GENERAL INTRODUCTION
Physico-Chemical studies of molten salts and their mixtures have been a subject of extensive research for the last several years. Recently, much interest has been shown in investigating their industrial and technological applications.

Measurements of density, viscosity, surface tension, electrical conductance and ultrasonic velocity help in understanding the molecular interactions in molten salts as well as in their mixtures and also in correlating their behaviour with the corresponding thermodynamic properties. Since salts are expected to behave like liquids in their molten state, the theories of liquids are applied to study their thermodynamic behaviour. Considerable interest has also been shown in studying the thermodynamic behaviour of molten electrolytes in aqueous and non-aqueous media as well as in their corresponding mixtures.

Some of the hydrated salts and their aqueous solutions in higher concentrations are known to have greater tendency to form glasses. A knowledge of their solute-solvent interactions helps a glass technologist in deciding about the glass composition for achieving glasses with specific properties.

The excess properties like the excess volume, $V^E$, the excess free-energy, $G^E$, and the excess viscosity, $\eta^E$, obtainable from the temperature and concentration dependences of density and viscosity data, have been utilized adequately for explaining
the strength and the nature of intermolecular interactions in such systems\textsuperscript{11-14}. Several empirical relations\textsuperscript{15-19} have been used to represent the concentration dependence of viscosity of binary liquid mixtures. The observed positive deviations from the dependence of viscosity on mole fraction and the presence of maxima\textsuperscript{20,21} have been attributed to complex formation while the negative deviation has been ascribed to the presence of dispersive forces.

The viscosity of aqueous electrolytes has been studied since Arrhenius began his investigations of the properties of electrolytic solutions in 1880's. Three major reviews on the subject have appeared\textsuperscript{22-24} besides many empirical relations which have been proposed for explaining the behaviour of single and mixed electrolyte solutions. Einstein's\textsuperscript{22} relation is restricted to the dilute range of solute concentration and consequently neglects the solvent-solute interaction. Jones and Dole\textsuperscript{23} empirical equation accounts for both the solute-solute electrostatic interaction and the solvent-solute interaction.

According to Merker and Scott\textsuperscript{25} it is the difference between the microscopic and the macroscopic viscosities, depending upon the solvent-solute interaction, that determines the various values of Einstein's constant for different solute-solvent systems. Vand\textsuperscript{26} extended the Einstein's limiting theory of higher concentrations to such solutions.
To deal with the viscosity of concentrated solutions, Suryanarayana and Venkatesan\textsuperscript{24} empirically formulated an equation and demonstrated its validity for some electrolytes. Efforts have been made to explain the concentration dependence of viscosity in aqueous solutions of molten electrolytes. Although the concentration dependence of viscosity has been investigated for many aqueous solutions of molten electrolytes\textsuperscript{5,27} as well as molten electrolytes in a mixture of aqueous and non-aqueous media, the data in aqueous solutions of mixed electrolytes are insufficient to arrive at some definite conclusions in respect of the models employed for explaining their behaviour. This led to study the concentration dependence of viscosity in aqueous solutions of mixed molten electrolytes. The excess viscosities, $\eta^E$, of systems have also been evaluated. The positive sign of $\eta^E$ has been attributed\textsuperscript{20,28} to a very strong interaction between the unlike molecules while the magnitude of $\eta^E$ is considered a measure of the strength of interaction.

Nomoto\textsuperscript{29} and Bhimsen\textsubscript{30} et al. evaluated the ultrasonic velocity in binary liquid mixtures by using the Nomoto's relation. Van Dael and Vangeel\textsuperscript{31} have applied the ideal mixing relation for evaluating the ultrasonic velocities in liquids. Mishra\textsuperscript{32} and others\textsuperscript{33} studied the relative merits of both the relations for the computation of ultrasonic velocity in binary and ternary mixtures of liquids. Nomoto's as well as the ideal
mixing relations have been successfully applied to evaluate the ultrasonic velocity in mixtures of molten salts\(^{34-36}\). The free length theory (F.L.T) of Jacobson\(^{33}\) has been applied to evaluate the ultrasonic velocity in binary liquid mixtures\(^{37-45}\) as well as in molten salt mixtures\(^{36,46}\).

Schaafs\(^{47,48}\) collision factor theory has also been successfully employed to evaluate the ultrasonic velocity in binary liquid mixtures\(^{37,42-45}\) as well as in molten mixtures\(^{46}\).

The ultrasonic velocity data are employed to evaluate the thermodynamic properties including the compressibilities of molten salts\(^{34,35}\) and their binary\(^{46,49}\) and ternary mixtures\(^{36}\), and also of aqueous solutions of single\(^{4,46,50-52}\) and mixed molten electrolytes\(^{53}\).

Several workers\(^{54-63}\) have proposed a number of fluid equations of state based on the consideration of hard spheres. The equations of Lebowitz \textit{et al.}\(^{56}\) and Rao \textit{et al.}\(^{64,65}\) helped in evaluating the acoustical properties (velocity and absorption coefficients) of liquids. Mayer\(^{66}\) also applied this equation to investigate the molecular parameter relationship between the compressibility factor and the surface tension. Even though the Cornhan-Sterling equation\(^{61}\) has been found to be better suited for explaining such behaviours than the hard sphere models proposed by others\(^{67,68}\), the latter (hard sphere equation) is still being employed to obtain the equilibrium thermodynamic properties of pure liquids\(^{69}\), liquid mixtures\(^{70-72}\),
Surface tension, $\sigma$, is one of the various physical properties which has been investigated to understand the ionic interactions in binary molten salt mixtures. Many attempts have been made for the theoretical correlation of surface tension of liquids, molten salts and their binary mixtures with ultrasonic velocities. Guggenheim, on the basis of quasi-crystalline model derived equations for the ideal and regular solutions. In order to ascertain if certain molten salts are non-interacting or they result in complex formation due to their interaction, Heymann et al.\textsuperscript{75} employed the thermodynamic method of Guggenheim for this purpose. An improved model was developed by Hoar and Melford\textsuperscript{76}. Later on, Bertozzi and Sternheim\textsuperscript{77,78} showed that there are significant deviations from the earlier proposed equations when employed for the calculation of $\sigma$-values of alkali nitrates and binary halide systems and they introduced in 'Tobolsky Parameter' to account for the intermolecular interaction in their efforts to obtain good results.

On the basis of few existing phenomenological theories of liquid state, Lennard-Jones\textsuperscript{79} and Corner have calculated the $\sigma$-values by employing the free-volume theory while Reiss, Lebowitz and Frisch\textsuperscript{80} used the hypernated chain theory for this purpose while Eyring et al.\textsuperscript{81} employed the significant structure theory. These theories reproduce the experimental values of $\sigma$ reasonably well in the low density range unlike those obtained
in the higher density range. Ono\textsuperscript{82} has reviewed the methods available for the calculation of surface properties by employing such theories.

Nissen and Domelen\textsuperscript{83} have pointed out that the equations based on the regular solution theory assuming quasi-lattice model and random distribution of species, both in the bulk phase as well as in that of the surface phase, can be used to calculate the $\sigma$-values of binary molten mixtures.

On the whole, the results thus obtained using these models were found to be in good agreement with those of the experimental values while the deviations were attributed to non-coulombic interactions which invalidate the random mixing assumptions implicit in such theoretical considerations.

Addition of molten electrolytes to water is known to cause an increase in the surface tension, which corresponds to a desorption of salt from the surface region\textsuperscript{84}. As a consequence of this considerable interest has been shown in the evaluation of surface tension in aqueous solutions of molten electrolytes\textsuperscript{84}, but very few attempts have been made to evaluate the surface tension of aqueous solutions of mixed molten electrolytes.

Finally, the Flory's statistical theory\textsuperscript{85,86} has been employed to evaluate the reduced and the characteristic thermodynamic parameters which, in turn, can be employed for evaluating the interaction parameter, the viscosity, the surface tension,
the ultrasonic velocity and the excess thermodynamic properties.

Consequently, with a view to understanding the behaviour of aqueous solutions of mixed molten electrolytes the density, the viscosity and the ultrasonic velocities of \([x \text{NaSCN} + (1-x)\text{KSCN}] + R\text{H}_2\text{O} \ (R = 4 \text{ to } 7)\) are measured as functions of mole fraction of NaSCN, ionic concentration, \(R = (\text{water/salt})\) ratio and temperature. In addition, some of the thermodynamic functions, the relevant parameters, the surface tension and the viscosities have also been evaluated for the said purpose.