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Despite this extensive use of molten salts in thermal energy management, many basic and challenging issues about these materials are still unresolved, both at the experimental and theoretical level. One crucial issue is the temperature dependence of cP(T) in their liquid phase, especially concerning nitrate molten mixtures. Experimentally, different dependencies have been found, from increasing, to constant, to decreasing with T2,3,4,5,6,7. For this reason, round robin tests have been launched within the scientific and technological communities8.

In this rather complex experimental scenario, systematic theoretical results about thermostatic properties of molten salts and their mixtures are largely missing, while results for enthalpy H(T) and cP(T) of pure salts have been reported in ref.13.

As mentioned, empirical equations represent a key factor in establishing TES performances and have been crucial in the design of state-of-the-art CSP plants built recently14. Thus, revisiting them theoretically has a potentially high technological impact. Here, the validity of the mentioned cp relation will be investigated in detail and critically reconsidered.

Clearly, improvements in the performances of storage systems, connected to the energy production and reduction of electricity costs (e.g., of CSP plants), heavily rely on the maximum optimization of the thermodynamic properties of well known and new salt-based mixtures15,16,17,18,19.

In this work, we perform an extensive theoretical analysis, based on classical molecular dynamics (MD), of the temperature behavior of the thermostatic properties of NaNO3, KNO3 and their mixtures, with emphasis on the thermal behaviour of the specific heats cP(T) and cV(T), technologically relevant especially for the mixtures. The results are first compared with our new DSC experiments and with previous measurements. Then, they are interpreted by establishing a link between the solid-state approach to collective vibrational modes in liquids20,21,22,23, and the more standard gas-like approach24,25.

We will first investigate the thermostatic properties of pure potassium and sodium nitrates, in their solid and liquid regimes. Next, we will analyze the eutectic and "solar" mixtures with the ultimate goal to characterize the thermal behaviour of the specific heats in the liquid phases. All properties and methods to calculate them are described in the Methods Sections (MS).

Moreover, it is also the result of a MD procedure to locate TM, based on the temporal evolution of a two-phase system, as described in MS 1.4. The melting point is also reported in ref.13, where a different value (TM = 513 K ? 17 K) was obtained via a thermodynamic integration-based method.

Experimentally, solid KNO3 shows three polymorphic forms at P = 1 atm26: a stable form at 299 K, denoted a-KNO3; a stable phase generated by heating at T = 403 K, v-KNO3; and a third, different metastable phase g-KNO3 obtained by cooling down the system from high temperature, resulting from an alternative kinetic path.

As we aim to characterize the specific heats in various phases, we preliminary analyze the density r(T) and the enthalpy H(T) temperature behavior. The results are presented in Fig.2. Focusing first on the solid phases between 273 K and ?600 K, we find that by heating up the a-KNO3 phase from T = 273 K, the calculated density shows a strongly non-linear behavior in the range T = [273,400] K, Fig.2(a). This indicates the formation of a new phase, corresponding to v-KNO3. By cooling down the latter from T = 450 K, the density is once again non-linear, but the g-KNO3 phase is obtained at room temperature.

The overall agreement between experimental data and MD results in T = [400, 725] K is very good if compared to the accuracy found in the literature13, the difference being <8%. However, we note that the cP experimental data are in between the cP and cV theoretical results and show a slight tendency to decrease and oscillate with increasing T. This could be a consequence of experimental conditions closer to constant V than to constant P (sealed and small sample holder used in DSC experiments).

Finally, as shown in Fig.7, the T-behavior of the experimental isothermal compressibility kT(T) in the liquid regime is also well reproduced by our MD modeling. A similar self-consistency test as for KNO3, based on the use of three calculation procedures was performed for NaNO3. The test was successful, as shown by the coincidence of the black plot and the blue dot in Fig.7 and by the correct behavior of the Bhatia-Thornton structure factors SNN(k), tending to kT at the k -> 0 limit. Hence, all the considerations on accuracy and precision made for the KNO3 isothermal compressibility apply here too.

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