Review

The Li_2CO_3–Na_2CO_3–K_2CO_3 Eutectic Revisited: Challenges and Gaps in Thermophysical Property Data

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Review

The Li_2CO_3–Na_2CO_3–K_2CO_3 Eutectic Revisited: Challenges and Gaps in Thermophysical Property Data

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Abstract

Molten salts are increasingly regarded as promising fluids for high-temperature heat transfer, thermal energy storage, and advanced reaction processes, including concentrated solar power (CSP), molten salt oxidation (MSO), and next-generation nuclear reactors. Among these materials, the ternary eutectic mixture Li_2CO_3–Na_2CO_3–K_2CO_3 (32.12–33.36–34.52 wt%) has emerged as a leading candidate due to its wide operating temperature range and favourable thermodynamic characteristics. Despite its relevance, substantial inconsistencies and gaps remain in the available thermophysical property data, posing challenges for reliable design, modelling, and industrial deployment. This work revisits the Li_2CO_3–Na_2CO_3–K_2CO_3 eutectic through a critical assessment of the literature from its reported melting point at 397 °C (670 K) up to approximately 1200 K. Using a methodology inspired by IUPAC-supported strategies previously applied to common liquids such as water and hydrocarbons, we examine the quantity, quality, and coherence of existing measurements. Reference correlations are proposed only where the data are sufficiently robust to justify them. The analysis highlights a pressing need for more accurate and comprehensive measurements—particularly for heat capacity, thermal conductivity, and viscosity—to enable the development of reliable standard reference correlations. Addressing these data deficiencies is essential for advancing the safe and efficient use of molten carbonates in high-temperature energy technologies.

Keywords: molten carbonates; Li_2CO_3–Na_2CO_3–K_2CO_3 eutectic; thermophysical properties; concentrated solar power; high-temperature heat transfer; viscosity; thermal conductivity; heat capacity; reference correlations; critical review

1. Introduction

Molten alkali carbonates—both pure and mixed—are regarded as highly promising fluids for high-temperature applications, including concentrated solar power (CSP), molten salt oxidation (MSO), and advanced nuclear reactor technologies. Aneke and Wang (2016) [1] provided an excellent review of energy storage technologies and their real-life applications, comparing various storage approaches in terms of technical performance, deployment scale, number of operational projects, and technological maturity. Their analysis concluded that molten salts (MS) will continue to dominate the thermal energy storage sector for large-scale applications.

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In a comprehensive two-part review, Frangini and Masi (2016) [2,3], the properties and applications of molten carbonates were examined in detail. Owing to their unique physical and physicochemical characteristics, molten carbonates were shown to be highly attractive for a wide range of energy technologies operating under diverse conditions. Key advantages highlighted in that analysis include high thermal and moisture stability, reusability, elevated electrical conductivity, and relatively low corrosiveness toward metals under controlled atmospheres. Their eutectic mixtures also exhibit reduced melting points, lowering the risk of freezing in pipelines and equipment.

Among these materials, the ternary eutectic mixture Li_2CO_3–Na_2CO_3–K_2CO_3 has been selected as an alternative high-temperature heat transfer fluid (HTF) and energy storage medium for a molten salt solar-thermal pilot plant, Prieto et al. (2020) [4]. Its adoption enabled an increase in operational temperatures from the conventional limit of 565 °C (typical of alkali nitrates) up to 700 °C. For proper equipment design and reliable process modelling, accurate thermophysical property data are essential. However, a survey of the literature reveals significant scatter among published measurements, as shown in a previous review by our group, Nunes et al. (2019) [5]. This situation has not improved substantially since then, despite the publication of some new datasets. Consequently, there remains a strong need for high-quality experimental measurements of the most relevant thermophysical properties for heat transfer and thermal storage—namely density, heat capacity, thermal conductivity, and viscosity—and for the establishment of recommended reference values for these properties.

Measuring thermophysical properties of molten carbonates presents several challenges. Some arise from their intrinsic behavior at high temperatures, while others stem from limitations of the experimental techniques themselves, including high measurement uncertainties and material incompatibilities between molten samples and instrument components, often metallic. These factors contribute to inconsistencies and reduce confidence in the available data.

In a recent study, Tasidou et al. (2019) [6] proposed reference correlations for the viscosity of 13 inorganic molten salts, reporting expected uncertainties between 2% and 7%. However, none of the carbonate salts or their eutectic mixtures were included due to the poor quality of the available viscosity measurements. More recently, the same group [7] proposed a viscosity correlation for the ternary carbonate eutectic, although it relied on Arrhenius modelling of experimental data that had not been reported by the original authors.

The aim of this paper is to provide a critical assessment of the existing thermophysical property data for the ternary eutectic molten carbonate system. Our approach follows a strategy inspired by methodologies pioneered under IUPAC for common liquids, such as hydrocarbons and water [8–10]. Historical recommendations for the thermophysical properties of molten salts date back to the work of George Janz and co-workers in the 1960s and 1970s [11]. The present work does not replace the need for new, accurate measurements of the heat capacity, viscosity, and thermal conductivity of the Li_2CO_3–Na_2CO_3–K_2CO_3 eutectic—a task currently underway in our laboratory—but aims to clarify the state of the literature and identify critical gaps.

2. Ternary Eutectic of Molten Carbonates

The ternary lithium carbonate, sodium carbonate, and potassium carbonate (Li_2CO_3-Na_2CO_3-K_2CO_3) is a mixture with the composition 43.5-31.5-25.0 mol% respectively (or 32.12-33.36-34.52 wt%) and with a melting point of T_m = 397 °C (670 K), determined by DSC [15]. Along the text, it will be denoted as $(LiNaK)_2CO_3$, or LiNaK (in equations).

3. Data Analysis

3.1. Density

Table 1 summarizes the available works for the molten ternary eutectic $LiNaK)_2CO_3$ density. Included are the reference data of Janz et al (1979) [11].

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Table 1. Existing Literature Data for the Density of Molten Ternary Eutectic $(LiNaK)_2CO_3$, at P = 0.1 MPa.

Measuring Method[a]	Declared Relative Uncertainty[b]	Temperature Range /K	Purity of Sample[c], /%	Number of Data Points/ Equation	Reference
A	0.01	680 - 860	NA	Equation	Janz et al. (1979) [11]
A	---	677 - 1081	NA	Equation	Araki et al. (1987) [12]
DBA	0.005	882 - 1158	NA	#4	Liu et al. (2003) [13]
MBP	0.003	770 - 1163	99.5	Equation	Kojima et al. (2008) [14]
A	---	723 - 893	99.9	#4	An et al. (2016) [15]

[a] A – Archimedean; DBA – Double-bob Archimedean; MBP – Maximum bubble pressure; [b] Standard uncertainty; [c] NA – Not available.

Figure 1 shows the plotted data. For comparison purposes, to show the effect of composition on the density of the melt, Sang et al. (2019) [16] data for a ternary carbonate mixture with a different composition (4-4-2 mass ratio) have been included.

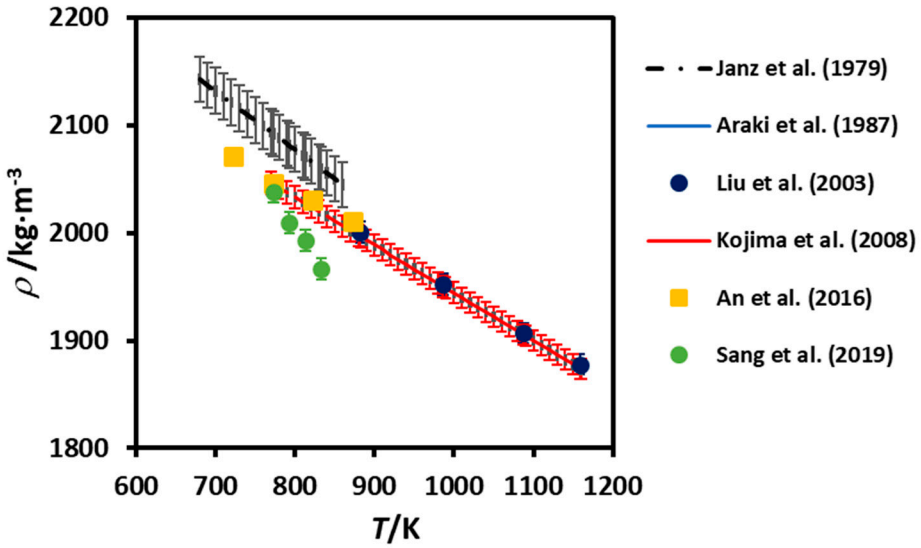

Figure 1. Density of the ternary eutectic $(LiNaK)_2CO_3$ as a function of temperature. Lines are equations given by the authors. Error bars display the declared uncertainty whenever it is known.

Based on the plotted data, we have chosen the data of Araki et al (1987) [12], Liu et al. (2003) [13], Kojima et al. (2008) [14], and An et al. (2016) [15], to establish a correlation for the density of the $(LiNaK)_2CO_3$ system. For data sets where only regression lines were available, a total of eight points were selected to cover the temperature range. The obtained equation is (1):

$$\rho(\mathrm{kg \cdot m^{-3}}) = (2387.52 \pm 7.39) - (0.44466 \pm 0.00802)(T/\mathrm{K}), \quad (1)$$

The correlation has a root mean square deviation of 5.29 kg·m^{-3}, can accommodate the data used within ± 0.3 %, and can be applied in the temperature interval 650 < T/K < 1150. Figure 2 shows the deviation of the different data sets relative to this correlation. It can be seen that the older recommendations of Janz et al. (1979) [11] are significantly higher than this correlation.

In Table 2, we propose recommended values for the density of molten ternary eutectic $(LiNaK)_2CO_3$, obtained from Eq. (1), with an uncertainty of 0.5%.

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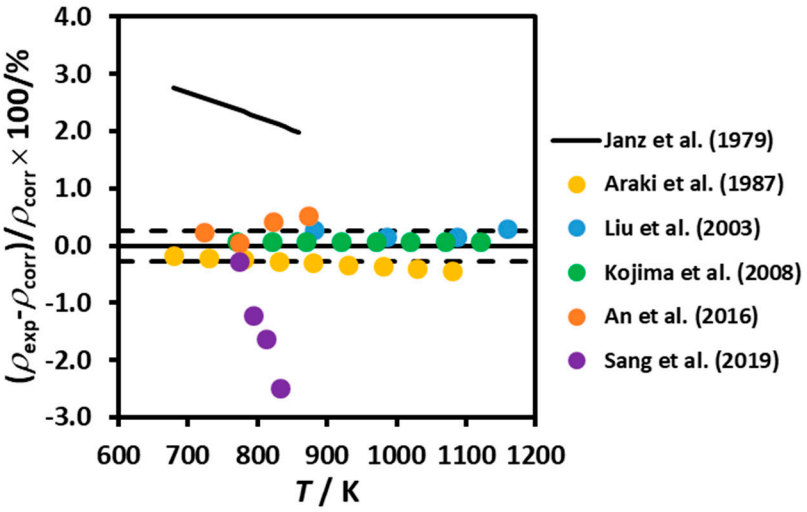

Figure 2. Deviation of different sets of data relative to the proposed correlation, Eq. (1).

Table 2. Recommended Values for the Density of Molten Ternary Eutectic $(LiNaK)_2CO_3$, at P = 0.1 MPa.

T/K	ρ/kg·m^{-3}	*T*/K	ρ/kg·m^{-3}
680	2085.1	950	1965.1
710	2071.8	980	1951.8
740	2058.5	1010	1938.4
770	2045.1	1040	1925.1
800	2031.8	1070	1911.7
830	2018.5	1100	1898.4
860	2005.1	1130	1885.1
890	1991.8	1160	1871.7
920	1978.4		

It is interesting to quote a result obtained by Kojima et al. (2008) [14] that the molar volume of the eutectic can be calculated to within 0.1 cm^3·mol^{-1} from the pure component molar volumes at the same temperature, using the molar additivity rule, at 1073 K. This means that the excess volume on the formation of the eutectic is almost negligible. However, this is not a real physical situation, as pure salts are solid at the temperatures of the eutectic, melting at temperatures above the eutectic (996 K for Li_2CO_3; 1124 K for Na_2CO_3 and 1164 K for K_2CO_3), and liquid metastable states of molten salts at temperatures 500 K below the melting point are not known to exist. To try to understand if this "pseudo" ideal solution was valid at the full liquid temperature range of the eutectic, for which there are available density data of the pure components, we have calculated the "pseudo" molar volume of the pure components, by extrapolating linearly the pure carbonates density data given by Kojima et al. (1999) [17] to the molten eutectic temperature, and calculating the molar volume of the eutectic mixture using the additivity rule for the ideal solution (no differences in interionic interactions between the three cations and between these and the carbonate ions), as explained in Eq. (2) to (4).

$$V_{m,LiNaK} = \frac{\bar{M}_{LiNaK}}{\rho_{LiNaK}}, \tag{2}$$

$$V_{m,Li2CO3} = \frac{M_{Li2CO3}}{\rho_{Li2CO3}}, V_{m,Na2CO3} = \frac{M_{Li2CO3}}{\rho_{Li2CO3}}, V_{m,K2CO3} = \frac{M_{Li2CO3}}{\rho_{Li2CO3}}, \tag{3}$$

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$$V_m^{id} = x_{Li2CO3} \times V_{m,Li2CO3} + x_{Na2CO3} \times V_{m,Na2CO3} + x_{K2CO3} \times V_{m,K2CO3}, \quad (4)$$

where x_i (i = Li_2CO_3, Na_2CO_3, and K_2CO_3) is the mole fraction of each salt in the eutectic mixture and $\bar{M}_{LiNaK}$ is the average molar molecular weight of the eutectic salt. The difference between the molar volume of the molten eutectic and that of an "pseudo" ideal solution of the individual molten salts at the same temperature is the excess volume of the mixture, defined in Eq. (5):

$$V_m^{\mathrm{E}} = V_{m,LiNaK} - V_m^{id}, \quad (5)$$

Using the recommended values from Table 1 for the density of $(LiNaK)_2CO_3$ and the densities for the pure salts obtained from [17], it can be shown that the excess volume is negligible at all temperatures, smaller than 0.001 $cm^3 \cdot mol^{-1}$. Therefore, Eqs. (4) and (5) can also be used to calculate the density of LiNaK, with $V_m^E = 0$. This result also confirms the similarity of the interactions of alkali metal ions between them and with carbonate ions. However, care must be taken not to make any conclusions from this result, as, as said above, we are not dealing with an ideal solution in thermodynamic terms, where the components must be liquid at the same temperature than the mixture.

3.2. Heat Capacity

Like for density, the available data for the heat capacity of the ternary eutectic is scarce. Table 3 resumes the available data. Figure 3 shows the plotted data. Included are data of Sang et al. (2017, 2019) [16,20] for a different composition (as stated before). The scarcity of data, especially in extended temperature intervals, does not allow the establishment of a correlation for the heat capacity, and further measurements are in progress in our laboratory. There are three sets of data in the temperature interval from 723 to 873 K, but unfortunately, with different temperature slopes. An et al. (2016) [15] suggested a constant value of Cp = 1.61 $J \cdot g^{-1} \cdot K^{-1}$. Figure 4 shows the deviations of the other data from this reference value.

Table 3. Existing Literature Data for the Heat Capacity of Molten Ternary Eutectic $(LiNaK)_2CO_3$, at P = 0.1 MPa.

Measuring Method[a]	Declared Relative Uncertainty[b]	Temperature Range /K	Purity of Sample[c] /%	Number of Data Points/ Equation	Reference
DC	0.01	680 - 1100	----	Equation	Janz et al. (1979) [11]
DSC	---	723 - 873	99.9	Equation	An et al. (2016) [15]
STA	--	703 - 1023	----	Equation	Liao et al. (2010) [18]
DSC	0.01	723 - 873	> 99	Equation	Zhang et al. (2017) [19]
DSC	0.03	773 - 833	99.7	#4; #3	Sang et al. (2019a, 2019b) [16,20]

[a] DC – Drop calorimetry; DSC – Differential scanning calorimetry; STA – Simultaneous thermal analysis; [b] Standard uncertainty.

The data of Liao et al. (2010) [18] is far higher than all the other. Even for the other sets, the percent deviation can only be accommodated within ± 20% of the chosen reference point. It means that improved measurements of this property are a task that must be pursued.

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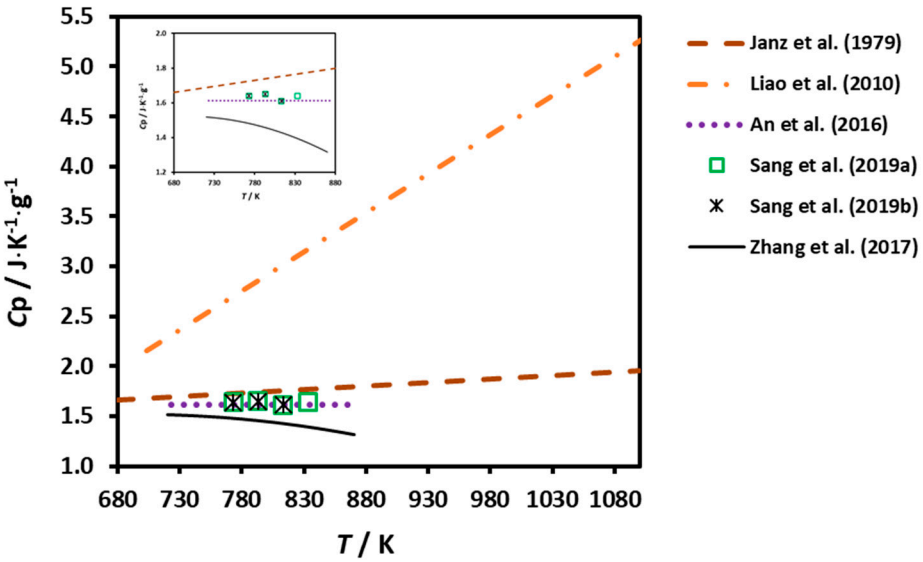

Figure 3. Heat capacity of the ternary eutectic $(LiNaK)_2CO_3$. Lines are equations given by the authors. The inset represents the data in a limited temperature range to enhance visibility.

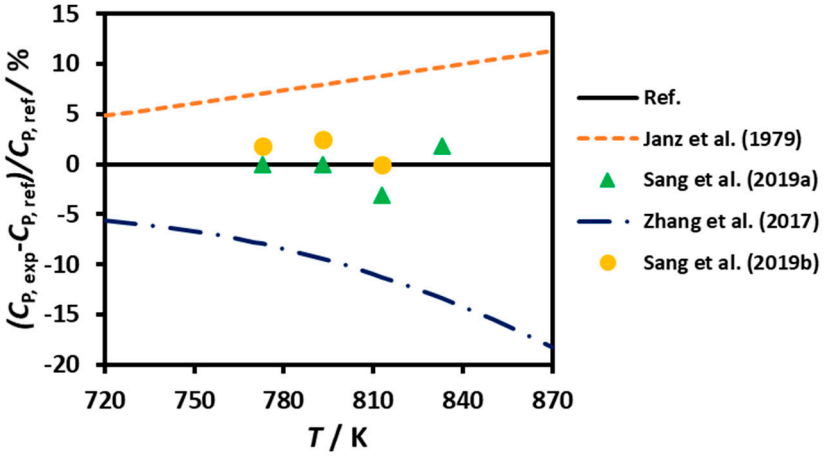

Figure 4. Deviation of different sets of data relative to the proposed reference value [15].

3.3. Thermal Conductivity

Contrary to other properties, Janz et al. (1979) do not provide a reference correlation for the thermal conductivity of the $(LiNaK)_2CO_3$ system. Table 4 lists the available data.

Figure 5 shows the thermal conductivity as a function of temperature for the different sets of data. As can be seen, the differences between different sets of data are too large, an obvious consequence of not so well-designed experimental apparatus or not accounting for additional methods of heat transfer present in the measurements, namely, radiation. There are 3 sets of data with similar trends, those of Araki et al. (1987) [12], Sang et al. (2019) [16], and Grosu et al. (2021) [24], all obtained with the Laser Flash technique, but new data, preferably obtained with a direct method to measure the thermal conductivity, is necessary. This technique measures directly the thermal diffusivity, and if density, ρ and specific heat capacity, C_p are available, the thermal conductivity, λ, can be obtained from the definition through Eq. (6):

$$\lambda = \alpha \rho Cp, \tag{6}$$

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Table 4. Existing Literature Data for the Thermal Conductivity of Molten Ternary Eutectic $(LiNaK)_2CO_3$, at P = 0.1 MPa.

Measuring Method[a]	Declared Relative Uncertainty[b]	Temperature Range /K	Purity of Sample[c] /%	Number of Data Points/ Equation	Reference
LF (Calc)	--	673 - 764	NA	Equation	Araki et al. (1987) [12]
FRS (Calc)	--	680 - 1030	NA	Equation	Otsubo et al. (1998) [21]
THW	0.03	803 - 943	NA	Plot	Zhang et al. (2002) [22]
LF (Calc)	--	723 - 873	NA	#4	An et al. (2016) [15]
SSCC	0.03	670 - 930	pure	Equation	Dokutovich et al. (2018) [23]
LF (Calc)	0.03	773 - 833	> 99	#4	Sang et al. (2019) [16]
LF (Calc)	0.05	693 - 823	NA	#4	Grosu et al. (2021) [24]

[a] LF(Calc) – Laser flash, calculated from thermal diffusivity, heat capacity, and density; FRS (Calc) – Forced Rayleigh scattering, calculated from thermal diffusivity, heat capacity, and density; THW – Transient hot wire; SSCC – Steady state coaxial cylinders; [b] Standard uncertainty; [c] NA – not available.

In our opinion, it is premature to suggest any reference correlation for this property at this time, as further measurements are necessary. Measurements based on the transient hot-strip method [25,26], using a special designed platinum thin-film sensor, are under way in our laboratory and we hope to present data soon.

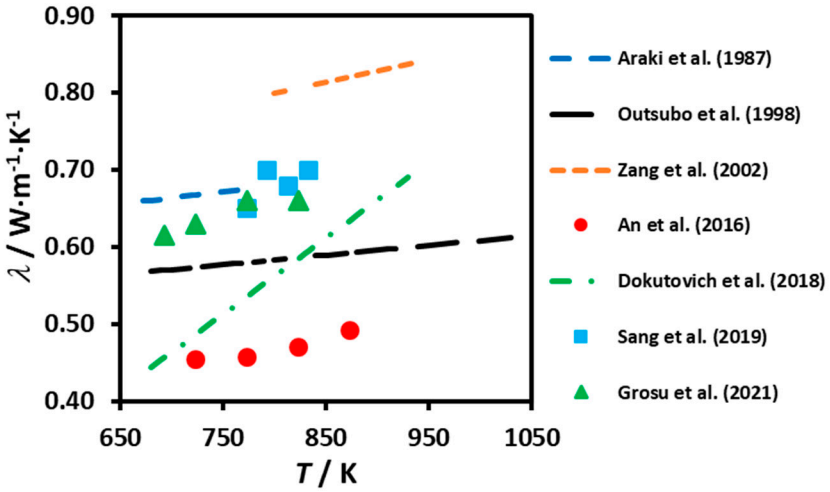

Figure 5. Thermal conductivity of the ternary eutectic $(LiNaK)_2CO_3$. Lines are equations given by the authors. The points from Grosu et al. (2021) [24] were estimated from the available plot.

3.3. Viscosity

Viscosity was first measured using an oscillating cylinder viscometer by Janz and Saegusa (1963) [27], and these measurements were the basis for the first reference correlation proposed by Janz et al. (1979) [11]. However, these measurements were later revised, using an improved instrument, and found to have serious errors. Table 5 summarizes the available data for the viscosity of the ternary melt, and Figure 6 displays the variation of viscosity with temperature for all data sets.

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Table 5. Existing Literature Data for the Viscosity of Molten Ternary Eutectic $(LiNaK)_2CO_3$, at P = 0.1 MPa.

Measuring Method[a]	Declared Relative Uncertainty[b]	Temperature Range /K	Purity of sample[c] /%	Number of Data Points/ Equation	Reference
OCY	0.25	760 - 870	NA	Equation	Janz et al. (1979) [11]
OCY	0.03	890 - 1175	98.4	#15	Ejima et al. (1987) [28,29]
RC	NA	700-970	NA	Diagram	Liao et al. (2010) [18]
RC	0.05	690 - 1120	99.8	#6	Kim et al. (2015) [30]
RC	NA	740 - 920	99.9	Equation	An et al. (2016) [15]
RC	NA	740 - 890	99.9	Equation	An et al. (2017) [31]
RPP	NA	673 - 773	99	Diagram	Grosu et al. (2021) [24]

[a] OCY – Oscillating cylinder; RC- Rotating cylinder; RPP - Rheometer, parallel plate; [b] Standard uncertainty; [c] NA - Not available.

It is clear from Figure 6 that the data sets of Janz et al. (1979) [11] and Liao et al. (2010) [18] deviate very much from the other data, namely in the lower temperatures of the melt. Recently, as mentioned in the introduction, Tasidou et al. (2019) [7] proposed a reference correlation for the viscosity of the ternary melt for 740 < T/K< 1140. They considered the data of Ejima et al. [28,29] and the data of An et al. (2017) [31] as primary data, although data points used were taken from the correlation. The proposed reference, Eq. (7), was:

$$\eta\,(\mathrm{mPa\cdot s}) = 0.1901\exp\left(\frac{2660.0}{T(\mathrm{K})} + \frac{726990}{T(\mathrm{K})^2}\right), \qquad (7)$$

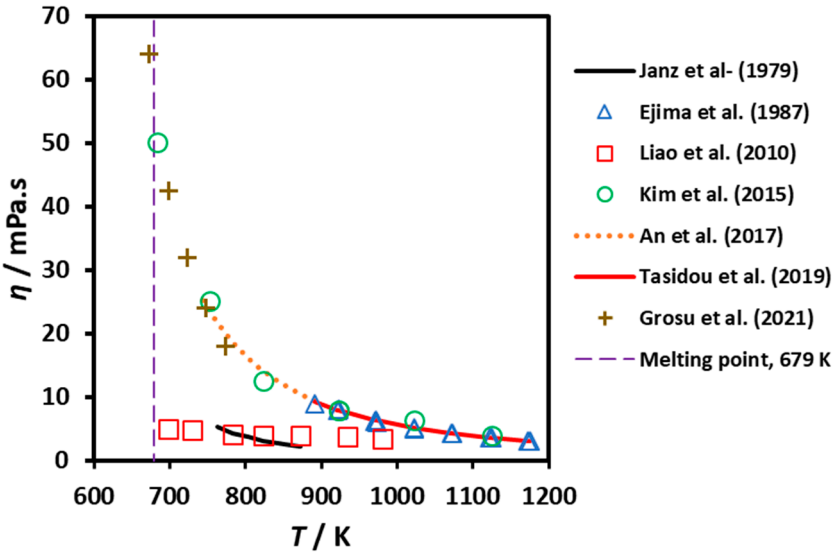

Figure 6. Viscosity of the ternary eutectic $(LiNaK)_2CO_3$. Lines are equations given by the authors. The points from Liao et al. (2010) [18] and Grosu et al. (2021) [24] were estimated from the available plot.

An uncertainty of 3% was claimed. An et al. (2016) [15] also published the viscosity of this melt early, and a slightly different equation was presented, not differing more than 0.5 % from the previous equation.

Figure 7 shows the percent deviation of the other data from the reference correlation proposed by Tasidou et al. (2019) [7], in the temperature interval considered. It includes the other existing experimental points. Deviations of the data used in the correlation never amount to more than 3 %. However, the data of Janz et al. (1979) [11] and Liao et al. (2010) [18] deviate considerably from the proposed equation
(- 80 to - 40%), probably due to deficient measurements. Data of Kim et al. (2015) [30] must have an

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uncertainty greater than the claimed 5%, but it follows the temperature variation trend. The data of Grosu et al. (2021) [24] also deviates very much, showing a completely different slope with temperature (and consequently a wrong activation energy for flow). However, it must be said that most of this data was obtained at temperatures below the correlation limit, recommending new experimental data with an absolute viscometer, currently undertaken in our laboratories, with an estimated expanded uncertainty of 1.2 %, with the oscillating cup viscometer.

The equation presented by Ejima et al. [28] is in the form of the common Arrhenius equation, normally obeyed by fluids with Newtonian behavior. The equation obtained, Eq. (8), using a reagent and commercial-grade eutectic, was:

$$\eta\ (\mathrm{mPa \cdot s}) = 0.1038 \exp \frac{(33140)}{RT(\mathrm{K})}\ , \tag{8}$$

However, the equation of Tasidou et al. (2019) [7] extends the applicability of Eq. (4) to much lower temperatures, although with the graphical data of An et al. (2017) [30]. Since the method used by Ejima et al. (1987) [28,29] is an absolute method and experimental data are available, it can be considered as the reference for the viscosity in the interval of temperatures from 890 to 1175 K. New data on the viscosity of molten ternary eutectic $(LiNaK)_2CO_3$ is necessary, namely for temperatures between the melting point and 920 K, obtained with an absolute method. These data are currently being obtained in our laboratory and will be published soon.

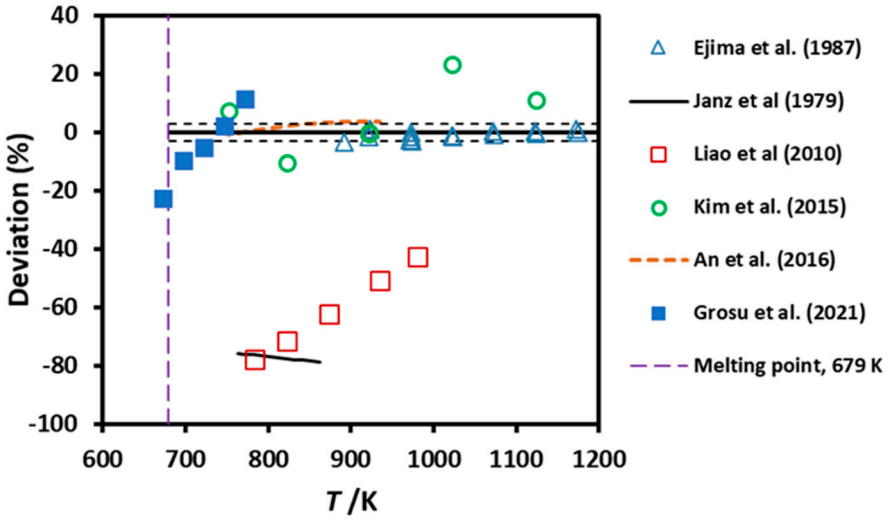

Figure 7. Deviation (in %) of different sets of data relative to the proposed reference data of Tasidou et al. (2019) [7], for the temperature range 670 < *T*/K< 1200. Dashed lines represent the claimed uncertainty of this correlation.

4. Conclusions

This report provides a critical assessment of the available thermophysical property data for the ternary molten carbonate eutectic $(LiNaK)_2CO_3$, a material of growing relevance for high-temperature heat transfer and thermal energy storage in concentrated solar power, molten salt oxidation, and advanced nuclear reactor technologies. Despite its technological importance, the quality and consistency of published data have not improved substantially since our previous review (Nunes et al., 2019) [5], although some new datasets have appeared.

Among the key properties evaluated, **density** remains the only one for which a reliable reference correlation can be proposed. Because density is an equilibrium thermodynamic property, the literature includes sufficiently consistent measurements, making Eq. (1) suitable for estimating its temperature dependence. For the **specific heat capacity at constant pressure**, a constant value of Cp

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= 1.61 $J \cdot g^{-1} \cdot K^{-1}$ can be suggested, although this does not differ significantly from the classical recommendation of Janz et al. (1979) [11]. In contrast, no meaningful reference data can be established for the **thermal conductivity**, as most measurements are derived from thermal diffusivity experiments that yield contradictory temperature trends ($d\lambda/dT$ either positive or negative), as observed previously for conductive liquids [5]. Regarding **viscosity**, the correlation recently proposed by Tasidou et al. (2019) [6] is applicable only within the range 890–1175 K, corresponding to the absolute measurements of Ejima et al. (1987) [27,28], obtained using the oscillating-cylinder method. Older recommendations by Janz et al. (1979) [11] are significantly lower and inconsistent with these more reliable data.

Overall, there is a clear need for **new high-quality measurements of heat capacity, thermal conductivity, and viscosity** for the molten ternary eutectic $(LiNaK)_2CO_3$, ideally extending up to at least 1100 K (827 °C), to enable the development of robust standard reference correlations essential for accurate process design in CSP and MSO applications.

Nevertheless, following the insights of Starke et al. (2024) [32], it appears increasingly likely that operating CSP plants above 650 °C may not yield net economic benefits. Although higher temperatures improve the thermal efficiency of Rankine cycles, this gain is offset by the substantially higher cost of the required high-temperature materials - such as nickel-based superalloys (Inconel 625 or Haynes 230) used in receivers, piping, and storage tanks. This economic constraint may ultimately limit the operating temperature range in which advanced molten carbonate systems can be deployed.

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Abbreviations

The following abbreviations are used in this manuscript:

CSP	Concentrated solar power
MSO	Molten salts oxidation
A	Archimedean
DBA	Double-bob Archimedean
MBP	Maximum bubble pressure
DC	Drop calorimetry
DSC	Differential scanning calorimetry
STA	Simultaneous thermal analysis
LF (Calc)	Laser flash, calculated from thermal diffusivity, heat capacity, and density
FRS (Calc)	Forced Rayleigh scattering, calculated from thermal diffusivity, heat capacity, and density
THW	Transient hot wire
SSCC	Steady state coaxial cylinders
OCY	Oscillating cylinder

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RC	Rotating cylinder
RPP	Rheometer, parallel plate
NA	Not available

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