Acoustical and Compressibility Parameters of Glycylglycine−FeCl$_3$ in Aqueous Ethanol Mixture at $T = (293.15, 303.15, \text{and } 313.15)$ K

M. S. Santosh and D. Krishna Bhat*

Physical Chemistry Division, Department of Chemistry, National Institute of Technology Karnataka, Surathkal, Srinivasanagar Post, Mangalore-575025, India

Ultrasonic velocity measurement is a reliable procedure that allows quick and easy determination of solvent concentrations in mixtures. This paper presents data of ultrasonic velocities and isentropic compressibilities of (glycylglcyine−FeCl$_3$) in aqueous ethanol mixture at $T = (293.15, 303.15, \text{and } 313.15)$ K. Various acoustical parameters were calculated, because of their importance in the study of specific molecular interactions. A less-compressible phase of the fluid and a closer packing of molecules is observed with changes in the intermolecular free length. Quantitative investigation suggests that the mixing of ethanol with an aqueous glycylglycine−FeCl$_3$ solution causes the rupture of hydrogen bonds, with increasing $\delta\kappa_S$ and L_f values (where the former represents the change in isentropic compressibility and the latter represents the intermolecular free length). Theoretically computed values of isentropic compressibility in the studied mixture using different models indicate the superiority of the very complex procedure.

Introduction

In recent years, the measurement of ultrasonic velocity has been successfully employed in understanding the nature of molecular interactions in pure liquids and liquid mixtures. Ultrasonic velocity measurements are highly sensitive to molecular interactions and can be used to provide qualitative information about the physical nature and strength of molecular interaction in liquid mixtures.[1−3] The ultrasonic velocity of a liquid is fundamentally related to the binding forces between atoms or molecules, and it has been successfully employed in understanding the nature of molecular interactions in pure liquids, as well as binary and ternary mixtures.[4,5] Variations in the ultrasonic velocity and related parameters have shed much light upon the structural changes associated with liquid mixtures of weakly[6] or strongly interacting components.[7] The study of molecular associations in binary mixtures that have alcohol as one component is of particular interest, since alcohols are strongly self-associated liquids with a three-dimensional network of hydrogen bonds[8] and can be associated with any other group that has some degree of polar attraction.[9] A brief survey of the literature has shown that few attempts have been made to obtain ultrasonic velocity data for binary liquid mixtures.[10−14] However, acoustical studies of dipeptide−transition-metal salt in aqueous ethanol mixtures are still scant. Our purpose is to discuss the dependence of isentropic compressibility on mixing, and that of other acoustic parameters on the composition and functionality, to provide a better understanding of the factors that contribute to the special behavior in incorporating such macromolecules into mixtures, where the solvent has a shorter chain and different molecular nature. Because of their importance in industrial design, different procedures[14,15] and theories[16−18] for the isentropic compressibilities were applied, and the obtained results are analyzed and commented upon.

In continuation of our previous studies,[19−23] we have attempted to explain the physicochemical behavior of glycylglycine−FeCl$_3$ in aqueous ethanol mixtures, to explore the strength and nature of interactions between the components by deriving various thermodynamic parameters from ultrasonic velocity (u) and density (ρ) data. Numerous parameters such as intermolecular free length (L_f), van der Waals' constant (b), molecular radius (r), geometrical volume (B), molar surface area (Y), molar sound velocity (R), collision factor (S), and specific acoustic impedance (Z) were computed.

Experimental Section

Materials. Glycylglycine and iron(III) chloride hexahydrate (with a purity of 0.99 (mass fraction)) were used in our studies and obtained from Sigma−Aldrich Germany, Ltd. Commercially available glycylglycine of the highest purity was used without further purification. $FeCl_3 \cdot 6H_2O$ was used after drying for 72 h in a vacuum desiccator at room temperature. Deionized, doubly distilled degassed water with a specific conductance of $<1.29 \times 10^{-6}\ \Omega^{-1}\ cm^{-1}$ was used for the preparation of all solutions. Ethanol (analytical reagent (AR) grade, 99.9% purity) used after redistillation to ensure maximum purity was purchased from Changshu Yanguan Chemicals, China. The solutions were prepared on a weight basis, using a Mettler balance that had a precision of ±0.01 mg. Care was taken to avoid evaporation and contamination during mixing. The water temperature was controlled within ±0.01 K, using a thermostat. In our studies, glycylglycine and FeCl$_3$ were kept constant at 0.020 mol kg^{-1} and 0.25 mol kg^{-1}, respectively, and the composition of ethanol was varied in terms of mole fraction (x_1). The estimated uncertainty for the mole fraction of ethanol was determined to be $<1 \times 10^{-4}$. To prevent formation of air bubbles, all solutions were preheated in sealed Eppendorf tubes to 5 °C above the measurement temperature, before filling the ultrasonic and densimetric cells.

Methods. The ultrasonic velocity of pure components and their mixtures were measured using a variable-path fixed frequency interferometer (Model-83, Mittal Enterprises, New Delhi, India). It consists of a high-frequency generator and a measuring cell. The ultrasonic velocity measurements were taken at a fixed frequency of 2 MHz. The capacity of the measurement cell was 7 mL. The calibration of ultrasonic interferometer was done by measuring the velocity in AR-grade benzene and carbon tetrachloride. The maximum estimated error in ultrasonic velocity measurements was

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* To whom correspondence should be addressed. Tel.: +91-9481271262. Fax: 0824-2474033. E-mail: denthajekb@gmail.com.

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Table 1. Ultrasonic Velocity, Density, Isentropic Compressibility, and the Change in Isentropic Compressibility for Glycylglycine + $FeCl_3$ in an Aqueous Ethanol Mixture at T = (293.15, 303.15, and 313.15) K, against the Mole Fraction of Ethanol

mole fraction of ethanol, x_1	ultrasonic velocity, u (m/s)	density, ρ ($\times 10^{-3}$ kg/m^3)	isentropic compressibility, κ_S (TPa^{-1})	change in isentropic compressibility, $\delta\kappa_S$ (TPa^{-1})
		T = 293.15 K		
0.0000	1468.91	1.0012	462.90	−220.79
0.0857	1510.82	0.9843	445.08	−191.93
0.1715	1569.65	0.9715	417.78	−163.54
0.2572	1604.79	0.9587	405.02	−134.12
0.3430	1650.53	0.9456	388.19	−106.26
0.4288	1691.48	0.9293	376.10	−77.47
0.5145	1744.26	0.9151	359.17	−48.65
0.6003	1788.12	0.9074	344.67	−20.42
		T = 303.15 K		
0.0000	1531.87	0.9914	429.84	−205.29
0.0857	1575.42	0.9743	413.53	−178.77
0.1715	1629.07	0.9597	392.63	−151.58
0.2572	1674.22	0.9463	377.00	−124.47
0.3430	1708.22	0.9337	367.03	−95.69
0.4288	1754.40	0.9199	353.18	−69.38
0.5145	1800.68	0.9064	340.25	−41.74
0.6003	1845.53	0.8982	326.87	−15.62
		T = 313.15 K		
0.0000	1622.85	0.9741	389.79	−184.93
0.0857	1664.62	0.9586	376.47	−159.35
0.1715	1705.47	0.9458	363.50	−133.68
0.2572	1748.36	0.9325	350.82	−107.82
0.3430	1782.27	0.9200	342.18	−82.55
0.4288	1835.59	0.9064	327.43	−57.79
0.5145	1879.14	0.8972	315.64	−32.41
0.6003	1913.93	0.8910	306.38	−8.18

determined to be ±0.08%. The temperature was controlled by circulating water around the liquid cell from a thermostatically controlled adequately stirred water bath. Densities were measured using a digital densitometer (Model Density 30PX, Mettler−Toledo) with an uncertainty of ±0.001 g/cm^3. The densitometer was calibrated using double-distilled water. The sample and reference resonator cells with minimum volumes of 0.5 cm^3 were thermostatted with an accuracy of ±0.01 K, and a previously described differential technique was employed for all measurements.[24] The physical parameters for aqueous ethanol solutions of glycylglycine−$FeCl_3$ were measured at temperatures of 293.15, 303.15, and 313.15 K. The measured density values and their uncertainties agree well with the literature values.[25] Each measurement was repeated three times, and the reported values are an average of all three trials.

Results

The experimental data regarding the ultrasonic velocity, density, isentropic compressibility, and change in isentropic compressibility (the latter of which is calculated by means of eq 1) are presented in Table 1.

$$\delta Q = Q_{\text{mix}} - \sum_{i=1}^{N} x_i Q_i \tag{1}$$

In this equation, δQ represents the variation of magnitude Q in the value of the isentropic compressibility (κ_S), which is calculated by the Laplace equation from density and ultrasonic velocity; Q_i is the magnitude of pure solvent, x_i is the mole fraction, and N is the number of components in the mixture. The derived magnitudes of the binary mixtures were fitted using the Redlich−Kister expression:[26]

$$\delta Q = x_i x_j \left[\sum_{p=0}^{S} B_p (x_i - x_j)^p\right] \tag{2}$$

where B_p is the fitting parameter obtained by the least-squares method of the fitting algorithm,[27] and S is the degree of the polynomial expansion, which was optimized by means of the Bevington test.[28] The root-mean-square deviations were computed using eq 3, where z is the value of the property, and n_{DAT} is the number of experimental data points.

$$\sigma = \left[\frac{\sum_{i=1}^{n_{\text{DAT}}} (Z_{\text{exp}} - Z_{\text{pred}})^2}{n_{\text{DAT}}}\right]^{1/2} \tag{3}$$

In Figure 1, the changes in isentropic compressibility are plotted against the mole fraction of ethanol. The fitting parameters for the curves and the σ deviation values are listed in Table 2.

To explore the strength and nature of interaction between the components, various acoustical parameters are derived from the ultrasonic velocity and density data. The parameters intermolecular free length (L_f), van der Waals' constant (b), molecular radius (r), geometrical volume (B), molar surface area (Y), molar sound velocity (R), collision factor (S), and specific acoustic impedance (Z) are derived from the following set of equations:

$$L_f\,(\text{nm}) = \left(\frac{K}{u\rho^{1/2}}\right) \tag{4}$$

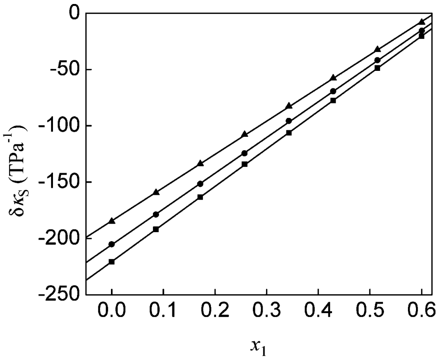

Figure 1. Changes in isentropic compressibility ($\delta\kappa_S$) for (glycylglycine + $FeCl_3$) (1) in an aqueous ethanol (2) mixture against the mole fraction of ethanol (x_1) at temperatures of (■) 293.15 K, (●) 303.15 K, and (▲) 313.15 K.

Table 2. Redlich−Kister Parameters (TPa^{-1}) for Changes in Isentropic Compressibility of Glycylglycine + $FeCl_3$ in an Aqueous Ethanol Mixture at T = (293.15, 303.15, and 313.15) K

	Value		
parameter	T = 293.15 K	T = 303.15 K	T = 313.15 K
B_0	−976.32	−926.59	−892.38
B_1	824.91	765.47	673.39
B_2	−801.39	−745.02	−663.84
B_3	724.16	698.43	636.91
B_4	−412.52	−361.67	−315.32
σ	0.34	0.33	0.32

$$b\ (\mathrm{cm^3/mol}) = \left(\frac{M}{\rho}\right) - \left(\frac{\mathrm{R}T}{\rho u^2}\right)\left[\left(1 + \frac{Mu^2}{3\mathrm{R}T}\right)^{1/2} - 1\right] \qquad (5)$$

$$r\ (\times 10^{10}\ \mathrm{m}) = \left(\frac{3b}{16\pi N}\right)^{1/3} \qquad (6)$$

$$B\ (\mathrm{m^3/mol}) = \left(\frac{4}{3}\right)\pi r^3 N \qquad (7)$$

$$Y\ (\times 10^{12}\ \mathrm{m^2}) = (36\pi NB^2)^{1/3} \qquad (8)$$

$$R\ (\mathrm{cm^3\ mol^{-1}\ m\ s^{-1}}) = \frac{Mu^{1/3}}{\rho} \qquad (9)$$

$$S = \frac{uV}{Bu_\infty} \qquad (10)$$

$$Z\ (\times 10^{-3}\ \mathrm{kg\ m^{-2}\ s^{-1}}) = u\rho \qquad (11)$$

where L_f is the free length of ideal mixture, K the Jacobson constant (which is temperature dependent; $K = (93.875 + 0.375\ T) \times 10^{-8}$),[16] M the molar mass of the solute, and N the number of components in the mixture. V is the molecular volume, u_∞ is taken to be 1600 m/s, and R and π are common universal constants. The above parameters are listed in Table 3.

The compressibility solvation numbers were calculated using the following equation:[29]

$$n_S = \left(\frac{n_{\mathrm{solvent}}}{n_{\mathrm{solute}}}\right)\left(1 - \frac{\kappa_S}{\kappa_S^0}\right) \qquad (12)$$

where n_{solvent} and n_{solute} are the mole number of solvent and the mole number of solute, respectively. The variation of compressibility solvation number, as a function of mole fraction of ethanol (x_1), is shown in Figure 2.

Estimation Models. The estimation of different acoustical properties of binary or multicomponent mixtures recently have been the subject of study, with different empirical or semiempirical models being applied to optimize industrial equipment and understand the mixing process. Experimental data for isentropic compressibility of the studied mixture were compared with values determined by different procedures. The models of Danusso and Nomoto, which are described by eqs 13 and 14, respectively,[15]

$$\kappa_S\ (\mathrm{TPa^{-1}}) = \left(\frac{1}{M\rho}\right)\left(\sum_{i=1}^{N}\frac{n_i M_i}{\rho_i^2 u_i^2}\right) \qquad (13)$$

$$\kappa_S\ (\mathrm{TPa^{-1}}) = \left(\frac{1}{\rho}\right)\left(\frac{\sum_i^N n_i R_i}{\sum_i^N n_i u_i}\right)^{-6} \qquad (14)$$

(where $R = u^{1/3}\sum_i^N n_i V_i$) and the Collision Factor Theory (CFT) and the Free Length Theory (FLT),[16−18] as described by eqs 15 and 16, respectively, for isentropic compressibilities were applied.

$$\kappa_S\ (\mathrm{TPa^{-1}}) = \left(\frac{1}{\rho^3}\right)\left(\frac{M}{u_\infty \sum_i^N x_i S_s \sum_i^N x_i B_s}\right)^2 \qquad (15)$$

$$\kappa_S\ (\mathrm{TPa^{-1}}) = \frac{L_f^2}{K^2} \qquad (16)$$

The Collision Factor Theory (CFT) is dependent on the collision factors among molecules, as a function of temperature in the pure solvent or mixture. The pertinent relationships in these calculations and the theoretical basis were described by Nutschkuhnkies[17] and Schaaffs.[18] The collision factor (S) and characteristic molecular volume (B) of the mixture used in the CFT calculations were estimated using the experimental ultrasonic velocities, which have been given in this paper, and the molar volumes of Santosh et al.[20] The Free Length Theory (FLT) estimates the isentropic compressibility of a mixture, using the free displacement of molecules, as a function of temperature. In the past few years, different authors have compared the relative merits of the existent theories with FLT, resulting in greater deviations of computed isentropic compressibilities from the experimental values. The deviations of each procedure for the studied mixture are given in Table 4, using the parameters from Table 3 and literature data.[29]

Discussion

The change in isentropic compressibility, as a function of mole fraction of ethanol for the studied binary mixture, is shown in Figure 1. It is observed that the change in isentropic compressibility is negative for all compositions and temperatures. This indicates that the mixture is more compressible than the corresponding ideal mixture. The molecules present in the solution interact with water readily and show strong solute−solvent interactions. When ethanol is introduced into the solution, the ethanol molecules weaken the interactions between water, glycylglycine, and $FeCl_3$. This leads to greater compressibility within the solution, and the solute molecules undergo a closer packing to retain the possible interaction with water. This causes a decrease in the intermolecular free length (L_f) and enhances collision within the interacting molecules. The decrease in L_f is inversely proportional to the ultrasonic velocity (u) and may

Table 3. Acoustic Parameters for Glycylglycine + $FeCl_3$ in an Aqueous Ethanol Mixture, Relative to the Mole Fraction of Ethanol

mole fraction of ethanol, x_1	free length of ideal mixture, L_f (nm)	van der Waals constant, b (cm^3/mol)	molecular radius, r ($\times 10^{10}$ m)	geometrical volume, B (m^3/mol)	molar surface area, Y ($\times 10^{-12}$ m^2)	molar sound velocity, R (cm^3 mol^{-1} m s^{-1})	collison factor, S	specific acoustic impedance, Z ($\times 10^{-3}$ kg m^{-2} s^{-1})
				$T = 293.15$ K				
0.0000	4.297	48.382	2.987	12.623	1.327	564.237	4.002	1470.672
0.0857	4.282	51.426	3.112	13.476	1.456	571.748	4.054	1487.100
0.1715	4.268	54.827	3.354	14.725	1.526	579.269	4.086	1524.914
0.2572	4.254	57.921	3.523	15.330	1.632	586.710	4.123	1538.512
0.3430	4.240	60.462	3.792	16.418	1.747	594.271	4.165	1560.741
0.4288	4.226	63.729	3.920	17.633	1.849	601.722	4.198	1571.892
0.5145	4.211	66.385	4.135	18.579	1.933	609.283	4.213	1596.172
0.6003	4.197	69.711	4.397	19.217	2.012	616.749	4.274	1622.540
				$T = 303.15$ K				
0.0000	4.151	62.142	3.462	14.652	1.545	630.274	3.723	1518.695
0.0857	4.137	64.237	3.698	15.233	1.623	636.505	3.771	1534.931
0.1715	4.123	66.157	3.813	16.317	1.736	642.826	3.800	1563.418
0.2572	4.109	69.056	4.026	17.284	1.867	649.157	3.845	1584.314
0.3430	4.095	71.484	4.223	18.643	1.924	655.438	3.884	1594.965
0.4288	4.081	74.265	4.441	19.926	2.056	661.779	3.932	1613.872
0.5145	4.067	76.912	4.638	20.427	2.172	668.060	3.967	1632.136
0.6003	4.050	79.049	4.875	21.689	2.225	673.892	3.999	1657.655
				$T = 313.15$ K				
0.0000	4.097	73.482	3.921	16.384	1.764	698.203	3.380	1580.818
0.0857	4.076	75.627	4.173	17.478	1.825	706.560	3.422	1595.704
0.1715	4.055	77.934	4.385	18.921	1.973	714.917	3.463	1613.033
0.2572	4.038	79.685	4.519	19.265	2.069	723.274	3.491	1639.688
0.3430	4.019	81.157	4.762	20.458	2.187	731.631	3.536	1663.778
0.4288	3.994	83.275	4.924	21.627	2.275	739.988	3.570	1685.964
0.5145	3.977	85.883	5.187	22.912	2.364	748.345	3.617	1705.311
0.6003	3.952	88.366	5.372	23.847	2.489	756.831	3.654	1445.959

Table 4. Root-Mean-Square Deviations for Predicted Isentropic Compressibility from Experimental Data for Glycylglycine + $FeCl_3$ in an Aqueous Ethanol Mixture at T = (293.15, 303.15, and 313.15) K

	Root-Mean-Square Deviation			
model/theory	equation	T = 293.15 K	T = 303.15 K	T = 313.15 K
Danusso	eq 13	0.49	0.56	0.69
Nomoto	eq 14	0.32	0.36	0.40
Collision Factor Theory, CFT	eq 15	0.37	0.34	0.33
Free Length Theory, FLT	eq 16	3.11	3.05	2.98

be attributed to the overall increase of cohesion that is caused by glycylglycine−$FeCl_3$ and glycylglycine−water interactions in the solution. Because of these interactions, the ultrasonic velocity increases and the compressibility of the solution decreases. Such behavior lends support to the strong solute−solvent and solute−solute intermolecular/interionic interactions in the above system.[30] From these considerations, it is clear that there is a strong association in the system studied. The variation in L_f indicates that structural rearrangement in the liquid mixture has taken place, toward a less-compressible phase of the fluid, and a closer packing of molecules is observed.[31] A qualitative investigation of the system under study indicates that (a) the molecules are associated through hydrogen bonding and (b) the mixing of ethanol in an aqueous glycylglycine−$FeCl_3$ solution will induce rupture of the hydrogen bonds, with a subsequent increase in the $\delta\kappa_S$ and L_f values. However, because of the simultaneous formation of hydrogen bonds between OH groups of unlike molecules, there is a compensating effect, resulting in an overall increase in $\delta\kappa_S$ and L_f values, as evident from Figure 1 and Table 3. These values indicate that the sound wave covers a shorter distance, because of a decrease in L_f, as a result of packing. It has been suggested[32,33] that the change in isentropic compressibility ($\delta\kappa_S$) and intermolecular free length (L_f) decrease with increasing strength of interaction between the component molecules. As expected, the values of specific acoustic impedance (Z) again support our view that the interactions between unlike molecules are quite obvious.[34] Also, it is observed from Table 3 that all the acoustic parameters except L_f increase with the mole fraction of ethanol (x_1). The decrease in L_f and the increase in Z with increasing concentration indicate that there is significant interaction between the solute and solvent molecules, and, as a result, the structural arrangement is considerably affected. This can be explained on the basis of hydrophobic interaction between solute and solvent molecules, which increases with the intermolecular distance, leaving relatively wider gaps between the molecules, and thus becomes the main cause of impediment to the propagation of ultrasound waves. From these measurements, two factors must be derived: first, the entire mixture shows lower values of compressibility than ideality, and second, a consequence of the molecular characteristics of the solvent and similar dispersive interaction at the aliphatic end, which leads to the creation of a more structured packing of molecules. The addition of ethanol to an aqueous solution that contains glycylglycine and $FeCl_3$ has a tendency to enhance dispersive interaction and the creation of better-packed clusters with a consequent increase in ultrasonic velocity and a decrease in density and isentropic compressibility.

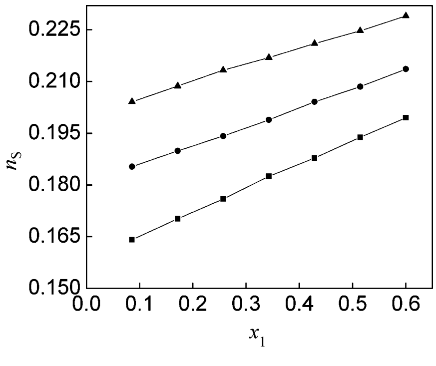

Figure 2. Variation of compressibility solvation number for (glycylglycine + $FeCl_3$) (1) in an aqueous ethanol (2) mixture, relative to the mole fraction of ethanol (x_1) at temperatures of (■) 293.15 K, (●) 303.15 K, and (▲) 313.15 K.

The compressibility solvation number is shown in Figure 2. From Figure 2, it can be concluded that (a) coordination is mainly dependent on volume of the solvent, and (b) there is an increase in compressibility solvation number with increase in temperature (this evolution was truncated in alcohol mixtures by phase separation). Similar results are computed for different temperatures by each procedure, which can be observed in Table 4. The accuracy achieved was examined using the Nomoto model and CFT, and an overestimation of experimental data was obtained in the studied system at higher solvent compositions. The results of the comparison of predicted values with experimental data are summarized in Table 4, in which the predicted isentropic compressibilities are shown. The model proposed by the Danusso model predicts values with deviations of <0.49 for the mixture. Since the Danusso model assumes additivity of the molar volume, this equation more accurately predicts values for mixtures that show ideal behavior. From the deviation in computed data, we arrive at the conclusion that the application of the CFT produces close agreement with the experimental data reported in this paper, showing this procedure as an accurate tool for isentropic compressibility data for nonideal systems.

Conclusion

The ultrasonic velocity (u) and density (ρ) values for glycylglycine−$FeCl_3$ in aqueous ethanol mixtures have been reported. The experimental values have been used to compute various acoustical parameters and the variations have been interpreted in terms of solute−solvent interactions. The simultaneous formation of hydrogen bonds between OH groups of unlike molecules causes a compensating effect, resulting in an overall increase in the change in isentropic compressibility ($\delta\kappa_S$) and in the intermolecular free length (L_f). An increase in specific acoustic impedance (Z) indicates that there is significant interaction between the solute and solvent molecules and, as a result, the structural arrangement is considerably affected. Also, relatively wider gaps between the molecules become the main cause of impediment to the propagation of ultrasound waves. Various models and theories have been used to correlate experimental values with predicted values of isentropic compressibilities and among them, the Collision Factor Theory (CFT) produced close agreement with the experimental data.

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