

# Molten salts database for energy applications

R. Serrano-López<sup>a,\*</sup>, J. Fradera<sup>a</sup>, S. Cuesta-López<sup>a,\*\*</sup>

<sup>a</sup>Science and Technology Park. I+D+I Building. Room 63. Plaza Misael Bañuelos s/n, 09001, Burgos (Spain)

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## Abstract

The growing interest in energy applications of molten salts is justified by several of their properties. Their possibilities of usage as a coolant, heat transfer fluid or heat storage substrate, require thermo-hydrodynamic refined calculations. Many researchers are using simulation techniques, such as Computational Fluid Dynamics (CFD) for their projects or conceptual designs. The aim of this work is providing a review of basic properties (density, viscosity, thermal conductivity and heat capacity) of the most common and referred salt mixtures. After checking data, tabulated and graphical outputs are given in order to offer the most suitable available values to be used as input parameters for other calculations or simulations. The reviewed values show a general scattering in characterization, mainly in thermal properties. This disagreement suggests that, in several cases, new studies must be started (and even new measurement techniques should be developed) to obtain accurate values.

*Keywords:* properties, molten salt, CFD, CSP, coolants, energy

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## Nomenclature

### *Symbols and Units*

T	Temperature, °K
M.P.	Melting point, °K
v	Velocity, m/s
$\rho$	Density, kg/m <sup>3</sup>
$\eta$	Dynamic viscosity, Pa·s
$\nu = \frac{\eta}{\rho}$	Kinematic viscosity, m <sup>2</sup> /s
$\lambda$	Thermal conductivity, W/(m·°K)
Cp	Heat Capacity, J/(kg·°K)

### *Salt Mixtures*

#### *Fluorides*

2LiF – BeF <sub>2</sub>	FLiBe
LiF – NaF – KF	FLiNaK
LiF – NaF – BeF <sub>2</sub>	FLiNaBe
NaF – NaBF <sub>4</sub>	Denoted by us as NaFNaB
KF – ZrF <sub>4</sub>	Denoted by us as FluZirK

#### *Chlorides*

KCl – MgCl <sub>2</sub>	Denoted by us as CloKMag
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#### *Nitrates*

NaNO <sub>3</sub> – KNO <sub>3</sub>	Solar Salt
NaNO <sub>3</sub> – NaNO <sub>2</sub> – KNO <sub>3</sub>	Hitec <sup>®</sup>

### *Acronyms and Abbreviations*

ACS	American Ceramic Society
AHTR	Advanced High-Temperature Reactor
ARE	Aircraft Reactor Experiment
CFD	Computational Fluid Dynamics
CSP	Concentrated Solar Power

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\*Corresponding author. Tel: (+34) 947258907. Universidad de Burgos, Spain.

\*\*Principal corresponding author. Tel: (+34) 947259062. Universidad de Burgos, Spain.

*Email addresses:* robertosl@ubu.es  
(R. Serrano-López), jfradera@ubu.es (J. Fradera),  
scuesta@ubu.es (S. Cuesta-López)

CSPonD	Concentrated Solar Power on Demand
FHR	Fluoride Salt Cooled High-Temperature Reactor
HR	Homogeneous Reactor
HTS	Heat Transfer Fluid
HTX	Heat Exchanger
IHX	Intermediate Heat Exchanger
INL	Idaho National Laboratory
MSRE	Molten Salt Reactor Experiment
NREL	National Renewable Energy Laboratory
ORNL	Oak Ridge National Laboratory
TES	Thermal Energy Storage

## 1. Introduction

Different heat transfer systems are being studied around the use of molten salt technology as working fluid. The initial development of salts features and use was carried out at Oak Ridge National Laboratory (ORNL) for aircraft propulsion purposes [1]. The ORNL team build demonstration reactors between 1950's and 1960's (ARE and MSRE), and continued their research during several years in order to design a Molten Salt Breeder Reactor [2]. During these years, a great amount of technical reports about salt candidates and tests were written, revised and archived, including a review of the final experimental successes [3]. The experience collected in that decades has been the subject of a renewed interest, due to potential advantages of liquid salt coolants in different ways [1, 4, 5]:

- high volumetric heat capacity,
- high boiling point and low vapor pressure,
- no undesirable chemical exothermic reactions between different zones of energy plants and power cycle coolants (core, heat exchange loop),
- optical transparency during inspection operations,
- ability to dissolve actinides,
- great insensitivity to radiations.

Molten salts also offer nuclear breeding capability by optimizing mixture and giving a on-line processing to the fuel salt carrier [2], and certain compositions can be used as neutron moderators (e.g. those containing Be). The advantages of salt coolants enable them for being used in heat transport loops, and would open new nuclear and non-nuclear applications [4, 6-8] :

- fission power plants (both liquid or solid fuel, mainly in high-temperature reactors),
- fusion or hybrid reactors,
- hydrogen production,
- long distance heat transport,
- nuclear fuel reprocessing,
- chemical industry,
- oil refineries,
- shale oil processing, etc.

There are also advanced studies about the use of fluids in the power cycle, transport and heat storage in high-efficiency solar power facilities [9-12]. Even future nuclear-propelled space aircrafts are being now studied around the same concepts [13]. Developments achieved in every of these fields could be generalized for civil and industrial uses in the future with the benefits of scale economy and standardization of processes.

The present work intends to be a thorough review of the current knowledge on molten salt properties, with the aim of giving to the researcher a key to evaluate their behavior. These data may be of interest in both experimental as numerical simulation tasks, such Computational Fluid Dynamics (CFD), and can be applied for thermal exchange, storage or piping systems [14].

## 2. Background

As has already be mentioned, the initial studies about salt properties were developed by scientists involved in chain reactions. Harold Urey, a nuclear chemist, and Eugene Wigner, a theoretical physicist, both implicated in the Manhattan Project, were the early promoters of the Homogeneous Reactors (HR). First molten fluorides ideas came into the chain reaction community by 1945 [15]. Therefore, ORNL Chemical Technology

Division was searching since the late forties for a homogeneous liquid solution suitable for use as a fuel for the aircraft reactor of the Aircraft Nuclear Propulsion Project. A fluid able for being used at high temperature and low pressure was needed, with the requirement of radiation stability and actinides solubility [16]. It is assumed that Edward Bettis and Raymond Clare Briant were the persons who, years later, cut the Gordian knot by suggesting again the use of fluorides with the Molten Salt Reactor concept [17, 18].

Although initial tests with HR were developed through sulphates, the pressure of Cold War shoved US Atomic Energy Commission to start the Aircraft Nuclear Propulsion Program [18]. In this case, new requirements were solved by the use of liquid alkali fluorides instead of solid fuel rods. Several developments in hi-tech materials and salt chemistry (e.g. dry fluorination) were added to knowledge during this experiences. From the very beginning, structural materials corrosion appeared as the most important issue to resolve [16]. Lithium and beryllium salts were very early considered to allow self-moderated fuels [19], and FLiNaK appeared simultaneously in heat transfer studies [20, 21]. Using graphs and charts, Poppendingck [22] analyzed some useful properties for different coolants, including molten salts.

During the forties and the fifties some reports were also written for ceramic applications (see, e.g., [23–28]). Since 1964, there have been different compilations of phase equilibria diagrams at the National Bureau of Standards (edited by ACS) [29]. This multi-volume work is continuously improved performing the cumulative specialized ACerS-NIST Phase Equilibria database (ACerS-NIST Phase Equilibria Diagrams), which is also on-line nowadays (<http://ceramics.org/publications-and-resources/phase-equilibria-diagrams>).

Ricci [30] and Thoma [31] made the first efforts to compile specific available equilibria data for different kind of chemical systems for nuclear applications. Maybe the first review of fused salt properties was made by Grimes [32] for nuclear reactors coolants (FLiBe, FLiNaK, and FLiNaBe). Grimes included measured values and correlations for Melting Point (MP), density ( $\rho$ ), heat capacity ( $Cp$ ) and viscosity ( $\eta$ ) for these salts.

During sixties, several achievements were made around the Molten Salt Reactor concept, including the referred use of salts as coolants in heat exchange

systems. Most of the reports came from ORNL Divisions. Blanke et al. [33] studied Li and Be fluorides density and viscosity. Grimes [34] gave some values for FLiBe [66-34 %] at 854 °K ( $\rho$ ,  $Cp$ ,  $\eta$ ,  $\lambda$ ). Thereafter, Grimes [35] discussed the requirements of coolant fluids for the MSBR providing values for NaFNaB [4-96 %] and FLiNaBe [5-56-42 %, and 26-41-36 %] among other salt mixtures at 727 °K ( $\rho$ ,  $Cp$ ,  $\eta$ ,  $\lambda$ ) [35].

Cantor [36–40] studies are also a classical reference for salt properties. Two mixtures were listed in 1968 [37], FLiBe [66-34 %] and NaFNaB [92-89 %], based in a critical review of own and other measurements. Then FLiBe was again studied in 1969 [39], giving density and viscosity correlations for different melt compositions. Some fluoride mixtures (FLiBe, NaFNaB) were revised also by Cantor in 1973 [40].

McDuffie et al. [41] reviewed again coolant requirements for heat exchange for nuclear technology purposes, summarizing physical properties of different mixes of fluorides, chlorides, nitrates, and fluoroborates. Similar works were conducted by Sanders (1971) [42] (NaFNaB, FLiBe, FLiNaK, nitrates), and Kelmers et al. (1976) [43] (NaFNaB, FLiBe). The use of low-cost molten salts as heat transfer fluids and their potential for thermal energy storage were also early discussed; Silverman and Engel made a review of Solar Salt and Hitec<sup>®</sup> capabilities in 1977 [44].

Janz started in 1968 a huge effort to compile a general database for molten salts [45, 46] relevant to energy storage [47–49]. Reports were published by the Office of Standard Reference Data at the National Bureau of Standards (OSRD-NBS), including a comprehensive compilation of eutectic compositions for salts. This series made a critical review of measurement methods and correlations, and were progressively written and published between 1968 and 1981.

Janz also worked in cumulative results for the salts properties between 1972 and 1983 (the Molten Salts Standards Program) [50–56]. This general database, subsequently reprinted and summarized in 1988 [57], is widely used today for any purpose due to its updated recommendations and re-examined best values for making density and viscosity predictions [58].

Lately, an important assessment of some liquid salts was conducted by Williams et al. in 2006 at ORNL [59–61], focusing their works in the Advanced High-Temperature Reactor (AHTR) ini-

tial concept development (primary and secondary coolants). This work included classical Janz, Cantor and ORNL entrances in most usual salt mixtures for nuclear reactors. However, Be-containing salts were in these case excluded because of their higher cost and toxicity. These studies also advised about taking into account  $\text{LiCl} - \text{KCl} - \text{MgCl}_2$  due to its low-cost. Further, Williams included additional measurements for relatively unexplored Zr and Al fluoride salts [62].

During these last years, Winconsin-Madison University has joined with Shell Company to perform the on-line Molten Salts Database. This American institution is currently working to maximize molten salts potential in energy issues (in support of fusion reactor and Very-High-Temperature Reactor (VHTR) concepts), with emphasis in FLiNaK and CloKMag mixtures[63]. Additional efforts are being conducted in the Idaho National Laboratory (INL) [64, 65].

Other institutions have funded knowledge on liquid salts behavior and selection criteria, like the International Atomic Energy Agency (IAEA) [66], the International Science and Technology Center (ISTC) by the support of European Community [67], and the European Commission through ALISIA (Assessment of LIquid Salts for Innovative Applications) [8].

### 3. Selection of salts

The advantages of molten salts as Heat Transfer Fluid (HTF) and Thermal Storage System (TES) promise a great development during next decades. The cost for the required volume of heat exchangers and pumps are highly reduced by the use of liquid salts instead of other coolants due to their higher volumetric heat capacity without the need of pressurizing. It has been reported that melting points and heat capacities increase in the following order: nitrates, chlorides, carbonates, and fluorides [68]. In any case, fluid salts provide the potential for improved heat transfer and reduced pumping powers and volume of the heat exchanger compared with helium. Molten salts have a 25% higher volumetric heat capacity than pressurized water, nearly five-times that of liquid sodium [69], and more than twice than lead or lead-bismuth eutectic [6, 70].

Long term corrosion, compatibility with available structural materials, potential toxicity and final costs are key research issues. In some cases, stability requirements reduce the number of chance

possibilities; in others, avoid freezing maybe the critical issue [71]. So the salt must be properly selected in order to agree the particular conditions of use.

Be mixtures require very special and expensive handling efforts due to toxicity, and so has been discarded from some reports [61]. For this reason FLiNaK has emerged, in several cases, as main alternative because of its low toxicity, excellent heat transfer properties, and chemical properties similar to those of FLiBe [72]. In nuclear applications, purity of Li is an additional requirement with primary coolants (at least 99,995% of  $^7\text{Li}$  is needed to avoid decrease reactivity feedback, due to tritium generation by the neutron absorption of  $^6\text{Li}$  isotope), and even with secondary coolants in case of a leak at Heat Exchanger (HTX) loop. For this reason other possibilities (other than FLiBe and FLiNaK) are now being explored for the latest versions of VHTR Intermediate Heat Exchanger (IHX), e.g. FluZirK liquid salt, wich has a relatively low toxicity, and does not include Li [73]. Williams discussed the influence of the price of the components with different salt mixtures [61]. His conclusions determined that magnesium chlorides are the least expensive of all, while fluorides, fluoroborates and Li-containing mixtures increase the price of the coolant (Williams excluded Be because of the same issues explained above). Hence, with a multi-criteria analysis including technical and economic factors, an additional study of  $\text{MgCl}_2$ -containing salts should be recommended (despite of inferior heat-transfer metrics of chlorides). The ideas previously summarized have suggested both FluZirK and CloKMag as possible working fluids in heat transfer piping systems. Several studies have been focused towards the behavior of the new potential fluids [74–77].

Zirconium fluorides were used in the past given the fact of their high solubility for actinides and oxygen getter. In addition, Zr has been proposed to mitigate materials corrosion by controlling the red-ox potential of the salts, but the activation products complicate their ability to be used as primary coolants [63]. Heavy halide salts (bromides, iodides) have poor heat transfer metrics [61], but chlorides, as well as nitrites and nitrates may be useful respect to raw materials cost. Oxygen-containing salts, such as nitrite-nitrate salt mixtures are not suitable as primary coolants in nuclear plants [78]. Chlorides were avoided in the past due to corrosion, but there are renewed expectatives around them in AHTR design.

New salt compositions are probably going to be tested for large scale energy systems, and even nanofluids could emerge as future solutions increasing  $C_p$  of usual base fluids by the addition of nanoparticles in low percentages [79]. However, after a literature survey, classical mixtures appear in most of conceptual and commercial designs of both nuclear and non nuclear applications. For instance, recent reports [8, 66, 80] have reviewed most usual proposals for nuclear reactors, including GEN-IV concepts.

Synergistic efforts in heat transport liquids are now being developed for both molten and liquid salt concepts, especially taking into account new possibilities in TES and Concentrated Solar power (CSP) technologies [10, 81]. The need of decarbonizing the energy mix current century has attracted and renewed the attention to different studies started decades ago, when nuclear and aerospace research was growing up [82, 83]. In the other hand, first commercial CSP-TES systems are using common compositions as working fluids, whose behavior is well known and have more competitive cost. These salt mixtures take in advance the accrued accomplishments since middle past century in heat transfer and storage, looking for compromise between thermal efficiency and economy [71]. For CSP and TES, the raw materials cost of nitrogen-based salt-compositions makes them currently the most competitive solution (Solar salt and Hitec<sup>®</sup>). Some Li-containing mixtures and other additives are also been investigated and patented for future development [84, 85]. In this case nitrate-nitrites are not only cheaper, but also much less corrosive than fluorides or chlorides, while thermal capabilities could be improved on a compromise solution in the medium term.

As have been mentioned in section 2, several experimental works have been carried out for fluoride, chloride and nitrate salts (single, binary and ternary mixtures) since the ARE project in ORNL. The working fluid selection must take into account different circumstances, so there are different candidate salt mixtures for different applications. Firstly, in case of primary nuclear coolant the ideal salt must have: (i) a melting point well below the coldest point in the circuit, (ii) a boiling point and thermal stability well above any credible accident condition temperatures, (iii) a low vapor pressure, (iv) a low viscosity at operating temperature, (v) a large heat capacity, good thermal conductivity, low Prandtl number, and (vi) a large change in spe-

cific volume with temperature to effectively drive natural circulation cooling. Chemical and neutron behavior may be also constraints in the fluid selection, so that the neutron-absorption cross section of coolant should be lower enough to guarantee stability under radiation. Moreover, the coolant selection have to ensure compatibility with structural materials of core, loop, and piping system [86].

Fuel-salts require in addition an adequate actinide solubility at the working core temperature, and the outlined features have also to be properly identified for the fuel mixture.

Secondary coolants have no nuclear constraints, and the evaluation criteria is mainly based on heat transport performance. But the same chemical system should be employed in both sides of HTX, so that compatibility of structural alloy with two different species is not required at the working temperature. Therefore, requirements are simpler than for primary coolant: low melting and boiling point, low vapor pressure, thermal stability and conductivity, low viscosity, large heat capacity, material compability, and preferably non/low toxicity. Some of them have been discussed as “Figure of merits” (FOM) by Idaho National Laboratory (INL) [87]. Whatever function proposed (primary, fuel or secondary coolant), commercial availability and industrial processing cost must also be take in account. The cost and especial care demanded for Be handling have been also referred occasionally by experimental researchers as an additional use constraint, at least during initial knowledge development of high temperatures performance [59, 61].

As oxygen-containing salts, nitrite-nitrate salt mixtures are not suitable as primary coolants [78]. Although latest reports agree on avoid chlorides for MSR, and nitrites-nitrates are also unlisted [80], both of them are frequently referred for heat exchange coolants in energy applications. Williams [61] recommended the ternary eutectic  $\text{LiCl} - \text{KCl} - \text{MgCl}_2$  for additional study because of its potential and raw-material cost, and this salt has been recognized also by Beneš and Konings [80, 88] as FLiNaK alternative HTF in the VHTR concept. As mentioned above, current Fluoride-Salt-Cooled High-Temperature (FHR) version includes FluZirK as secondary coolant [73].

This section have checked the most important criteria used for molten salt selection, that can be briefly summarized in the following general ideas:

- high volumetric heat capacity compared with

other coolants

- mandatory  $^7\text{Li}$  purity requirements in nuclear applications
- the cost is lower for chlorides, and higher for fluorides, fluoroborates and Li or Be containing mixtures
- Zr provides actinides solubility, oxygen getter capability and red-ox control to salt mixtures, but also have undesirable activation products
- O-containing salts are not suitable as primary coolants
- Zr-fluorides are also being studied as secondary coolants, and chlorides may be a low cost alternative to take into account.

#### 4. Molten salt thermo-physical properties

In this section, a review of the most relevant thermo-physical properties for design calculations is shown for the most common and recent coolant or HTS composition choices:

1. FLUORIDES:  $2\text{LiF}-\text{BeF}_2$ , (hereafter FLiBe),  $\text{LiF}-\text{NaF}-\text{KF}$  (hereafter FLiNaK),  $\text{LiF}-\text{NaF}-\text{BeF}_2$  (hereafter FLiNaBe),  $\text{NaF}-\text{NaBF}_4$  (hereafter NaFNaB),  $\text{KF}-\text{ZrF}_4$  (hereafter FluZirK)
2. CHLORIDES:  $\text{KCl}-\text{MgCl}_2$  (hereafter CloK-Mag)
3. NITRATES:  $\text{NaNO}_3-\text{KNO}_3$  (hereafter Solar Salt), and  $\text{NaNO}_3-\text{NaNO}_2-\text{KNO}_3$  (hereafter Hitec<sup>®</sup>)

Selected properties, by order, are summarized for each salt mixture: melting point, density, viscosity, heat capacity, thermal conductivity. Most usual molar compositions are shown in brackets, in Table 1. When available, possible differences have been checked by using different compositions of the same salt by the same work or author, in order to analyze its behavior in molar terms and the differences among the temperature range of measurement.

Phase diagrams and useful properties have been also recently reviewed by Beneš and Konings [80, 88] for nuclear fission applications, including critical discussion of recent values and measurements of FLiBe, FLiNaK and NaFNaB.

The currently useful mixtures for solar energy are mainly nitrates and nitrites. Beneš et al. [89] used a

encapsulation technique with Solar Salt, measuring phase diagram. Ferri et al. proposed selected properties of Solar Salt in RELAP5-3D code for solar parabolic collectors [90], and Bauer et al. reviewed thermo-physical correlations [91]. Both Solar Salt and Hitec<sup>®</sup> mixtures have been used in other heat transfer investigations [71, 79, 92–94].

Other particular references are listed for each property, in order to make a complete review of the cases. The general procedure has involved a global plot of all the correlations found, verifying consistency and agreement among them. The aim of this analysis is to create a set of physical properties for technical use in system codes, computational simulations, or experimental workbenches. Data may be also used to observe dispersion and agreement in properties predictions, before make decisions on critical parameters of design in energy projects.

##### 4.1. Melting point (M.P., °K)

As has already been commented, phase diagrams for molten salts have been continuously investigated and reviewed in order to detect the most useful compositions for each mixture. Table 1 shows different values for melting point parameter for selected or more common molar compositions. In the case of FLiNaBe there are not many experimental values for the usually referred compositions (0.31-0.31-0.38) or (0.33-0.33-0.33).

##### 4.2. Density ( $\rho$ , $\text{kg}/\text{m}^3$ )

In general, a good agreement is verified by all selected mixtures for density correlations. Recommended values are summarized in Table 2, and Fig. 4. Density of FLiBe has been studied and experimented for different compositions since 1956. The slope of the temperature function in the reviewed bibliography agrees with very little variation. Changes in molar percentages do not suppose great differences (e.g., Cantor et al. [39], Janz et al. [51], Powers et al. [100]), with the exception of values offered by Grimes et al. [32] for 50%-50% molar mixture, and recently cited by Korkut and Hançerlioğulları [101]. In this case, a significant disagreement is observed, and correlated values are apparently too large, and the deviation from average is over 11.8 %.

The standard molar composition (0.66-0.34) has been correlated by Blanke et al. [33], Cantor et al. [37] and [40]. Cantor [40] is recommended for

**Table 1:**Melting point ( $^{\circ}\text{K}$ ) proposed or used by different authors for all the studied salts.

	Salt mixture	FLiBe	FLiNaK	FLiNaBe	NaFNaB	FluZirK	CloKMag	Solar Salt	Hitec <sup>®</sup>
Refer.	Usual Composition	(0.66- -0.34)	(0.465-0.115- -0.42)	(0.31-0.31- -0.38)	(0.08- -0.92)	(0.58- -0.42)	(0.68- -0.32)	(0.66- -0.34)	(0.07-0.49- -0.44)
Hoffman and Lones (1955) [21]			727.4						
Cohen and Jones (1957) [95]			727						
Grimes et al. (1958) [32]			727	611					
Thoma (1959) [31]		727	727	588		663			
Cantor et al. (1968) [37]		731			658				
Barton et al. (1971) [96]					654				
Janz et al. (1972) [50]								495	
Cantor (1973) [40]					658				
Janz et al. (1974) [51]		731.9			657				
Janz et al. (1975) [52]							699		
Janz et al. (1978) [47]		729	727		657	693	710		
Vriesema (1979) [97]			727						
Janz and Tomkins (1981) [49]			727				708		415
Abe et al. (1981) [98]		732.1							
Janz and Tomkins (1983) [56]					657				
Mlynariková et al. (2008) [99]					658.7				
Beneš et al. (2010) [89]								496	

use, in agreement with Beneš and Konings [80]. However, this last work does not offer the correct temperature function, differing from the original proposal. So best values are obtained for the range  $T \in [788 - 1094]$  (see Eq. 1). The deviation from the average is around 0.89 % for the mentioned selected correlation, while the global standard deviation for the three plotted values (Fig. 1a) is about 17.3.

$$\rho (\text{kg/m}^3) = 2413.03 - 0.4884 \cdot T(^{\circ}\text{K}) \quad (1)$$

The density of eutectic FLiNaK has been experimentally measured and estimated in different occasions, as reported by Grimes et al. [32], Janz and Tomkins [49], Chrenková et al. [102], Cibulkova et al. [103] and Salanne et al. [104] for several temperature ranges. Williams et al. [60], Ambrosek et al. [105], and Korkut and Hançerlioğulları [101] used Grimes et al. as a reference. However, and as suggested by Beneš and Konings [88], data of Chrenková et al. are near and parallel to ideal density behavior (lightly minor than ideal). Although Salanne et al. estimation is nearest in timeline, it is based on Molecular Dynamics (MD) simulations and the calculated values are more distant if compared with the ideal behavior. An excellent correlation between the function reported by Chrenková and the one from Janz and Tomkins can also be observed; the standard deviation is 17.6 for the six plotted functions (Fig. 1b). Results of Cibulková et al. (Eq. 2) are close to Chrenková et al. Both of them are suggested as best currently values, giving a 0.38 % deviation from average values. Any of this two functions may be extrapolated for the whole range  $T \in [933 - 1170]$ :

$$\rho (\text{kg/m}^3) = 2579.3 - 0.6237 \cdot T(^{\circ}\text{K}) \quad (2)$$

The usually suggested composition for ternary FLiNaBe in recent papers is (0.31-0.31-0.38) or (0.33-0.33-0.33). The nearest correlation found was published by Williams et al. [60], by the method of additive molar volumes. Moreover, Beneš and Konings [80] suggested a different composition for the fuel matrix in MSR using FLiNaBe–AnF<sub>4</sub> (0.203-0.571-0.212-0.013). Other researchers have proposed other expressions, e.g. Grimes et al. [32] for (0.35-0.27-0.38), Powers et al. [100] for (0.20-0.35-0.46), and Khokhlov et al. [106] for (0.22-0.567-0.213) among others. All of them offer a similar

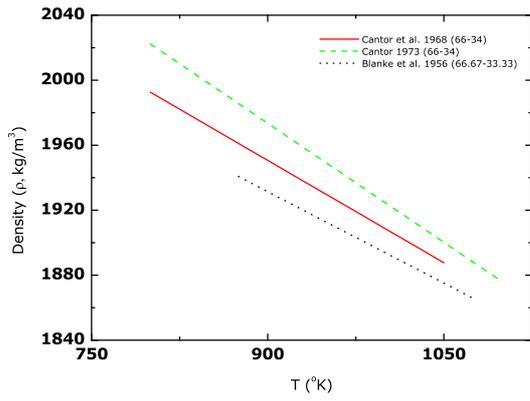
slope (Fig. 1c). The differences among the compositions result in a standard deviation value of 64.9, but estimations like those of Williams et al. (Eq. 3) are in halfway regarding the others (only 0.07 % of deviation from the average) and are suggested for the temperature range  $T \in [800 - 1025]$ :

$$\rho (\text{kg/m}^3) = 2435.85 - 0.45 \cdot T(^{\circ}\text{K}) \quad (3)$$

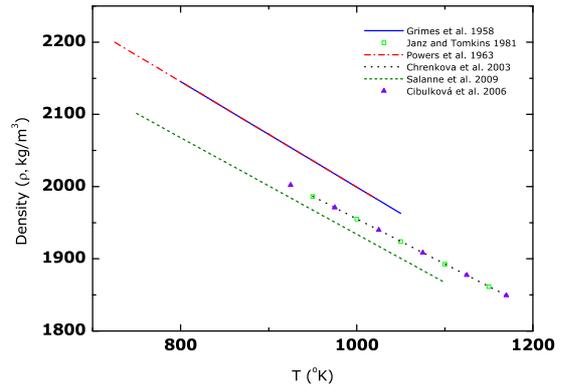
Two correlations have been found for NaFNaB, Cantor et al. [37] and [40]. Both equations are similar (Fig. 2a) giving 1.47 for standard deviation at the overlap interval, and results can be estimated by the second one for  $T \in [673 - 864]$  in order to obtain differences from the average minor than 0.06 %:

$$\rho (\text{kg/m}^3) = 2446.2 - 0.711 \cdot T(^{\circ}\text{K}) \quad (4)$$

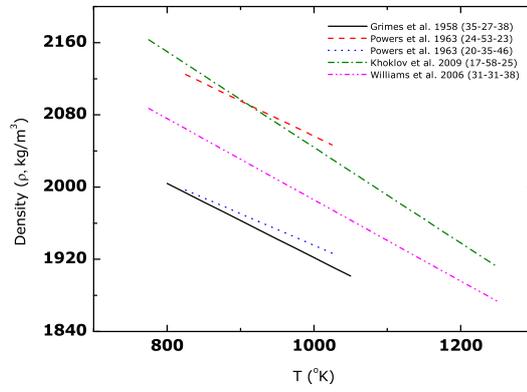
FluZirK is being used recently in latest FHR concepts and proposals with (0.58-0.42) molar relation. For this salt, no correlations were made till 1988 to our best knowledge. Several data were formerly reported by Janz et al. [51] and then by Janz [57] at 1233.2 °K. Different estimations were provided by Darienko et al. [107, 108] from 33 to 80 mol% of ZrF<sub>4</sub>, and also an estimation may be calculated through the kinematic and dynamic viscosity relationship given in Darienko et al. [109]. The graphical analysis of the parameters given in the latter one shows an anomalous value for the 33 mol% ZrF<sub>4</sub> compositions (whose standard deviation is higher, as for the 60 and 80 mol%). But, if we interpolate  $\rho$  between 40 and 45 % for the usual 42 mol% the final curve fits with the others, which shows the accuracy in this molar concentration range. More recently a new correlation was published by Williams [61]. Studying the similarity among them (Fig. 2b), the slope is almost the same in all functions. Darienko et al. results are in a very good agreement in the three reported papers. However, the equation given by Williams is quantitatively far from the others. Indeed, standard deviation grows to 95.8 if taking into account Williams correlation, while deviation goes to 7.34 when calculating this value without this expression. When plotting all values together, proposals of Darienko et al. [108] (Eq. 5) are more coherent to those initially published by Janz et al. and lately by Janz. The deviation from average is about 0.17 % if we ignore Williams correlation; but this value raises to 1.7 % if we use this expression for average estimation. Hence, although



a FLiBe.

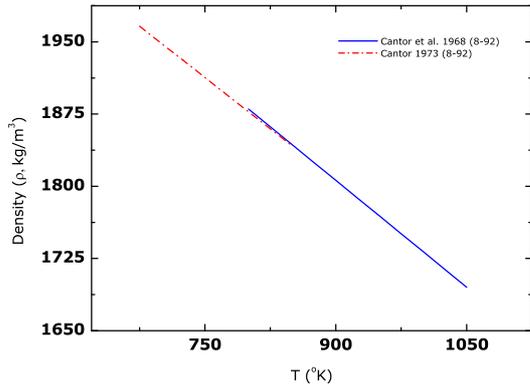


b FLiNaK.

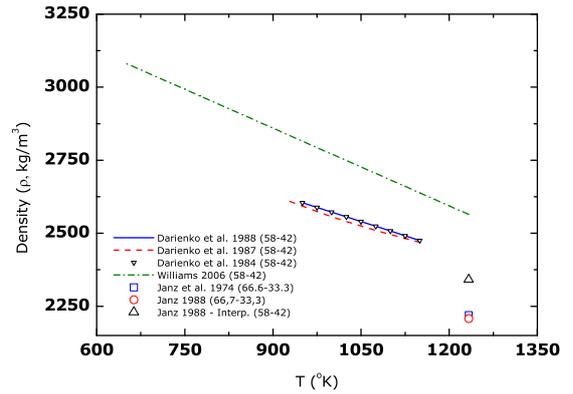


c FLiNaBe.

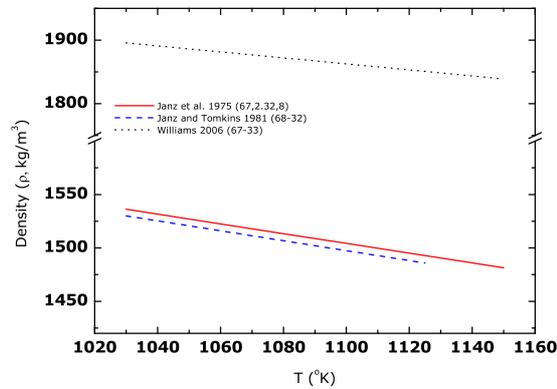
**Fig. 1:** Comparison of density variation with respect to temperature for the salts: FLiBe (a), FLiNaK (b), and FLiNaBe (c). Suggested values for each three salts are very near to average, giving a deviation below 0.9 %.



a NaFNaB.



b FluZirK.



c CloKMag.

**Fig. 2:** Reviewed density correlations for NaFNaB (a), FluZirK (b) and CloKMag (c). A very good agreement is observed for NaFNaB (a). Plot for FluZirK (b) includes two different values correlated by Janz at 1233.2 °K for the (0.67-0.33) molar ratio, and an extrapolated value for the (0.58-0.42) composition. Williams expressions give too high density values in the case of FluZirK and CloKMag mixtures, when compared with other measurements.

Williams expression has been used in recent papers such as Anderson and Sabharwall [74], the most suitable correlation for the range  $T \in [953 - 1150]$  reads as follows:

$$\rho (\text{kg/m}^3) = 3217.44 - 0.6453 \cdot T (\text{°K}) \quad (5)$$

Classical works of Janz and Tomkins [49], Janz et al. [52] included several expressions to estimate the density for CloKMag, from 25 to 42.2 mol% of  $\text{MgCl}_2$ . Also Williams [62] has reported another expression, but values obtained with it are too far from the other ones (Fig. 2c). Standard deviation is about 4.64 if avoiding Williams correlation, while this value raises up to 221.6 when taking into account this expression. Hence, Janz et al. (Eq. 6) is suggested for future works in the range  $T \in [1017 - 1174]$ , with a 0.22 % deviation from average (without Williams):

$$\rho (\text{kg/m}^3) = 2007 - 0.4571 \cdot T (\text{°K}) \quad (6)$$

Parameters for the so called Solar Salt (also known as draw salt) have been either published for the equimolar composition (near 1:1 molar ratio), and for the cheaper commercial mixture (0.64-0.36), or 60-40 wt%. However, there are no significant differences in terms of density. This useful mixture was reported by Janz et al. [50] giving a second order equation in terms of temperature, and which allows to take into account the molar percentage of  $\text{KNO}_3$ . Janz [57] includes linear expressions for the density from 30 to 50 mol% of  $\text{KNO}_3$ . The eutectic composition of this salt was also studied by James and Liu [110], Carling et al. [111] and Nissen [112]. Carling et al. reported two correlations, taking into account the thermal decomposition of nitrate to nitrite at extended time experiments. This maybe the reason of the second order proposal of Janz et al., but calculated values do not exactly agree in both cases. The correlation given by Nissen has been chosen by Zavoico [113] at Sandia National Laboratories, and is the current reference for the System Advisor Model (SAM) at the National Renewable Energy Laboratory (NREL). James and Liu values are slightly below from all the others, and the curve has also a different slope. For the two possibilities given by Carling et al., Nissen appears as the average value (Fig. 3a) with 0.03 % of deviation, and 13.10 as global standard deviation. As the values are so closer for the (0.64-0.36) commercial salt, negligible error is done by using the same correlation for both mixtures. The higher

range of measurement reported by Nissen (Eq. 7) respect to Janz et al. suggests best accuracy. Therefore, for  $T \in [573 - 873]$  and the (0.50-0.50) composition, the Nissen expression is as follows:

$$\rho (\text{kg/m}^3) = 2263.641 - 0.636 \cdot T (\text{°K}) \quad (7)$$

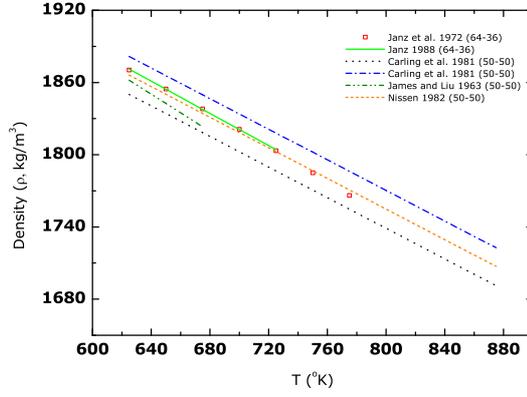
The last salt reviewed is the commercially denoted as Hitec<sup>®</sup>, which has been largely used as HTF by chemical industry. Several studies can be mentioned, with very little differences (Fig. 3b) among their mathematical expression and nearly the same after calculating values, e.g., Kirst et al. [114] (as cited by Gaune [115]), Janz and Tomkins [49], Yang and Garimella [116], Wu et al. [117] and Boerema et al. [118]. Additionally, values of SAM for the Hitec<sup>®</sup> composition [119] have been correlated (Eq. 8). This last system is being commonly used as standard, giving a 0.09 % deviation from the average. Using all the mentioned expressions, a value of 2.55 is calculated as standard deviation. Hence the recommended correlation for the temperature range  $T \in [448 - 773]$  reads:

$$\rho (\text{kg/m}^3) = 2279.799 - 0.7324 \cdot T (\text{°K}) \quad (8)$$

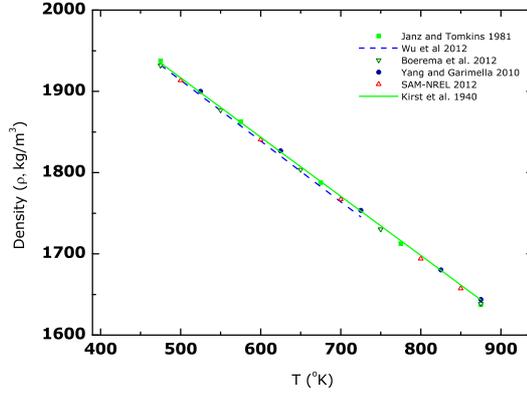
Summarizing for density property, (Table 2) shows the selected functions for each salt with the calculated percentage of deviation from the average values. Best fit is obtained for nitrates, followed by NaFNaB and FLiNaBe salts. Nevertheless, tabulated expressions provide acceptable estimations, even for FLiBe and FLiNaK (which show the higher scattering among reviewed data). A graphical sketch of selected functions is plotted in Fig. 4. This figure provides a quickly view for all the studied salts: FluZirK appears as the heaviest one, while CloKMag is the lightest of them. Molten salt density decreases as temperature increases in all the cases analyzed.

#### 4.3. Dynamic viscosity ( $\eta$ , Pa·s)

Most of selected salts follow an Arrhenius behavior in their temperature range. However, some nitrate salts are susceptible to decompose at high temperatures or extended time heat expositions. The non-Arrhenius behavior has been reported for some chloride mixtures by Boon et al. [120], discussed by Nissen [112] for equimolar Solar Salt, and



a Solar Salt.



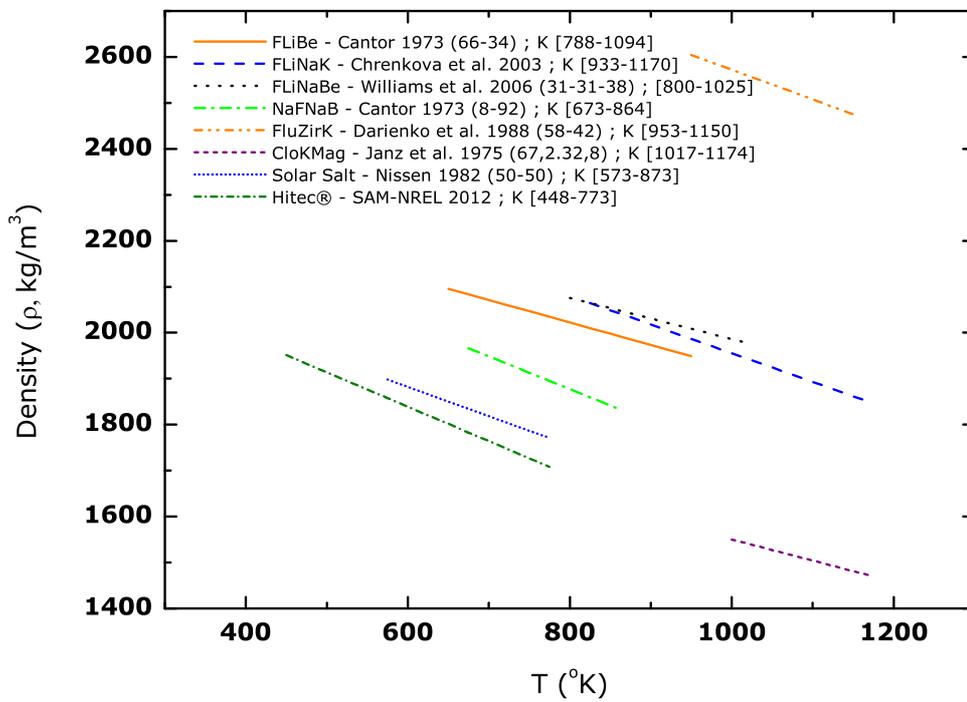
b Hitec<sup>®</sup>.

**Fig. 3:** Density correlations for nitrates: Solar Salt (a), and Hitec<sup>®</sup> mixture (b).

**Table 2:**

Density correlations ( $\text{kg/m}^3$ ) as temperature function suggested for studied salts, including references and deviation from the global average data.

Salt mixture	Reference	Ref. num.	Selected correlation	Temp. Range	% Dev.
FLiBe	Cantor 1973 (0.66-0.34)	[40]	$2413.03-0.4884 \cdot T$	[788-1094]	0.89 %
FLiNaK	Chrenkova et al. 2003 (0.465-0.115-0.42)	[102]	$2579.3-0.624 \cdot T$	[933-1170]	0.38 %
FLiNaBe	Williams et al. 2006 (31-31-38)	[60]	$2435.85-0.45 \cdot T$	[800-1025]	0.07 %
NaFNaB	Cantor 1973 (0.08-0.92)	[40]	$2446.2-0.711 \cdot T$	[673-864]	0.06 %
FluZirK	Darienko et al. 1988 (0.58-0.42)	[108]	$3217.44-0.6453 \cdot T$	[953-1150]	0.17 %
CloKMag	Janz et al. 1975 (0.672-0.328)	[52]	$2007-0.4571 \cdot T$	[1017-1174]	0.22 %
Solar Salt	Nissen 1982 (0.50-0.50)	[112]	$2263.628-0.636 \cdot T$	[573-873]	0.03 %
Hitec <sup>®</sup>	SAM-NREL 2012 (0.07-0.49-0.44)	[119]	$2279.799-0.7324 \cdot T$	[448-773]	0.09 %



**Fig. 4:** Graphical density comparison of the different mixtures studied by selected correlations, showing a similar negative slope.

lately described by Bradshaw [121] for some multi-component nitrates. The Fulcher expression (Eq. 9, also known as VFTH) can be used for correlation in this cases, but several authors still use a simple polynomial regression.

$$\log_{10}(\eta) = A + \frac{B}{T - T_0} \quad (9)$$

Cantor et al. [39] also conjectured about the non-Arrhenius behavior in FLiBe when applying low temperatures, which can be extrapolated for other molten salts. However, working range is higher to preclude this, far enough from the double of temperature of ideal glass transition point.

Around FLiBe viscosity, Salanne et al. [122] explained the reason of pure BeF<sub>2</sub> high viscosity based in a MD study. As BeF<sub>2</sub> concentration increases in a FLiBe solution, different species are created, resulting in a polymer of several BeF<sub>4</sub><sup>2-</sup> units at the highest percentages. Different investigations have been carried out about this physical property. Blanke et al. [33], Cantor et al. [37], Desyatnik et al. [123] measured viscosity for several molar concentrations of BeF<sub>2</sub>, while Cantor et al. [39], Cohen and Jones [95] and Abe et al. [98] studied only one mixture. Williams et al. [60] gave a correlation based on Cantor et al. [37], but the expression is one order of lower magnitude (Fig. 5a). The global standard deviation grows from 0.0004 to 0.0034 when including Williams expression. Janz et al. [51] and Janz and Tomkins [49] already used Cantor et al. correlations for the molten salt database.

Globally, the agreement is nearly perfect [88] among all values with the exception of Williams correlation, showing a rising curve when plotted all data in a X (% BeF<sub>2</sub>), Y (log  $\eta$ ) graph. For T > 1050, although viscosity becomes almost constant, data are near parallel (Fig. 5a). Percentage of deviation from the average is around 4.97 % for Cantor et al. and T  $\in$  [800 – 1050], and near to 7 % for Abe et al. at the same temperature interval. From these results, Abe et al. [98] is suggested for T > 1050 °K, while Cantor et al. [37] (Eq. 10) is selected for the range T  $\in$  [800 – 1050]:

$$\eta(\text{Pa} \cdot \text{s}) = 0.000116 \cdot \exp\left(\frac{3755}{T(^{\circ}\text{K})}\right) \quad (10)$$

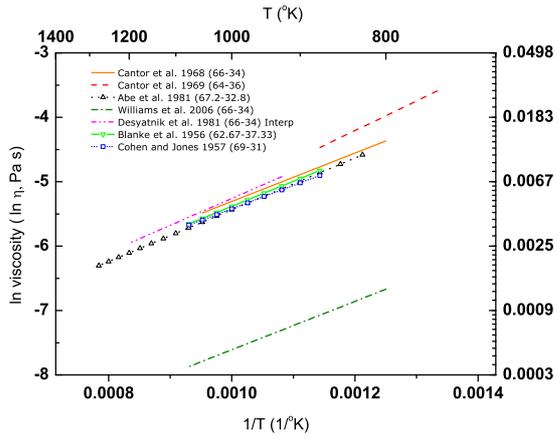
Cohen and Jones [95] offered measurements for FLiNaK viscosity, later reported by Grimes et al. [32], Williams [62], Powers et al. [100], Korkut and

Hançerlioğulları [101]. Vriesema [97] used a different data (kinematic viscosity from a private communication of Oye, H. A.), and these values have been correlated using the same density of his heat transfer experiments. More recently, results provided by Chrenková et al. [102] and correlation listed by Janz and Tomkins [49] give nearly the same numbers (Fig. 5b), and both of them can be over-ranged by the equation of Cibulkova et al. [103] up to 1163 °K. After a comparison, and taking into account the discussed argument for FLiNaK density, we suggest the correlation of Chrenková et al. (Eq. 11) for the range T  $\in$  [773 – 1163] (assuming negligible extrapolation error because of nearness to Cibulková et al. values). However, if using all reviewed data, the deviation from the average raises to a 9.87 % due to the closeness of all values:

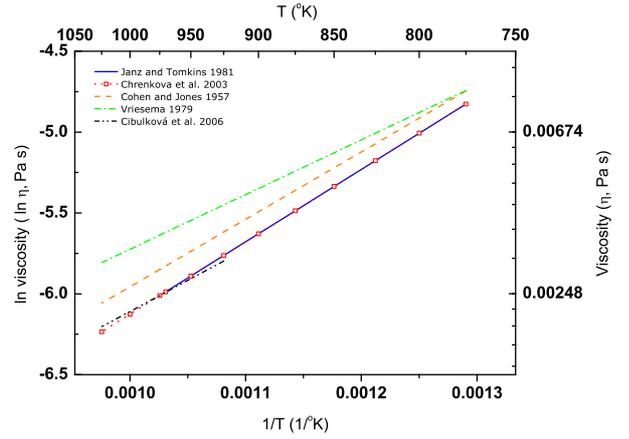
$$\eta(\text{Pa} \cdot \text{s}) = 0.0000249 \cdot 10^{\left(\frac{1944}{T(^{\circ}\text{K})}\right)} \quad (11)$$

The viscosity of FLiNaBe was measured by Cohen and Jones [95] for different compositions, then reported by Grimes et al. [32] for (0.35-0.27-0.38) molar mixture, and also correlations were given by Powers et al. [100] for some other compositions. Khokhlov et al. [106] made calculations using additive law of molar volumes of simple LiF and BeF<sub>2</sub>, with binary LiF – BeF<sub>2</sub> and NaF – BeF<sub>2</sub>. As discussed by Zherebtsov and Ignatiév [67] the experimental data of early ORNL measurements are in good agreement with the more recent values. In addition, Ignatiév et al. [124] plotted a multi-comparison graph showing a very good agreement with the Institute of High Temperature Electrochemistry (IHTE) modeling equations used in the International Science & Technology Center (ISTC) #1606-Project, when temperatures are over 600°K. MSR application has been reviewed by Beneš and Konings [80] pointing to a recommended molar composition (0.22-0.5666-0.2133), but they reproduced the equation corresponding to (0.1433-0.59-0.2667) which must be taken into account. In other nuclear applications, the most referred promising mixture is near to the equimolar. To the author's knowledge, no investigation has been performed on this particular salt.

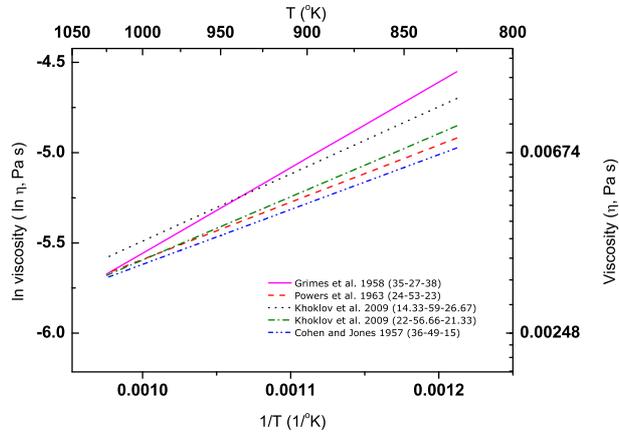
All reviewed correlations show a good agreement for temperatures higher to 950°K (Fig. 5c), where the composition dependence do not imply so much scattering. Therefore, searching for a global expression, accuracy of predictions for the range T  $\in$  [823 – 1023] is legitimated with the following



a FLiBe.



b FLiNaK.



c FLiNaBe.

**Fig. 5:** Comparison of viscosity variation with respect to temperature for FLiBe (a), FLiNaK (b), and FLiNaBe (c), according to empirical correlations of different studies and authors.

approach, which give a 12.35 % of deviation from the overall average:

$$\eta (\text{Pa} \cdot \text{s}) = 0.0000338 \cdot \exp\left(\frac{4738}{T(^{\circ}\text{K})}\right) \quad (12)$$

Two formulations have been reported for NaFNaB at different temperature ranges. Cantor et al. [37] made a extrapolation for the behavior of sodium iodide based in experimental values for NaFNaB published by Wittenberg et al. [125] at Mound Laboratory. This latter correlation was defined for  $T \in [400 - 700]$ . Thereafter Cantor [38] gave a new equation for the range  $T \in [682 - 810]$ , which has been subsequently cited in later reports as Cantor [40], Janz et al. [51], Janz [57]. Janz and Tomkins [56] expanded the temperature window between 799-906 °K, but reviewing this reference there are no apparently reasons to include this new temperature range (Fig. 6a). So we suggest the function reported by Cantor [38] but only for the range  $T \in [682 - 810]$ . Reviewed data give 0.003 for standard deviation at the overlap interval, while suggested expression shows a 8.97 % of deviation from average in the same interval:

$$\eta (\text{Pa} \cdot \text{s}) = 0.0000877 \cdot \exp\left(\frac{2240}{T(^{\circ}\text{K})}\right) \quad (13)$$

Janz et al. [51] proposed viscosity values for two compositions of FluZirK at 1253°K. Later, Janz [57] reported an expression to calculate this property at the same temperature for a wide range of compositions, between 0-33,3 mol% of ZrF<sub>4</sub>. More recently, Darienko et al. [109] measured FluZirK viscosity for concentrations from 0 to 80 mol% of ZrF<sub>4</sub>, and Williams et al. [60] proposed a exponential correlation for the promising (0.58-0.42) composition as a function of temperature. Differences have been analyzed, after interpolating data from Darienko et al. for the same (0.58-0.42) mixture (Eq. 14). Expression reported by Williams et al. shows noticeably lower values the others, which means a 0.0022 global standard deviation and about 82.6 % of deviance from the calculated average. However, our interpolation points to very similar values as previously listed by Janz and Janz et al. (Fig. 6b). Hence, although Williams correlation has been widely used and cited in many recent reports and papers (e.g., Beneš et al. [8], Williams [61], Samuel [66], Anderson and Sabharwall [74], Kubíková et al. [126], Scheele and Casella [127], Sabharwall et al. [128, 129]), the following interpolated expression

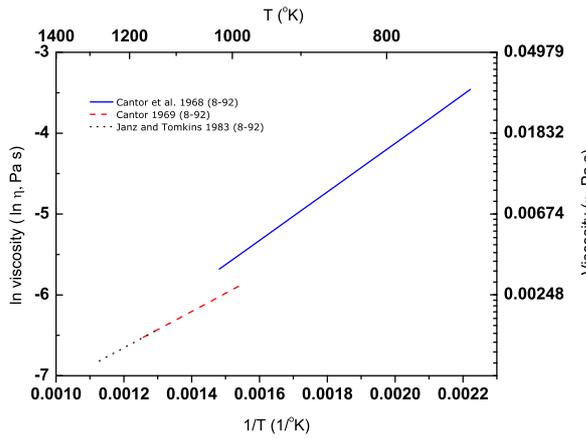
based in values of Darienko et al. is suggested in present work for the range  $T \in [921 - 1185]$ , reducing deviation from the average to 45.24 % when data of Williams are still included in calculations:

$$\eta (\text{Pa} \cdot \text{s}) = 0.0001084 \cdot 10^{\left(\frac{1581.2}{T(^{\circ}\text{K})}\right)} \quad (14)$$

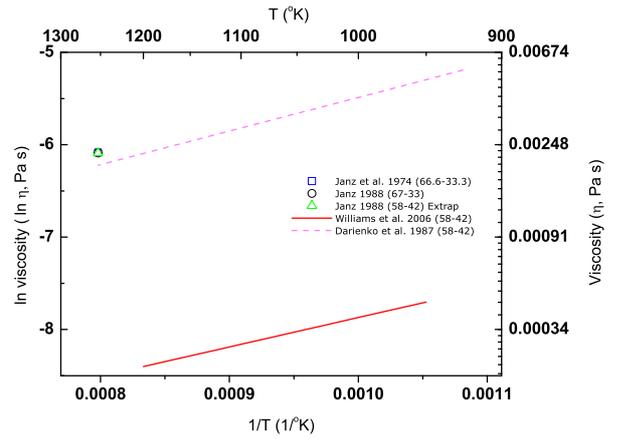
The viscosity of binary chloride CloKMag was correlated for different molar concentrations by Janz et al. [52] and Janz and Tomkins [49], giving one third order polynomial expression (including the usually accepted 0.68-0.32 composition) and other standard Arrhenius formulation. Also Janz [57] and Williams [61] reported two different correlations for the mentioned (0.68-0.32) composition. By calculating values we have compared all the possibilities (Fig. 6c). Williams expression appears with a lower order of magnitude, giving a global  $6.2 \cdot 10^{-4}$  standard deviation, while all the others are in good agreement. Standard deviation is reduced to  $5 \cdot 10^{-5}$  when ignoring Williams correlation. Therefore, Janz [57] (Eq. 15) is the recommended correlation for the whole range  $T \in [900 - 1030]$ , which means a 3.95 % of deviation from the average:

$$\eta (\text{Pa} \cdot \text{s}) = 0.0001408 \cdot \exp\left(\frac{2261.3}{T(^{\circ}\text{K})}\right) \quad (15)$$

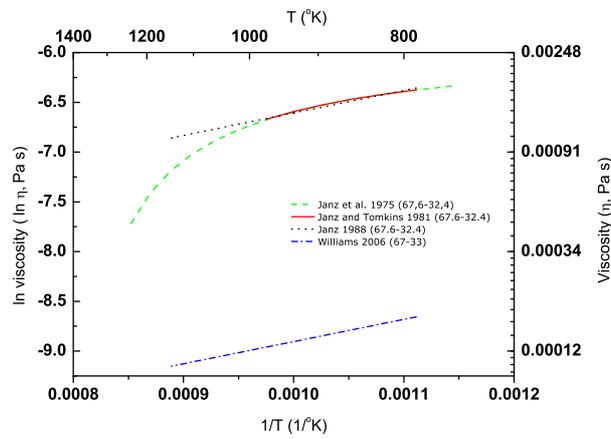
For the binary Solar Salt, negligible differences were found for viscosity between equimolar and commercial compositions. Initial measurements made by Murgulescu and Zuca were reported by Janz et al. [50] for the range  $T \in [525 - 725]$  after a critical review. These correlations included the 0, 25, 50, 75 and 100 mol% of NaNO<sub>3</sub>, with Arrhenius form for the two first and a third order polynomial for the others. New Arrhenius expressions were reported by Janz [57] for the same molar composition cases, revising again the measurements mentioned above. For the equimolar salt, experimental data was given by Nissen [112], making a polynomial correlation used lately by Zavoico [113] for the Basis Document of the *Solar Power Tower*. Data offered in SAM NREL [119] have been also correlated, using polynomial (Eq. 16) and Arrhenius forms. After a comparison, data chart of SAM give the same values as Nissen for a shorter temperature range (Fig. 7a). A value of 6E-05 has been calculated as global standard deviation. Apparently, the equation given by Nissen shows the high temperature behavior of nitrates regarding to decomposition (as mentioned previously), while this



a NaFNaB.

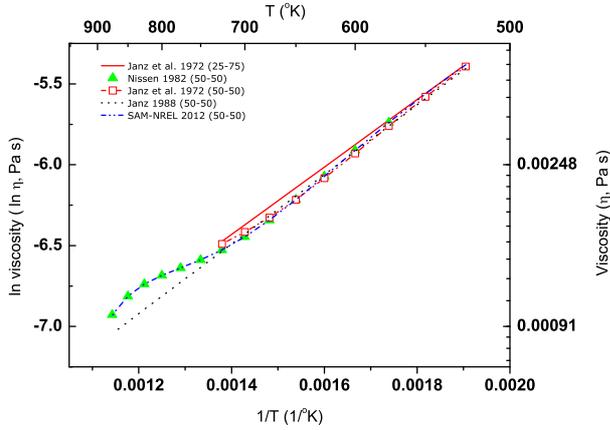


b FluZirK.

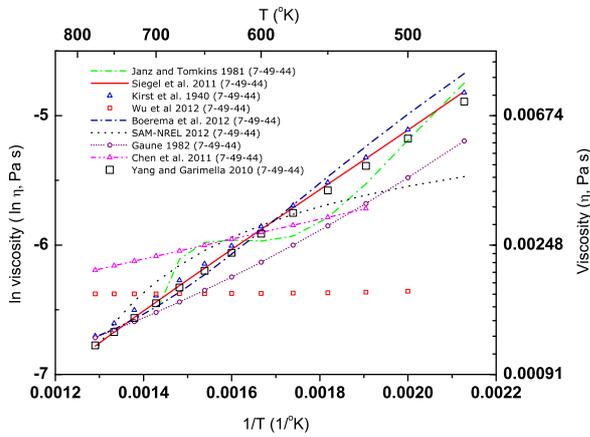


c CloKMag.

**Fig. 6:** Agreement among the reviewed correlations for NaFNaB viscosity (a). Graphical comparison (b) among different proposed functions and values correlated by Janz at 1253 °K for FluZirK viscosity. Functions of temperature for CloKMag viscosity (c). The last two (b & c) include the anomalous values obtained with the expressions reported by Williams.



a Solar Salt.



b Hitec<sup>®</sup>.

**Fig. 7:** Graphical comparison of viscosity functions for Solar Salt (a) and Hitec (b).

chemical mechanism is not reflected in the Arrhenius form. In any case, the effect of molar composition is almost negligible. The following polynomial correlation is suggested for the whole range  $T \in [573 - 873]$ , using data of Nissen, and giving a 2.33 % of deviation from the average values:

$$\eta (\text{Pa} \cdot \text{s}) = 0.07543937 - 2.77 \cdot 10^{-4} \cdot T(^{\circ}\text{K}) + 3.49 \cdot 10^{-7} \cdot T^2(^{\circ}\text{K}) - 1.47 \cdot 10^{-10} \cdot T^3(^{\circ}\text{K}) \quad (16)$$

Different correlations have been published for the Hitec<sup>®</sup> mixture. Kirst et al. [114] proposed a exponential behavior for temperatures between 473-773 °K, while Gaune [115] used a second order func-

tion of temperature for the same range, with Arrhenius global form. Janz and Tomkins [49] reported a third order polynomial correlation, but it shows a excessive slope at high temperatures regarding the others (probably due to thermal decomposition of nitrates). More recently, other authors have used other expressions, e.g. Yang and Garimella [116], Boerema et al. [118], Siegel et al. [130], Chen et al. [131] and Wu et al. [117]. Values of SAM NREL [119] have been also correlated to a linear expression in present work (Eq. 17). After a comparison, Siegel et al. gives approximately the same values as Kirst et al. (Fig. 7b).

Boerema et al. gives the higher viscosity of all the correlations at low temperatures, as well as Wu et al. gives the lowest one, and shows a crescent function of temperature. The expression provided by Yang and Garimella is selected as the most representative. This correlation was obtained by correlating Coastal Chemical data, giving values in coherence with Solar Salt (also a nitrate mixture). Standard deviation is calculated, giving a global  $3 \cdot 10^{-4}$  value. Hence, the suggested expression for the range  $T \in [525 - 773]$ , with 3.65 % of deviation from the average, is as follows:

$$\eta (\text{Pa} \cdot \text{s}) = \exp(-4.343 - 2.0143 \cdot ((\ln(T(^{\circ}\text{K}) - 273) - 5.011)) \quad (17)$$

In short, for dynamic viscosity, suggested expressions are listed in Table 3 for all the studied mixtures. Data for Solar salt give the smallest deviation from average values, while correlations for NaFNab show the higher scattering by far (when compared with the other salts). A graphical comparison is plotted in Fig. 8; all functions show a descent slope. Data for FLiBe are valid in a large temperature range, while viscosity correlations for the remainder mixtures are only useful for a short interval.

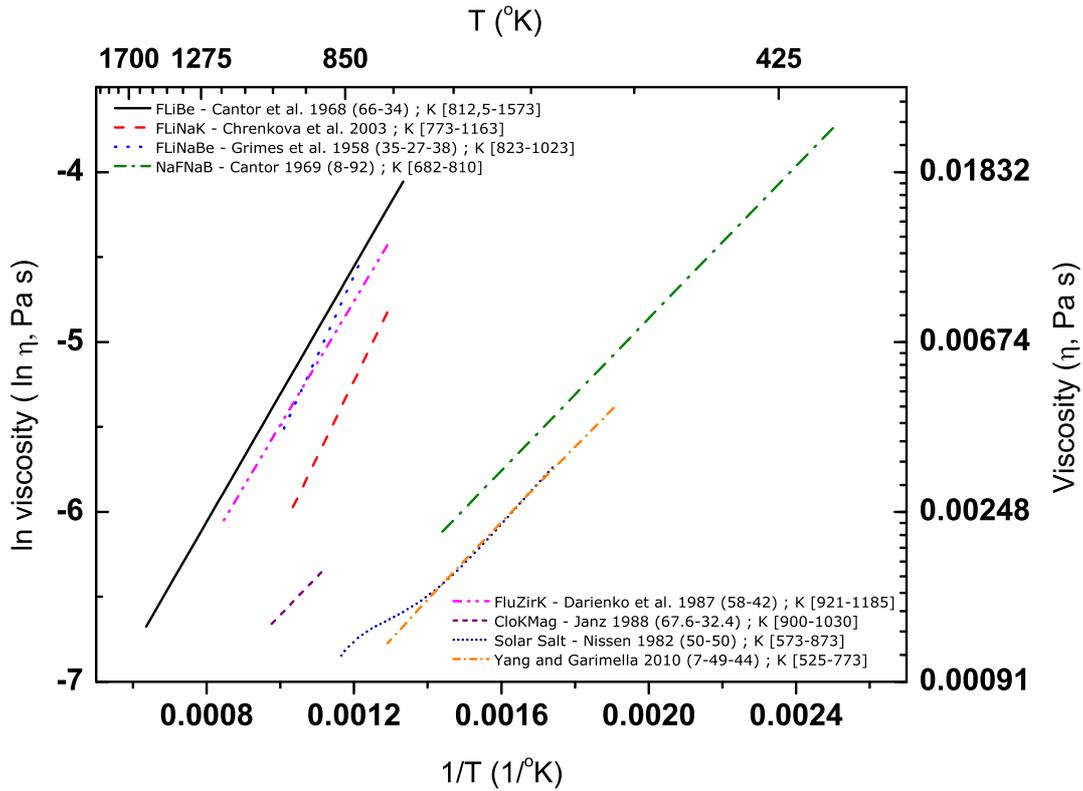
#### 4.4. Thermal conductivity ( $\lambda$ , $W \cdot m^{-1} \cdot K^{-1}$ )

The measurement of thermal properties in molten salts has been pointed out by many authors as an arduous task, specially in the case of conductivity. At high temperatures, there are important uncertainties associated to the heat transfer mode. Nieto de Castro [132] also identified the combined effect of factors involved in this kind of measurement, analyzing the different methods available: (i)

**Table 3:**

Viscosity correlations (Pa·s) as temperature function, including final deviation from the average values after ignoring both too high as too low values.

Salt mixture	Reference	Ref. Num.	Selected correlation	Temp. Range	% Dev.
FLiBe	Cantor et al. 1968 (0.66-0.34)	[37]	$1.16 \cdot 10^{-4} \cdot \exp(3755/T)$	[800-1050]	4.97 %
FLiNaK	Chrenkova et al. 2003 (0.465-0.115-0.42)	[102]	$2.49 \cdot 10^{-5} \cdot 10^{(1944/T)}$	[773-1163]	9.87 %
FLiNaBe	Grimes et al. 1958 (0.35-0.27-0.38)	[32]	$3.38 \cdot 10^{-5} \cdot \exp(4738/T)$	[823-1023]	12.35 %
NaFNaB	Cantor 1969 (0.08-0.92)	[38]	$8.77 \cdot 10^{-5} \cdot \exp(2240/T)$	[682-810]	8.97 %
FluZirK	Darienko et al. 1987 (0.58-0.42)	[109]	$1.084 \cdot 10^{-4} \cdot 10^{(1581.2/T)}$	[921-1185]	45.24 %
CloKMag	Janz 1988 (0.676-0.324)	[57]	$1.408 \cdot 10^{-4} \cdot \exp(2261.3/T)$	[900-1030]	3.95 %
Solar Salt	Nissen 1982 (0.50-0.50)	[112]	$0.075439 - 2.77 \cdot 10^{-4} \cdot (T-273)$ $+ 3.49 \cdot 10^{-7} \cdot (T-273)^2$ $- 1.474 \cdot 10^{-10} \cdot (T-273)^3$	[573-873]	2.33 %
Hitec <sup>®</sup>	Yang and Garimella 2010 (0.07-0.49-0.44)	[116]	$\exp(-4.343 - 2.0143 \cdot (\ln(T-273) - 5.011))$	[525-773]	3.65 %



**Fig. 8:** Global plot of selected expressions of viscosity for the different mixtures studied. A descent slope is showed by all the salts. The lowest viscosity values, in studied temperature intervals, correspond to nitrates and chlorides.

the sample purity and homogeneity, (ii) thermal stability of salt, (iii) interaction between the sample and both the surrounding gaseous atmosphere and the container material, (iv) temperature measurement sensor, and (v) other simultaneous heat transfer mechanisms such as convection and radiation. Some interesting thought was exposed by Nunes et al. [14], in order not to design under or over-dimensioned heat exchangers or industrial equipments. DiGuilio and Teja [133] discussed about the influence of electrical charges, the pressure dependence, the presence of a saturation curve, and the effect of size of ions in salt families. The method of measurement is also a key issue for molten salts. This circumstance was also argued by Diguilio and Teja and by Nieto de Castro, advising about examples of positive or negative slope for the same salt by different authors. In fact, Diguilio and Teja reported erroneous slope values for some nitrates and chlorides.

Among correlation methods, many formulations have been developed and reported with different assumptions, e.g., Bridgman and Kindcaid-Eyring equation [134], Mean-ionic-weight and Rao-Turnbull correlations [135], and Gustafsson and Rough-Hard-Sphere models [133]. Khokhlov et al. [106] proposed a standard function of temperature and molecular weight for multicomponent fluorides. In order to check its behavior, some correlations have been extended for all the selected salts in the present work.

Both experimental or estimated data have been found in the literature for the selected salt mixtures. After a global comparison, a low temperature dependence is showed (Fig. 9). Rao-Turnbull [136, 137] prediction shows acceptable values for melting point, but is very sensitive to the average number of ions per mole (a proposal of ions is listed in Cooke et al. [134] for different mixtures). The assumptions for this correlation made by Williams et al. [60] have been recalculated, obtaining different values other than in this work. Khokhlov et al. [106] functions appears always with a positive slope, which is not true for some mixtures, and the calculated values seem to be only coherent with measurements for fluorides and chlorides (but not with fluoborates or nitrates).

For FLiBe, we have plotted results and proposals from Cantor et al. [37], Williams et al. [60], Cooke et al. [134, 138], Kato et al. [139] and Khokhlov et al. [106], and recalculated Rao-Turnbull correlation. As the temperature dependence is relatively

low, we agree with Beneš and Konings [80] recommending  $\lambda = 1.1 \text{ W} \cdot \text{m}^{-1} \cdot \text{°K}^{-1}$ , with a 0.29 % deviation from the average value.

FLiNaK investigations show a wide range of data from 0.6 to  $4.5 \text{ W} \cdot \text{m}^{-1} \cdot \text{°K}^{-1}$ . The Rao-Turnbull value moves from 0.7 to 1.58 when the number of ions is changed between 1 or 2. Several measurements and estimations have been compared: Grele and Gedeon [20], Hoffman and Lones [21], Grimes et al. [32], Janz and Tomkins [49], Vriesema [97], Powers et al. [100], Kato et al. [139], Ewing et al. [140], Smirnov et al. [141] and Khokhlov et al. [106]. Kato et al. values are derived from thermal diffusivity data. Ewing et al. showed decreasing transmission coefficients, which was explained due to solution of container components.

For the usual composition Beneš and Konings [80] conclusions recommended a linear equation with positive slope. However, DiGuilio and Teja [133] discussed this behavior for the alkali halides (descent slope). In any case, there is good agreement among Kato et al. [139] and Smirnov et al. [141]. Therefore, a constant value of  $\lambda = 0.85 \text{ W} \cdot \text{m}^{-1} \cdot \text{°K}^{-1}$  is suggested in agreement with this two last authors and the expression of Khokhlov et al. [106]. The standard deviation is too high if values of Grele and Gedeon [20], Hoffman and Lones [21], Grimes et al. [32], Powers et al. [100] and Janz and Tomkins [49] are used in calculations, growing up to 1.29; but it decreases till 0.21 when this anomalous values are ignored. Using all data to compute the average the deviation of suggested value is about 51.29 %, but it only 10.78 % when anomalous values are not taken into account.

There are few estimations for FLiNaBe mixture, and most of them are for different compositions. Grimes [35, 142], Ignatiév et al. [143] and Khokhlov et al. [106] have been revised in the present work. Because of the lack of experimental data, a constant value of  $\lambda = 0.70 \text{ W} \cdot \text{m}^{-1} \cdot \text{°K}^{-1}$  is selected for the (0.31-0.31-0.28) mixture, which is coherent with Rao-Turnbull correlation with  $n=1$ . The deviation from average is around 18.21 % for the suggested heat capacity.

Several values have been published for NaFNaB with (0.08-0.92) molar composition, such as Cantor et al. [37], Cooke et al. [134, 138], Grimes [142] and Khokhlov et al. [106]. Analyzing the proposed functions, thermal conductivity can be evaluated by Rao-Turnbull equation ( $n=2$ ), giving  $\lambda = 0.47 \text{ W} \cdot \text{m}^{-1} \cdot \text{°K}^{-1}$  with 0.44 % of deviation from average.

To our best knowledge, no measurements about FluZirK have been published. A standard value, for (0.58-0.42) molar composition, can be estimated in coherence with Rao-Turnbull expression ( $n=1$ ) and Khokhlov et al. correlation. Hence  $\lambda = 0.30 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$  is found to be a good constant value (19.61 % of deviation from the average).

The thermal conductivity of CloKMag have been reported by Janz and Tomkins [49] for the (0.66-0.34) and (0.71-0.29) molar compositions. Williams et al. [60] calculated a value of  $0.39 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$  by simple mole-fraction average of the pure-compound data, which has been recently suggested by Anderson and Sabharwall [74]. By using again the Rao-Turnbull estimation ( $n=2$ ,  $n=1$ , and  $n=1.5$ ), the values obtained are 0.55, 0.25 and  $0.40 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$  respectively. The estimation function of Khokhlov et al. [106] is near parallel to data obtained by Janz and Tomkins [49] correlation, and agrees with Rao-Turnbull ( $n=1.5$ ) and Williams et al. calculations. Therefore, a constant value for thermal conductivity of  $\lambda = 0.55 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$  is suggested with temperature independence, in coherence with Rao-Turnbull estimation for  $n=2$ . Deviation from average is about 14.02 %, and 0.16 is the computed standard deviation.

The scattering of data is even larger for nitrate compositions, as they have been studied extensively. Thermal decomposition of this kind of salts must be taken also into account, which can be significantly enhanced by controlling the atmosphere, as recently discussed by Olivares [144]. Several reports for Solar Salt (with different molar compositions) have been reviewed, and also the SAM data have been correlated by linear regression: Janz et al. [48], McDonald and H. Ted Davis [145], Omotani et al. [146], Tufeu et al. [147] and Zavoico [113]. According to DiGuilio and Teja [133], temperature dependence must show negative slope. This feature is only followed by Omotani et al. and Tufeu et al. expressions. Khokhlov et al. [106] function does not match the measured values. In general, correlated values give a maximum of 0.58 and a minimum of  $0.42 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$  for the range  $T \in [600 - 730]$ , and Rao-Turnbull gives  $0.47 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ . The latter value agrees with Tufeu et al. and Omotani et al. correlations, and DiGuilio and Teja [133] predictions. Hence, a constant  $\lambda = 0.45 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$  is found to be a good choice, showing a 10.12 % of deviation from average.

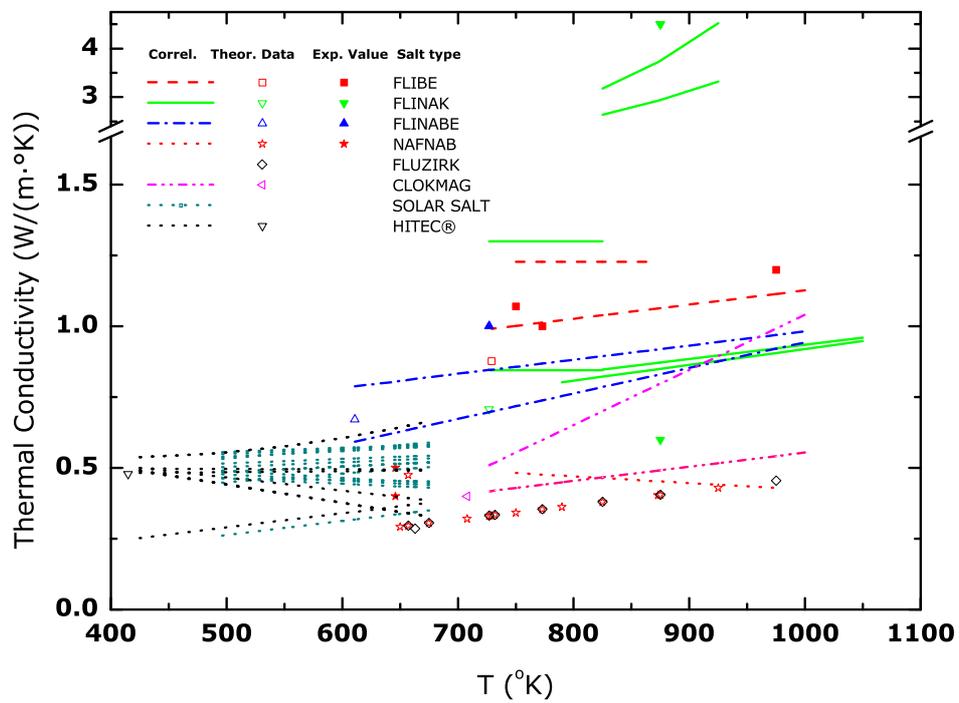
Finally, Hitec<sup>®</sup> correlations have been reviewed.

SAM database has been also accessed, in order to compare values with Janz and Tomkins [49], Yang and Garimella [116], Wu et al. [117], Cooke et al. [134], Tufeu et al. [147], Omotani and Nagashima [148], Santini et al. [149] and Boerema et al. [118]. The plotted functions of temperature show a dispersion around an average value, which is close to Rao-Turnbull estimation for  $n=2$  and giving  $\lambda = 0.48 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ . The latter is also very close to Tufeu et al. and Omotani et al. values. The global standard deviation for this property and salt has been computed about 0.097. Although there is a general disagreement among most recent reports, a suggested value of  $\lambda = 0.48 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$  is also in coherence with DiGuilio and Teja [133] arguments for  $\text{KNO}_3$  and nitrate mixtures. The deviation from average for this last value is calculated to be around 4.36 %.

Trying to make a brief summary for this property, Fig. 9 shows a global view of reviewed values for thermal conductivity and all studied salts. Although most of mixtures shows a high scattering when studied separately, some conclusions can be made with a general overview. For example, most of data are bounded into the range  $0.25 - 1.30 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ . In case of nitrate-nitrite salts, this interval can be reduced to  $0.25 - 0.70 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ , with independence of temperature value. For fluorides, data are bounded into  $0.25 - 1.30 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$  if anomalous values reported by Grele and Gedeon [20], Hoffman and Lones [21] and Janz and Tomkins [49] for FLiNaK are ignored. In general, more tests are needed so as to assess better or newer correlations for salts at high temperature conditions. Hence, with currently knowledge, a constant value may be a good option to get acceptable accuracy in calculations (Table 4).

#### 4.5. Heat capacity ( $C_p$ , $J\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$ )

Khokhlov et al. [106] reported a general correlation for multi-component fluorides. In order to assess further possibilities, this expression have been made extensive to the other molten salts in this work. Some other functions of temperature have been found for certain salts as plotted in Fig. 10. Cantor [36] proposed the assumption of temperature independence for this property, because of the small accuracy observed in experiments. Cantor modified the Dulong-Petit expression for molten fluorides, using an average value for heat capacity



**Fig. 9:** Graph summarizing the thermal conductivity correlations and estimated values proposed by different authors at different temperatures. Legend inside the graph shows: Lines defining different correlations reported for each kind of salt; Symbols representing point values for theoretical estimations (open symbols), or reported experimental measurements (filled symbols).

**Table 4:**

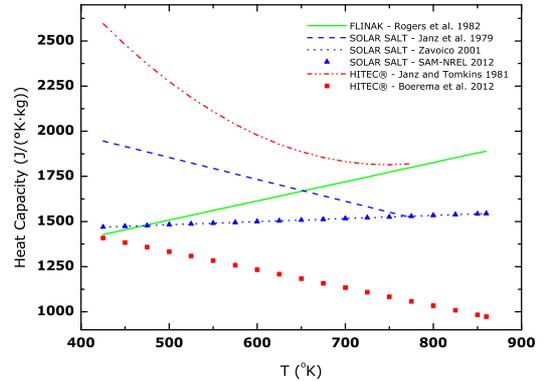
Summary of suggested constant values for thermal conductivity ( $\lambda$ ,  $\text{W} \cdot \text{m}^{-1} \cdot ^\circ\text{K}^{-1}$ ), with temperature independence. Table include the calculated standard deviation for studied data, and % of deviation of selected values from the average. Deviation from average for FLiNaK is too high if anomalous high values are not avoided in calculations (51.29 %), but decrease to 10.78 % when this values are not used.

Salt mixture	Constant value	Stand. Dev.	% Dev.
FLiBe	1.10	0.096	0.29 %
FLiNaK	0.85	0.206	10.78 %
FLiNaBe	0.70	0.105	18.21 %
NaFNaB	0.47	0.016	0.44 %
FluZirK	0.30	0.054	19.61 %
CloKMag	0.55	0.163	14.02 %
Solar Salt	0.45	0.067	10.12 %
Hitec <sup>®</sup>	0.48	0.097	4.36 %

per atom (33.472 J/°K). This proposal was used by Williams et al. [60], making a comparison with measurements at 973 °K. A global graph is showed in Fig. 11, including reported and estimated values. The suggested values for each salt are summarized in Table 5.

For FLiBe, values were reported by Grimes et al. [32] for (0.69-0.31) and (0.50-0.50) compositions at 973 °K. Cantor et al. [37] (from an internal report of Hoffman and Lones) and Douglas and Payne [150] also gave values for the promising (0.66-0.34) mixture. Heat capacity of FLiBe can be also obtained of Kato et al. [139] from thermal diffusivity. By comparison, we propose Cantor et al. [37] as a constant value, giving  $C_p = 2385 \text{ J} \cdot \text{kg}^{-1} \cdot ^\circ\text{K}^{-1}$  and a 5.26 % of deviation from the average.

First found data for FLiNaK were published by Poppendick [22], and also by Powers and Blalock at ORNL [151] using a Bunsen calorimeter. Grele and Gedeon [20] used  $2092 \text{ J} \cdot \text{kg}^{-1} \cdot ^\circ\text{K}^{-1}$  with temperature independence, based on data reported by Poppendick. Other proposals were made by Janz and Tomkins [49], Kato et al. [139] and Salanne et al. [104] (this last, from a first-principles deter-



**Fig. 10:** Correlations found for heat capacity in the case of FLiNaK (—), Solar Salt (---, ···, ▲) and Hitec<sup>®</sup> (-·-, ■) mixtures and usual compositions.

mination). Kato et al. values are based on thermal diffusivity data. Vriesema [97] used a constant value of  $1890 \text{ J} \cdot \text{kg}^{-1} \cdot ^\circ\text{K}^{-1}$ . Rogers et al. [152] also reported a correlation for this property with positive slope, as showed in Fig. 10. Published data are higher than ideal behavior in agreement with Beneš and Konings [80]. After verifying the strong correlation among the constant reported data (standard deviation of 208.46), a heat capacity of  $C_p = 1880 \text{ J} \cdot \text{kg}^{-1} \cdot ^\circ\text{K}^{-1}$  is suggested to be a good value with 6.07 % of deviation from average.

For the molten FLiNaBe, several values have been reported for different molar compositions. However, none of them for the equimolar or (0.31-0.31-0.38) compositions. Grimes et al. [32] gave  $2467 \text{ J} \cdot \text{kg}^{-1} \cdot ^\circ\text{K}^{-1}$  value for (0.35-0.27-0.38) at 973°K, and Powers et al. [100] reviewed for different molar percentages at the same temperature, as well as Grimes [35] at 727 °K. A global standard deviation of 158.15 has been calculated for reviewed data. For the selected composition (0.31-0.31-0.38), we suggest a constant heat capacity of  $C_p = 2200 \text{ J} \cdot \text{kg}^{-1} \cdot ^\circ\text{K}^{-1}$  which has a 0.48 % of deviation from average.

No measurements have been found for FluZirK. Only estimated values can be obtained by the mentioned Khokhlov et al. [106] and Cantor-Dulong-Petit correlations. A constant heat capacity of  $C_p = 1000 \text{ J} \cdot \text{kg}^{-1} \cdot ^\circ\text{K}^{-1}$  is prudently proposed with 5.36 % of deviation from the average of estimations.

Only two references have been found for NaFNaB

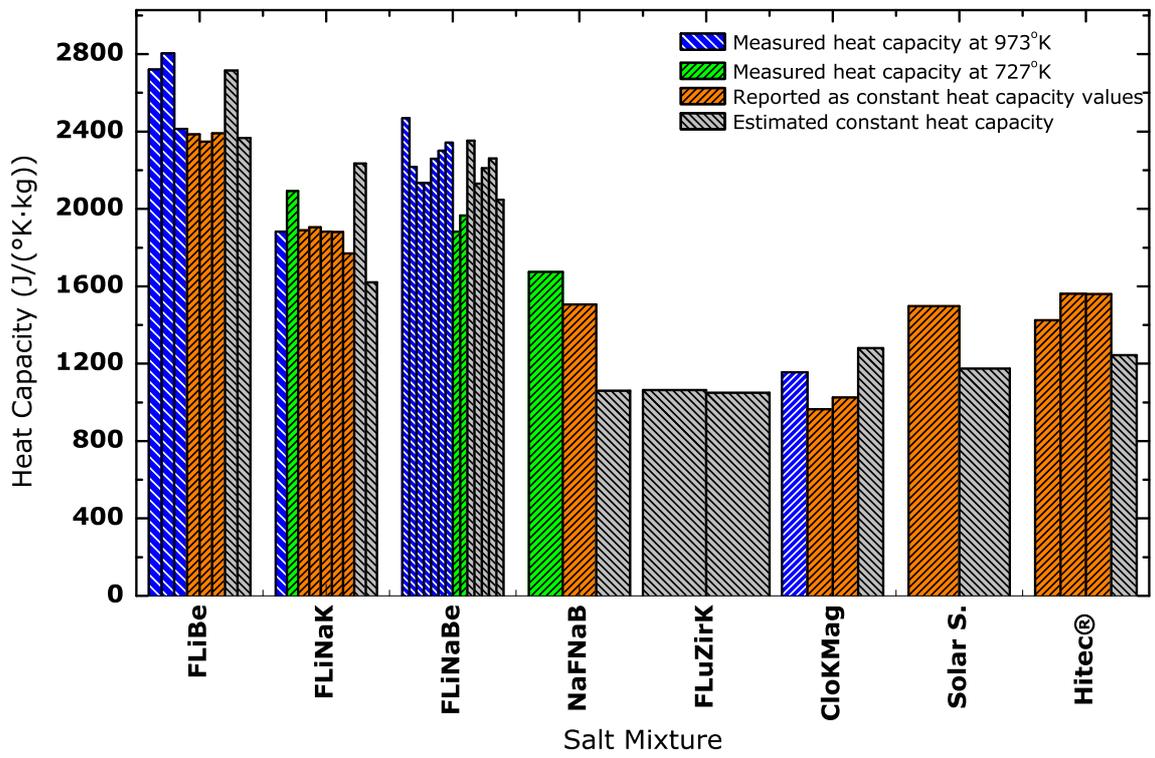


Fig. 11: Global comparison of reported values of heat capacity for selected salts and different compositions.

(0.08-0.92). Grimes [35] reported a value of  $1674 \text{ J}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$ , and as mentioned in Cantor et al. [37], Dworkin measured it giving  $C_p = 1506 \text{ J}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$ , which is the suggested value for this salt. Calculated deviation from the average is about 6.56 % in this case.

For CloKMag (0.666-0.334) and (0.418-0.582) compositions, Janz and Tomkins [49] reported 964 and  $1026 \text{ J}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$  respectively. Williams [61] also reported a measured value at  $973 \text{ K}$  of  $C_p = 1155 \text{ J}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$  for the mixture (0.67-0.33), which is suggested as good value (4.42 % of deviation from average).

The nitrate and nitrite salts, in addition to the correlations plotted in Fig. 10, have been reported several times elsewhere (e.g., Janz et al. [48], Janz and Tomkins [49], among others). The heat capacity of equimolar Solar Salt was fixed in 1498  $\text{J}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$  by Tufeu et al. [147]. Although the correlated SAM data NREL [119] and the expression reported by Zavoico [113] are exactly the same, some differences can be found for temperature dependence if compared with Janz et al. [48]. Agreement among the different investigations analyzed (standard deviation about 148.99) suggests that the actualized values offered by SAM may be a good choice with a 2.36 % of deviation from the global average, using the following expression:

$$C_p (\text{J}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}) = 1396.044 + 0.172\cdot T (\text{K}) \quad (18)$$

Finally, correlations for the commercial Hitec<sup>®</sup> have been reported by Janz and Tomkins [49], Hoffman and Cohen [153] and Boerema et al. [118], using different expressions. Constant values were reported by Wu et al. [117] and Yang and Garimella [116], and this latter gives the same number as found in the SAM database. The computed standard deviation is 473.23 when using all reviewed data. A value of  $C_p = 1560 \text{ J}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$  is suggested due to the agreement of recent proposals with early measurements, giving a 2.45 % of deviation from average.

## 5. Conclusions

The future use of molten salts as coolants or HTF requires a previous checking of transport and thermal behaviors. The use of computational packages allows validation of preliminary designs, and

**Table 5:**

Heat capacity ( $\text{J}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$ ), suggested as constant value with temperature independence, for the reviewed salts. A function is only offered for the Solar Salt mixture.

Salt mixture	$C_p$ value	Stand. Dev.	% Dev.
FLiBe	2385	191.89	5.26 %
FLiNaK	1880	208.46	6.07 %
FLiNaBe	2200	158.15	0.48 %
NaFNaB	1506	263.08	6.56 %
FluZirK	1000	9.43	5.36 %
CloKMag	1155	140.41	4.42 %
Solar Salt	$1396.044 + 0.172\cdot T$	148.99	2.36 %
Hitec <sup>®</sup>	1560	473.23	2.45 %

even complete piping systems, reactors, or heat exchanger loops. Moreover, numerical simulations need verified physical properties as an input. This work intends to be a refined compendium of data, aiming to feed computer aided engineering designs. After a intensive review of the different reports and published data for eight different mixtures, several discrepancies have been found for some correlations, and a lack of data for certain salts. Deviations reveal that density is the best known property, but both viscosity as values for thermal properties, which in some cases are only based in theoretical models, show scattering. By this order, FLiBe, nitrates and NaFNaB mixtures have a global acceptable accuracy (less than 5 % of global average deviations), followed by CloKMag, FLiNaK and FLiNaBe (less than 8 %). Although FluZirK has been recently suggested as suitable salt for certain applications, the level of knowledge about transport and thermal behavior is very short nowadays. Hence, the need of new studies is legitimated in order to obtain higher accuracy, mainly for thermal properties. Discussion about this latter parameters have shown that standard techniques must be refined or even developed in some particular cases, to avoid undesirable conditions or mechanisms which can disturb measuring procedures (such as interaction of sensors or dissolution of container materials).

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