y.; Wang, B.; Zhang, W.; Wu, X.; Zhang, S.; Zhao, Y.; Shi, E.; Zhang, R. Synthesis of Ultralong Carbon Nanotubes with Ultrahigh Yields. *Nano Lett.* **2023**, *23* (2), 523−532, DOI: 10.1021/acs.nanolett.2c03858.

(19) Bioucas, F. E. B.; Vieira, S. I. C.; Lourenço, M. J. V.; Santos, F. J. V.; Lopes, M. L. M.; de Castro, C. A. N.; Massonne, K. [C2mim][CH3SO3] − A Suitable New Heat Transfer Fluid? Part 1. Thermophysical and Toxicological Properties. *Ind. Eng. Chem. Res.* **2018**, *57*, 8541−8551, DOI: 10.1021/acs.iecr.8b00723.

(20) Paredes, X.; Queirós, C. S. G. P.; Santos, F. J. V.; Santos, A. F.; Santos, M. S. C. S.; Lourenço, M. J. V.; de Castro, C. A. N. Thermophysical Properties of 1-Hexyl-3-Methylimidazolium Bis-(Trifluoromethylsulfonyl)Imide, [C6mim][(CF3SO2)2N] − New Data, Reference Data and Reference Correlations. *J. Phys. Chem. Ref. Data* **2020**, *49*, No. 043101.

(21) Murshed, S. M. S.; Estellé, P. A state of the art review on viscosity of nanofluids. *Renewable Sustainable Energy Rev.* **2017**, *76*, 1134−1152.

(22) Shakeel, A.; Mahmood, H.; Farooq, U.; Ullah, Z.; Yasin, S.; Iqbal, T.; Chassagne, C.; Moniruzzaman, M. Rheology of Pure Ionic Liquids and Their Complex Fluids: A Review. *ACS Sustainable Chem. Eng.* **2019**, *7* (16), 13586−13626.

(23) Jóźwiak, B.; Boncel, S. Rheology of ionanofluids − A review. *J. Mol. Liq.* **2020**, *302*, No. 112568.

(24) Liu, J.; Wang, F.; Zhang, L.; Fang, X.; Zhang, Z. Thermodynamic properties and thermal stability of ionic liquid-based nanofluids containing graphene as advanced heat transfer fluids for medium-to-high-temperature applications. *Renewable Energy* **2014**, *63*, 519−523, DOI: 10.1016/j.renene.2013.10.002.

(25) Wang, F.; Han, L.; Zhang, Z.; Fang, X.; Shi, J.; Ma, W. Surfactant-free ionic liquid-based nanofluids with remarkable thermal conductivity enhancement at very low loading of graphene. *Nanoscale Res. Lett.* **2012**, *7*, No. 314, DOI: 10.1186/1556-276X-7-314.

(26) Wittmar, A.; Ulbricht, M. Dispersions of Various Titania Nanoparticles in Two Different Ionic Liquids. *Ind. Eng. Chem. Res.* **2012**, *51* (25), 8425−8433.

(27) Bender, C. R.; Salbego, P. R. S.; Wust, K.; Farias, C. A. A.; Beck, T. S.; Machado, G.; Vaucher, R. A.; Martins, M. A. P.; Frizzo, C. P. Interaction of pharmaceutical ionic liquids with TiO2 in anatase and rutile phase. *J. Mol. Liq.* **2018**, *269*, 912−919.

(28) Wittmar, A.; Gajda, M.; Gautam, D.; Dörfler, U.; Winterer, M.; Ulbricht, M. Influence of the cation alkyl chain length of imidazolium-based room temperature ionic liquids on the dispersibility of TiO2 nanopowders. *J. Nanopart. Res.* **2013**, *15*, No. 1463, DOI: 10.1007/s11051-013-1463-2.

(29) Wittmar, A.; Gautam, D.; Schilling, C.; Dörfler, U.; Mayer-Zaika, W.; Winterer, M.; Ulbricht, M. Stable zinc oxide nanoparticle dispersions in ionic liquids. *J. Nanopart. Res.* **2014**, *16*, No. 2341, DOI: 10.1007/s11051-014-2341-2.

(30) Chereches̨, E. I.; Prado, J. I.; Chereches̨, M.; Minea, A. A.; Lugo, L. Experimental study on thermophysical properties of alumina nanoparticle enhanced ionic liquids. *J. Mol. Liq.* **2019**, *291*, No. 111332.

(31) Chereches̨, E. I.; Prado, J. I.; Ibanescu, C.; Danu, M.; Minea, A. A.; Lugo, L. Viscosity and isobaric specific heat capacity of alumina nanoparticle enhanced ionic liquids: An experimental approach. *J. Mol. Liq.* **2020**, *317*, No. 114020.

(32) Hothar, M.; Sundén, B.; Wu, Z. Rheological behaviour of IoNanofluids based on [emim][DCA] and [emim][TCM]. *J. Mol. Liq.* **2022**, *348*, No. 118064.

(33) Ueno, K.; Imaizumi, S.; Hata, K.; Watanabe, M. Colloidal Interaction in Ionic Liquids: Effects of Ionic Structures and Surface Chemistry on Rheology of Silica Colloidal Dispersions. *Langmuir* **2009**, *25* (2), 825−831.

(34) Gao, J.; Mwasame, P. M.; Wagner, N. J. Thermal rheology and microstructure of shear thickening suspensions of silica nanoparticles dispersed in the ionic liquid [C4mim][BF4]. *J. Rheol.* **2017**, *61* (3), 525−535.

(35) Novak, J.; Britton, M. M. Magnetic resonance imaging of the rheology of ionic liquid colloidal suspensions. *Soft Matter* **2013**, *9* (9), 2730−2737, DOI: 10.1039/c3sm27409h.

(36) Paul, T. C.; Morshed, A.; Fox, E. B.; Khan, J. A. Thermal performance of Al2O3 nanoparticle enhanced ionic liquids (NEILs) for concentrated solar power (CSP) applications. *Int. J. Heat Mass Transfer* **2015**, *85*, 585−594.

(37) Kiani, S.; Taherkhani, F. Size and temperature dependency on structure, heat capacity and phonon density of state for colloidal silver nanoparticle in 1-Ethyl-3-methylimidazolium Hexafluorophosphate ionic liquid. *J. Mol. Liq.* **2017**, *230*, 374−383.

(38) Patil, V. S.; Cera-Manjarre, A.; Salavera, D.; Rode, C.; Patil, K.; de Castro, C. A. N.; Coronas, A. Ru-Imidazolium Halide IoNanofluids. Synthesis, structural, morphological and thermophysical properties. *J. Nanofluids* **2016**, *5* (2), 191−208.

(39) Wegner, S.; Janiak, C. Metal Nanoparticles in Ionic Liquids. *Top. Curr. Chem.* **2017**, *375* (4), No. 65, DOI: 10.1007/s41061-017-0148-1.

(40) Hermida-Merino, C.; Pereiro, A. B.; Araújo, J. M. M.; Gracia-Fernández, C.; Vallejo, J. P.; Lugo, L.; Piñeiro, M. M. Graphene IoNanofluids, Thermal and Structural Characterization. *Nanomaterials* **2019**, *9*, No. 1549, DOI: 10.3390/nano9111549.

(41) Shakeel, A.; Mahmood, H.; Farooq, U.; Ullah, Z.; Yasin, S.; Iqbal, T.; Chassagne, C.; Moniruzzaman, M. Rheology of Pure Ionic Liquids and Their Complex Fluids: A Review. *ACS Sustainable Chem. Eng.* **2019**, *7* (16), 13586−13626.

(42) He, Z.; Alexandridis, P. Nanoparticles in ionic liquids: interactions and organization. *Phys. Chem. Chem. Phys.* **2015**, *17* (28), 18238−18261.

(43) de Castro, C. A. N.; Lamas, A.; Magendran, C.; Almeida, R. M.; Paredes, X.; Santos, Â. F.; Lampreia, I. M. S.; Santos, F. J. V.; Lourenço, M. J. V. Thermophysical Properties of 1-Ethyl-3-methylimidazolium Dicyanamide, its mixture with water. Understanding Interactions and Structure of Ionic Liquid + Water Systems. *J. Chem. Eng. Data*, April 2024.

(44) Goodwin, J. W.; Hughes, R. W. *Rheology for Chemists − An Introduction*; The Royal Society of Chemistry: Cambridge, UK, 2000.

(45) Kazakov, A.; Magee, J. W.; Chirico, R. D.; Paulechka, E.; Diky, V.; Muzny, C. D.; Kroenlein, K.; Frenkel, M. *ILThermo - NIST Standard Reference Database #147 (V2.0)*; National Institute of Standards and Technology: Gaithersburg MD, 2022 http://ilthermo.boulder.nist.gov.

(46) Zheng, Y.; Zheng, Y.; Wang, Q.; Wang, Z. Density, Viscosity, and Electrical Conductivity of 1-Alkyl-3-methylimidazolium Dicyanamide Ionic Liquids. *J. Chem. Eng. Data* **2021**, *66* (1), 480−493.

(47) Fatima, U.; Riyazuddeen; Dhakal, P.; Shah, J. K. Comparative Study of Influence of Ethanol and 2,2,2-Trifluoroethanol on Thermophysical Properties of 1-Ethyl-3-methylimidazolium Dicyanamide in Binary Mixtures: Experimental and MD Simulations. *J. Chem. Eng. Data* **2021**, *66*, 101−115.

(48) Lepre, L. F.; Gomes, M. C.; Padua, A. A. H.; Ando, R. A.; Ribeiro, M. C. C. On the Regular Behaviour of a Binary Mixture of Ionic Liquids. *J. Phys. Chem. B* **2019**, *123*, 6579−6587.

(49) Vataščin, E.; Dohnal, V. Thermodynamic properties of aqueous solutions of [EMIM] thiocyanate and [EMIM] dicyanamide. *J. Chem. Thermodyn.* **2017**, *106*, 262−275.

(50) Larriba, M.; Navarro, P.; Garcia, J.; Rodriguez, F. Liquid−liquid extraction of toluene from n-heptane by {[emim][TCM] + [emim][DCA]} binary ionic liquid mixtures. *Fluid Phase Equilib.* **2014**, *364*, 48−54.

(51) Pamies, R.; Aviles, M. D.; Arias-Pardilla, J.; Carrion, F. J.; Sanes, J.; Bermudez, M. D. Rheological study of new dispersions of carbon nanotubes in the ionic liquid 1-ethyl-3-methylimidazolium dicyanamide. *J. Mol. Liq.* **2019**, *278*, 368−375, DOI: 10.1016/j.molliq.2019.01.074.

(52) Stoppa, A.; Buchner, R.; Hefter, G. How ideal are binary mixtures of room-temperature ionic liquids? *J. Mol. Liq.* **2010**, *153*, 46−51.

(53) Neves, C. M. S. S.; Kurnia, K. A.; Coutinho, J. A. P.; Marrucho, I. M.; Lopes, J. N. C.; Freire, M. G.; Rebelo, L. P. N. Systematic Study of the Thermophysical Properties of Imidazolium-Based Ionic Liquids with Cyano-Functionalized Anions. *J. Phys. Chem. B* **2013**, *117*, 10271−10283.

(54) Freire, M. G.; Teles, A. R. R.; Rocha, M. A. A.; Schroder, B.; Neves, C. M. S. S.; Carvalho, P. J.; Evtuguin, D. V.; Santos, L. M. N. B. F.; Coutinho, J. A. P. Thermophysical Characterization of Ionic Liquids Able To Dissolve Biomass. *J. Chem. Eng. Data* **2011**, *56* (12), 4813−4822.

(55) Larriba, M.; Navarro, P.; Garcia, J.; Rodriguez, F. Liquid-Liquid Extraction of Toluene from Heptane Using [emim][DCA], [bmim]-[DCA], and [emim][TCM] Ionic Liquids. *J. Ind. Eng. Chem.* **2013**, *52*, 2714−2720.

(56) Quijada-Maldonado, E.; van der Boogaart, S.; Lijbers, J. H.; Meindersma, G. W.; de Haan, A. B. Experimental densities, dynamic

viscosities and surface tensions of the ionic liquids seres 1-ethyl-3-methylimizazolium acetate and dicyanamide and their binary and ternary mixtures with water and ethanol at T = (298.15 to 343.15 K). *J. Chem. Thermodyn.* **2012**, *51*, 51−58.

(57) Schreiner, C.; Zugmann, S.; Hartl, R.; Gores, H. J. Fractional Walden Rule for Ionic Liquids: Examples from Recent Measurements and a Critique of the So-Called Ideal KCl Line for the Walden Plot. *J. Chem. Eng. Data* **2010**, *55* (5), 1784−1788.

(58) Huang, Q.; Loureno, T. C.; Costa, L. T.; Zhang, Y.; Maginn, E. J.; Gurkan, B. Solvation Structure and Dynamics of Li+ in Ternary Ionic Liquid-Lithium Salt Electrolytes. *J. Phys. Chem. A* **2019**, *123* (2), 516−527.

(59) Mei, X.; Yue, Z.; Ma, Q.; Dunya, H.; Mandal, B. K. Synthesis and electrochemical properties of new dicationic ionic liquids. *J. Mol. Liq.* **2018**, *272*, 1001−1018.

(60) Winterer, M. *Nanocrystalline Ceramics−Synthesis and Structure*, Springer Series in Materials Science; Springer Verlag: New York, 2002.

(61) de Castro, C. A. N.; Lamas, A.; Paredes, X.; Santos, Â.F.; Lampreia, I. M. S.; Santos, F. J. V.; Lourenço, M. J. V.; Graber, T.Thermophysical Properties of 1-dodecyl-3-methyl-1H-imidazolium bis(trifluoromethylsulfonyl)amide and its IoNanofluid with Graphene. Invited Manuscript - Special Issue in Honour of Maria Eugénia Macedo *J. Chem. Eng. Data* 2023 DOI: 10.1021/acs.jced.3c00398.

(62) Parmar, N.; Bendova, M.; Wagner, Z.; Jacquemin, J. A study of changes in the heat capacity of carbon nanotube-based ionanofluids prepared from a series of imidazolium ionic liquids. *J. Phys. Chem. Chem. Phys.* **2022**, *24*, 22181−22190.

https://doi.org/10.1021/acs.jced.4c00167
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