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# Volumetric, Acoustic and Viscometric Studies on the Behaviour of Homoeopathic Formulations of Ammonium Causticum at Different Temperatures



## Anil Kumar Nain<sup>1</sup>\*, Neha Chaudhary<sup>1</sup>, Anil Khurana<sup>2</sup>, Raj Kumar Manchanda<sup>2</sup> and Debadatta Nayak<sup>2</sup>

<sup>1</sup>Department of Chemistry, Dyal Singh College, University of Delhi, New Delhi, India

<sup>2</sup>Central Council for Research in Homeopathy, Ministry of AYUSH, New Delhi, India

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Corresponding author: Anil Kumar Nain, Department of Chemistry, Dyal Singh College, University of Delhi, New Delhi, India

#### Abstract

The densities  $\rho$ , ultrasonic speeds, u, and viscosities  $\eta$  of pure ethanol control and 32 formulations of ammonium causticum of potencies ranging from 1C to 200C (with intervals of 2C up to 30C and then with intervals of 10 C up to 200 C) have been measured at 293.15, 298.15, 303.15, 308.15, 313.15 and 318.15 K and atmospheric pressure. Using these experimental data, the isentropic compressibilities  $\kappa_s$ , intermolecular free length  $L_r$ , acoustic impedance Z, deviations in isentropic compressibility  $\Delta \kappa_s$ , deviations in intermolecular free length  $\Delta L_r$ , deviations in acoustic impedance  $\Delta Z$ , and deviations in viscosity  $\Delta \eta$ , have been calculated. The variations of these parameters with potency show anomalous behaviour at certain potencies. The results have been qualitatively discussed in terms of interactions of these ammonium causticum homoeopathic formulations. The results show that some potencies of ammonium causticum exhibit anomalous trends in investigated physicochemical properties and may show different behaviour in terms of efficacy when used in practice.

Keywords: Density; Ultrasonic Speed; Viscosity; Homoeopathic Medicines; Ammonium Causticum; Extremely Diluted Solutions

#### Introduction

The homoeopathic medicines are obtained through the combination of two processes, viz., a dilution of 1:100 followed by succession and these "extremely diluted solutions" show anomalous behaviour in medicinal efficacy. The efficacy of homoeopathic medicines is well supported by research evidence; however, there are controversies regarding improbability in biological activity of these medicines in which the source drug is diluted beyond Avagadro's limit, i.e., the highly diluted medicine formulation may be alike to the solvent. There have been a few research studies relating to reconnoiter the presence of drug in extremely diluted formulations [1-5] and its mechanism of action, but the question still exists unanswered. The physicochemical properties and derived parameters of electrolytes, amino acids, carbohydrates, drugs, etc. in aqueous and mixed-aqueous

solutions of have been helpful in characterizing the prevailing interactions, which are subsequently useful in understanding of solute-solvation/hydration behaviour of solute and preferential solvation of solute by the solvent [6-13].

The homoeopathic formulations are dilute solutions, therefore, their physical properties, like density, ultrasonic speed, viscosity, refractive index, etc. can be measured with varying potency and temperature. The physicochemical parameters derived from these experimental data provide valuable information regarding physicochemical behaviour and mechanism of drug action of these homoeopathic medicines. Recently, there have been few physicochemical studies on extremely diluted solutions of inorganic salts [14-17] and homoeopathic medicines [3,18-22] by using physicochemical methods. These studies provided interesting and convincing information on the behaviour of these extremely diluted solutions. To the best of our information, very few physicochemical studies on homoeopathic medicines using volumetric, acoustic and viscometric have been reported in the literature [3,18, 23, 24].

In continuation to earlier research on the physicochemical behaviour of extremely diluted homoeopathic formulations [25,26], here we report the results of our study on the physicochemical behaviour of homoeopathic formulations of ammonium causticum. Ammonium causticum is a homoeopathic dilution made from ammonia water. Ammonium causticum is used in the treatment of children with primary enuresis (bed-wetting since infancy). It is a powerful cardiac stimulant. It can be used as such in syncope, thrombosis, hemorrhage, snakebites, chloroform narcosis, and may be given by inhalation. It is also used for treating hearing loss, swelling and skin disorders. These considerations led us to undertake the present study on the physicochemical behaviour of extremely diluted homoeopathic formulations of ammonium causticum.

In the present study, the densities,  $\rho$ , ultrasonic speeds, u, and viscosities,  $\eta$ , of pure ethanol control (91% ethanol in water) and 33 formulations of ammonium causticum with potencies from 1C to 200C (with intervals of 2C till 30C, and then with intervals of 10C till 200C) at 293.15, 298.15, 303.15, 308.15, 313.15 and 318.15 K and atmospheric pressure. Using these experimental data, the

isentropic compressibilities,  $\kappa_{\rm s}$ , intermolecular free length,  $L_{\rm f}$ , acoustic impedance, Z, deviations in isentropic compressibility,  $\Delta\kappa_{\rm s}$ , deviations in intermolecular free length,  $\Delta L_{\rm f}$ , deviations in acoustic impedance,  $\Delta Z$  and deviations in viscosity,  $\Delta\eta$ , have been calculated. The results have been discussed qualitatively in terms of interactions/physicochemical behaviour of ammonium

causticum in these homoeopathic formulations.

#### **Experimental**

Ethanol control and homoeopathic formulations of various potencies of ammonium causticum used in the study were procured from Dr. Wilmer Schwabe India Pvt. Limited, India, which were prepared in accordance with Homoeopathic Pharmacopoeia of India [27]. The densities and ultrasonic speeds of the samples were measured by using high precision digital vibrating tube Density and Sound Analyzer (DSA 5000M, Anton Paar, Austria). The principle used in density measurement is based upon oscillating U-tube principle while the speed of sound is measured using a propagation time technique. This instrument is equipped with both density and ultrasonic cells, with reproducibility of  $\pm 1 \times 10^{-3}$  kg·m<sup>-3</sup> and  $\pm 1 \times 10^{-2}$  m·s<sup>-1</sup> for density and ultrasonic speed, respectively. The temperature for both cells was kept constant by using built in peltier thermostat within  $\pm 0.01$  K. The equipment

was calibrated with triply distilled degassed water and with dry air at atmospheric pressure [9,10]. The operating working frequency used for ultrasonic speed measurements is 3 MHz. The standard uncertainties related to the measurements of density, ultrasonic speed and temperature were found within  $\pm 0.05$  kg·m<sup>-3</sup>,  $\pm 0.5$  m·s<sup>-1</sup> and  $\pm 0.01$  K, respectively.

The viscosity measurements were done by using microviscometer (Lovis 2000M, Anton Paar, Austria) at temperatures, (293.15 -318.15) K, and atmospheric pressure p = 101 kPa. The temperature was controlled to  $\pm 0.02$  K by an automatic build in Peltier technique. The rolling ball principle was used in the measurement of viscosity, having a calibrated glass capillary with a steel ball as supplied by manufacturer. The calibration of capillary was accomplished by using viscosity standard fluids. The standard uncertainties for viscosity measurements and temperature were estimated to be within  $\pm 0.5\%$  and  $\pm 0.02$  K.

#### Results

The values of densities,  $\rho$ , ultrasonic speeds, u, and viscosities,  $\eta$  of homoeopathic formulations of ammonium causticum as function of potency (in centesimal) at different temperatures are listed in Tables 1-3 and are presented graphically in Figures

1-3 respectively. The values of the isentropic compressibility,  $\kappa_{s}$ 

, intermolecular free length,  $L_{\rm f}~$  , acoustic impedance, Z have been calculated by using the following relations

$$K_s = 1/u^2 \rho \tag{1}$$

$$L_{f} = K' / u \rho^{1/2}$$
 (2)

$$Z = u.\rho \tag{3}$$

where K' is temperature dependent constant [= (93.875 + 0.375T)  $\times 10^{-8}$ ]; T is the absolute temperature, The values of,  $\kappa_s$ ,  $L_f$  and Z are given in Tables 4-6.

The deviations in,  $\kappa_s$ ,  $L_f$ , Z and  $\eta$  due to addition of ammonium causticum with dilution and succussion are represented by deviations of these properties from ethanol control properties. The deviations in isentropic compressibility,  $\Delta \kappa_s$ , deviations in intermolecular free length,  $\Delta L_f$ , deviations in acoustic impedance,  $\Delta Z$  and deviations in viscosity,  $\Delta \eta$  have been calculated by using the following standard relations

$$\Delta \kappa_{\rm s} = \kappa_{\rm s} - \kappa_{\rm s}^{\rm o} \tag{4}$$

$$\Delta L_{\rm f} = L_{\rm f} - L_{\rm f}^{\rm o} \tag{5}$$

$\Delta Z = Z - Z^{\rm o}$	(6)	
$\Delta \eta = \eta - \eta^{\rm o}$	(7)	

ethanol control (91% ethanol in water). The variations of,  $\Delta\kappa_{\rm s}$  ,

 $\Delta L_{\rm f}$  ,  $\Delta Z$  ,and  $\Delta\eta~$  with potency, C of ammonium causticum and temperature are presented graphically in Figure 4-7, respectively.

where the superscript '' represents the values for pure

Table 1: The densities, p/ (kg m <sup>-3</sup> ) of ethanol control (0 potency, 91% ethanol in water) and 33 formulations of ammonium causticum in ethanol
control, as function of potency, C of ammonium causticum (in centesimal) at the temperatures (293.15-318.15) K and atmospheric pressure.

Potency	T/K					
(C)	293.15	298.15	303.15	308.15	313.15	318.15
1	869.103	864.829	860.516	856.132	851.688	847.18
2	827.977	823.621	819.215	814.756	810.237	805.657
4	830.877	826.148	821.829	817.463	812.845	808.369
6	827.961	823.785	819.329	814.97	810.419	805.922
8	827.82	823.468	819.064	814.607	810.089	805.511
10	827.975	823.618	819.21	814.749	810.227	805.646
12	827.784	823.43	819.026	814.566	810.049	805.47
14	827.772	823.417	819.111	814.652	810.135	805.46
16	827.808	823.46	819.056	814.597	810.079	805.498
18	828.033	823.68	819.278	814.823	810.303	805.724
20	828.049	823.695	819.293	814.839	810.322	805.745
22	828.123	823.679	819.475	814.919	810.306	805.725
24	828.1	823.745	819.342	814.887	810.369	805.792
26	828.096	823.744	819.342	814.886	810.37	805.794
28	827.969	823.613	819.211	814.755	810.238	805.66
30	828.632	823.947	819.763	815.14	810.694	806.16
40	831.907	827.563	823.261	818.604	814.086	809.504
50	831.784	827.396	823.198	818.543	814.025	809.48
60	831.679	827.331	823.131	818.576	814.058	809.378
70	831.591	827.34	823.042	818.485	813.965	809.349
80	831.471	827.232	822.92	818.464	813.846	809.308
90	831.347	827.098	822.797	818.39	813.753	809.141
100	831.133	826.982	822.683	818.226	813.61	809.003
110	831.016	826.825	822.563	818.111	813.512	808.915
120	830.88	826.671	822.402	817.942	813.319	808.844
130	830.663	826.452	822.241	817.8	813.202	808.68
140	830.458	826.21	822.008	817.551	812.933	808.452
150	830.249	825.997	821.807	817.338	812.722	808.201
160	830.112	825.808	821.427	817	812.455	807.904
170	829.821	825.536	821.134	816.678	812.16	807.611
180	829.527	825.172	820.78	816.413	811.94	807.316
190	829.152	824.898	820.495	816.041	811.523	806.943
200	828.878	824.529	820.128	815.673	811.159	806.579

Potency	Т/К							
(C)	293.15	298.15	303.15	308.15	313.15	318.15		
0	1165.93	1149.76	1133.54	1117.33	1101.38	1084.91		
1	1375.21	1360.04	1344.92	1329.38	1313.65	1297.75		
2	1256.81	1240.16	1223.38	1208.57	1190.73	1172.84		
4	1280.28	1261.9	1243.15	1226.37	1208.53	1191.65		
6	1265.01	1248.56	1231.87	1214.92	1198.01	1181.12		
8	1264.39	1248.02	1231.29	1214.52	1197.69	1180.81		
10	1264.38	1248.12	1231.34	1214.52	1197.67	1180.77		
12	1264.2	1247.56	1230.78	1213.98	1197.14	1180.25		
14	1264.5	1248.16	1231.1	1214.3	1197.47	1180.59		
16	1264.39	1248.04	1231.3	1214.51	1197.66	1180.77		
18	1264.02	1248.65	1231.97	1215.21	1197.84	1181.53		
20	1264.41	1248.27	1231.82	1215.07	1198.24	1181.35		
22	1264.72	1249.12	1232.18	1215.27	1198.17	1181.32		
24	1264.1	1248.44	1231.98	1215.23	1198.4	1181.53		
26	1264.4	1249.03	1232.32	1215.56	1198.72	1181.36		
28	1264.72	1248.08	1231.62	1214.86	1198.04	1181.17		
30	1265.42	1249.77	1232.3	1216.52	1198.92	1182.82		
40	1274.84	1258.91	1242.77	1225.98	1208.92	1192.51		
50	1274.01	1258.2	1242.11	1224.98	1208.4	1192.1		
60	1273.47	1257.44	1241.24	1224.05	1207.79	1191.58		
70	1272.81	1256.77	1240.9	1223.61	1207.46	1190.65		
80	1272.54	1256.27	1240.55	1223.24	1206.86	1190.32		
90	1271.89	1255.75	1239.84	1223.05	1206.45	1189.88		
100	1271.27	1255.38	1239.66	1222.4	1205.82	1189.2		
110	1270.9	1254.8	1239.19	1221.85	1205.38	1188.51		
120	1270.66	1254.58	1239	1221.24	1204.57	1187.93		
130	1270.42	1254.2	1238.6	1220.85	1204.28	1187.26		
140	1270.18	1253.8	1238.16	1220.64	1203.7	1187.02		
150	1269.85	1253.43	1237.51	1220.08	1203.16	1186.45		
160	1269.3	1252.87	1237.05	1219.4	1202.64	1185.8		
170	1268.9	1252.21	1236.39	1218.87	1202.09	1185.25		
180	1268.14	1251.53	1235.74	1218.38	1201.26	1184.71		
190	1267.69	1251.11	1234.99	1218	1200.78	1184.11		
200	1267.04	1250.44	1234.53	1217.3	1200.12	1183.49		

**Table 2:** The ultrasonic speeds, u/ (m s<sup>-1</sup>) of ethanol control (0 potency, 91% ethanol in water) and 33 formulations of ammonium causticum in ethanol control, as function of potency, C of ammonium causticum (in centesimal) at the temperatures (293.15-318.15) K and atmospheric pressure.

**Table 3:** The viscosities,  $\eta/(10^{-3} \text{ N s m}^2)$  of ethanol control (0 potency, 91% ethanol in water) and 33 formulations of ammonium causticum in ethanol control, as function of potency, C of ammonium causticum (in centesimal) at the temperatures (293.15-318.15) K and atmospheric pressure.

Potency	T/K							
(C)	293.15	298.15	303.15	308.15	313.15	318.15		
0	1.2072	1.0957	0.994	0.9055	0.8316	0.7642		
1	2.4562	2.1245	1.8405	1.6066	1.414	1.2582		

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41.94721.75441.56261.42511.28271.61.82081.60121.42541.27381.15141.81.81271.60471.42561.27771.14771.101.81291.6031.42471.27431.15291.121.81131.60421.42211.27691.15261.141.81311.60321.42631.27151.15131.161.81211.59941.42261.27121.15021.181.81181.60331.42731.27651.15191.201.81091.60251.42671.27311.1511.221.80781.60361.42411.27311.1511.	0381 1631 0303 0299 0312 0309 0301 0312 0308
61.82081.60121.42541.27381.15141.81.81271.60471.42561.27771.14771.101.81291.6031.42471.27431.15291.121.81131.60421.42211.27691.15261.141.81311.60321.42631.27151.15131.161.81211.59941.42261.27121.15021.181.81181.60331.42731.27611.15091.201.81091.60251.42671.27651.15191.221.80781.60361.42411.27311.1511.	0303 0299 0312 0309 0301 0312
8   1.8127   1.6047   1.4256   1.2777   1.1477   1.     10   1.8129   1.603   1.4247   1.2743   1.1529   1.     12   1.8113   1.6042   1.4221   1.2769   1.1526   1.     14   1.8131   1.6032   1.4263   1.2715   1.1513   1.     16   1.8121   1.5994   1.4226   1.2712   1.1502   1.     18   1.8118   1.6033   1.4273   1.2761   1.1509   1.     20   1.8109   1.6025   1.4267   1.2765   1.1519   1.     22   1.8078   1.6036   1.4241   1.2731   1.151   1.	.0299 .0312 .0309 .0301 .0312
10   1.8129   1.603   1.4247   1.2743   1.1529   1.     12   1.8113   1.6042   1.4221   1.2769   1.1526   1.     14   1.8131   1.6032   1.4263   1.2715   1.1513   1.     16   1.8121   1.5994   1.4226   1.2712   1.1502   1.     18   1.8118   1.6033   1.4273   1.2761   1.1509   1.     20   1.8109   1.6025   1.4267   1.2765   1.1519   1.     22   1.8078   1.6036   1.4241   1.2731   1.151   1.	0312 .0309 .0301 .0312
12 1.8113 1.6042 1.4221 1.2769 1.1526 1.   14 1.8131 1.6032 1.4263 1.2715 1.1513 1.   16 1.8121 1.5994 1.4226 1.2712 1.1502 1.   18 1.8118 1.6033 1.4273 1.2761 1.1509 1.   20 1.8109 1.6025 1.4267 1.2765 1.1519 1.   22 1.8078 1.6036 1.4241 1.2731 1.151 1.	.0309 .0301 .0312
14   1.8131   1.6032   1.4263   1.2715   1.1513   1.     16   1.8121   1.5994   1.4226   1.2712   1.1502   1.     18   1.8118   1.6033   1.4273   1.2761   1.1509   1.     20   1.8109   1.6025   1.4267   1.2765   1.1519   1.     22   1.8078   1.6036   1.4241   1.2731   1.151   1.	.0301 .0312
16   1.8121   1.5994   1.4226   1.2712   1.1502   1.     18   1.8118   1.6033   1.4273   1.2761   1.1509   1.     20   1.8109   1.6025   1.4267   1.2765   1.1519   1.     22   1.8078   1.6036   1.4241   1.2731   1.151   1.	.0312
18   1.8118   1.6033   1.4273   1.2761   1.1509   1.     20   1.8109   1.6025   1.4267   1.2765   1.1519   1.     22   1.8078   1.6036   1.4241   1.2731   1.151   1.	
20   1.8109   1.6025   1.4267   1.2765   1.1519   1.     22   1.8078   1.6036   1.4241   1.2731   1.151   1.	0308
22   1.8078   1.6036   1.4241   1.2731   1.151   1.	
	.0294
	.0283
24   1.8111   1.6073   1.4286   1.275   1.1522   1.	.0285
26   1.8118   1.6056   1.4256   1.2742   1.1518   1	.028
28   1.8371   1.6291   1.4497   1.2916   1.1585   1.	.0374
30   1.8829   1.6756   1.4912   1.3415   1.1914   1.	.0684
40 1.8743 1.6651 1.4855 1.3174 1.1853 1.	.0605
50 1.8646 1.6548 1.4756 1.3085 1.1753 1.	.0546
60   1.8559   1.6448   1.4673   1.3013   1.1686   1.	.0492
70 1.8461 1.6362 1.4586 1.2938 1.1623 1.	.0421
80 1.8361 1.6277 1.4541 1.2894 1.1584 1.	.0388
90   1.8272   1.6202   1.4477   1.2853   1.1559   1.	.0347
100 1.82 1.6141 1.4419 1.2788 1.1496 1.	.0302
110   1.8131   1.606   1.4363   1.2735   1.1457   1	.028
120   1.8058   1.6004   1.4296   1.2687   1.1408   1.	.0233
130   1.7985   1.5924   1.4246   1.2652   1.1364   1.	.0195
140   1.7939   1.5851   1.4182   1.2596   1.1319   1.	.0172
150   1.7868   1.5786   1.4129   1.2566   1.1297   1.	.0134
160   1.781   1.5722   1.4062   1.2523   1.1243   1.	.0092
170   1.7735   1.5657   1.3994   1.2481   1.1207   1.	.0067
180   1.769   1.56   1.3961   1.2438   1.1166   1.	.0035
190   1.7614   1.5543   1.3901   1.2405   1.1132   1.	.0002
200 1.7543 1.5472 1.3854 1.2361 1.1101 0.	

**Table 4:** Isentropic compressibilities,  $\kappa_s / (10^{-10} \text{ m}^2 \text{ N}^{-1})$  of ethanol control (0 potency, 91% ethanol in water) and 33 formulations of ammonium causticum in ethanol controls, as function of potency, C of ammonium causticum (in centesimal) at the temperatures (293.15-318.15) K and atmospheric pressure.

Potency		Т/К					
(C)	293.15	298.15	303.15	308.15	313.15	318.15	
0	9.101	9.405	9.726	10.063	10.414	10.789	
1	6.084	6.251	6.425	6.609	6.804	7.009	
2	7.646	7.894	8.156	8.403	8.705	9.023	
4	7.343	7.601	7.874	8.134	8.423	8.711	
6	7.547	7.787	8.043	8.313	8.597	8.894	
8	7.556	7.797	8.053	8.322	8.606	8.904	

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10	7.555	7.794	8.051	8.321	8.604	8.903
12	7.559	7.803	8.06	8.33	8.614	8.913
14	7.555	7.795	8.055	8.325	8.608	8.908
16	7.556	7.797	8.053	8.323	8.606	8.904
18	7.559	7.787	8.042	8.311	8.601	8.89
20	7.554	7.791	8.044	8.312	8.595	8.893
22	7.549	7.781	8.037	8.309	8.596	8.894
24	7.557	7.789	8.041	8.31	8.592	8.89
26	7.554	7.781	8.037	8.305	8.588	8.892
28	7.551	7.795	8.047	8.316	8.599	8.897
30	7.536	7.77	8.033	8.29	8.581	8.866
40	7.396	7.624	7.865	8.128	8.405	8.687
50	7.407	7.635	7.874	8.141	8.413	8.693
60	7.414	7.644	7.885	8.153	8.421	8.702
70	7.423	7.653	7.89	8.16	8.427	8.716
80	7.427	7.66	7.896	8.165	8.436	8.721
90	7.436	7.667	7.906	8.169	8.443	8.729
100	7.445	7.673	7.91	8.179	8.453	8.741
110	7.45	7.681	7.917	8.188	8.46	8.752
120	7.454	7.685	7.921	8.197	8.474	8.761
130	7.459	7.692	7.928	8.204	8.479	8.773
140	7.464	7.699	7.935	8.209	8.49	8.779
150	7.469	7.706	7.946	8.219	8.5	8.79
160	7.477	7.715	7.955	8.232	8.51	8.803
170	7.484	7.725	7.967	8.242	8.521	8.814
180	7.496	7.737	7.978	8.251	8.535	8.825
190	7.505	7.745	7.991	8.26	8.546	8.838
200	7.515	7.757	8	8.273	8.559	8.852

**Table 5:** Intermolecular free lengths,  $L_{\rm f}$  / (10<sup>-10</sup> m) of ethanol control (0 potency, 91% ethanol in water) and 33 formulations of ammonium causticum in ethanol control, as function of potency, C of ammonium causticum (in centesimal) at the temperatures (293.15-318.15) K.

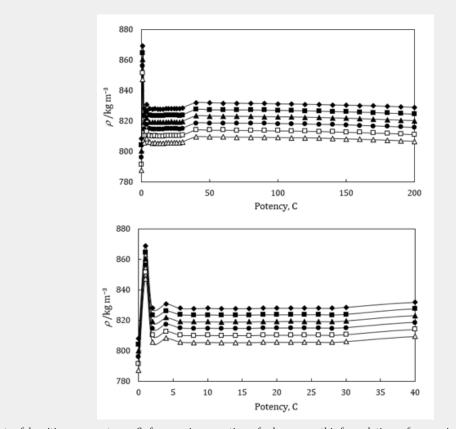
Potency	Т/К							
(C)	293.15	298.15	303.15	308.15	313.15	318.15		
0	6.149	6.308	6.473	6.644	6.819	7.002		
1	5.027	5.143	5.261	5.384	5.512	5.644		
2	5.636	5.779	5.928	6.071	6.234	6.404		
4	5.523	5.671	5.824	5.973	6.133	6.292		
6	5.599	5.74	5.886	6.038	6.196	6.358		
8	5.602	5.743	5.89	6.042	6.199	6.361		
10	5.602	5.742	5.889	6.041	6.198	6.361		
12	5.603	5.745	5.893	6.045	6.202	6.364		
14	5.602	5.743	5.891	6.043	6.2	6.363		
16	5.602	5.743	5.89	6.042	6.199	6.361		
18	5.603	5.74	5.886	6.038	6.197	6.356		

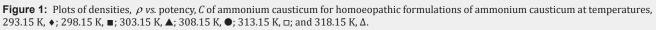
20	5.601	5.741	5.887	6.038	6.195	6.357
22	5.6	5.737	5.884	6.037	6.195	6.358
24	5.603	5.74	5.886	6.037	6.194	6.356
26	5.601	5.738	5.884	6.036	6.192	6.357
28	5.6	5.742	5.888	6.04	6.196	6.359
30	5.595	5.733	5.883	6.03	6.19	6.348
40	5.543	5.679	5.821	5.971	6.126	6.283
50	5.547	5.683	5.824	5.976	6.129	6.285
60	5.549	5.687	5.828	5.98	6.132	6.289
70	5.553	5.69	5.83	5.983	6.134	6.294
80	5.554	5.692	5.832	5.985	6.137	6.295
90	5.557	5.695	5.836	5.986	6.14	6.298
100	5.561	5.697	5.837	5.99	6.144	6.303
110	5.563	5.701	5.84	5.993	6.146	6.307
120	5.564	5.702	5.841	5.996	6.151	6.31
130	5.566	5.705	5.844	5.999	6.153	6.314
140	5.568	5.707	5.847	6.001	6.157	6.316
150	5.57	5.71	5.851	6.004	6.161	6.32
160	5.573	5.713	5.854	6.009	6.164	6.325
170	5.576	5.717	5.858	6.013	6.168	6.329
180	5.58	5.721	5.863	6.016	6.173	6.333
190	5.583	5.724	5.867	6.019	6.177	6.338
200	5.587	5.728	5.871	6.024	6.182	6.343

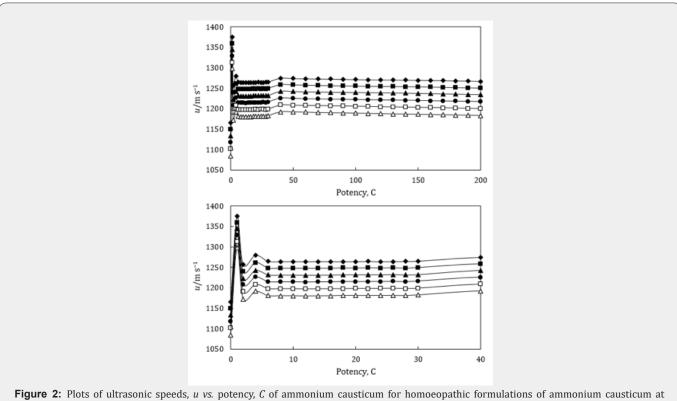
**Table 6:** Specific acoustic impedances, Z/ (106 kg m² s<sup>-1</sup>) of ethanol control (0 potency, 91% ethanol in water) and 33 formulations of ammoniumcausticum in ethanol control, as function of potency, C of ammonium causticum (in centesimal) at the temperatures (293.15-318.15) K.

Potency			T/K			
(C)	293.15	298.15	303.15	308.15	313.15	318.15
0	0.9424	0.9247	0.907	0.8894	0.8719	0.8543
1	1.1952	1.1762	1.1573	1.1381	1.1188	1.0994
2	1.0406	1.0214	1.0022	0.9847	0.9648	0.9449
4	1.0638	1.0425	1.0217	1.0025	0.9823	0.9633
6	1.0474	1.0285	1.0093	0.9901	0.9709	0.9519
8	1.0467	1.0277	1.0085	0.9894	0.9702	0.9512
10	1.0469	1.028	1.0087	0.9895	0.9704	0.9513
12	1.0465	1.0273	1.008	0.9889	0.9697	0.9507
14	1.0467	1.0278	1.0084	0.9892	0.9701	0.9509
16	1.0467	1.0277	1.0085	0.9893	0.9702	0.9511
18	1.0467	1.0285	1.0093	0.9902	0.9706	0.952
20	1.047	1.0282	1.0092	0.9901	0.971	0.9519
22	1.0473	1.0289	1.0097	0.9903	0.9709	0.9518
24	1.0468	1.0284	1.0094	0.9903	0.9711	0.9521
26	1.047	1.0289	1.0097	0.9905	0.9714	0.9519

28	1.0471	1.0279	1.009	0.9898	0.9707	0.9516
30	1.0486	1.0297	1.0102	0.9916	0.972	0.9535
40	1.0605	1.0418	1.0231	1.0036	0.9842	0.9653
50	1.0597	1.041	1.0225	1.0027	0.9837	0.965
60	1.0591	1.0403	1.0217	1.002	0.9832	0.9644
70	1.0585	1.0398	1.0213	1.0015	0.9828	0.9637
80	1.0581	1.0392	1.0209	1.0012	0.9822	0.9633
90	1.0574	1.0386	1.0201	1.0009	0.9818	0.9628
100	1.0566	1.0382	1.0198	1.0002	0.9811	0.9621
110	1.0561	1.0375	1.0193	0.9996	0.9806	0.9614
120	1.0558	1.0371	1.019	0.9989	0.9797	0.9609
130	1.0553	1.0365	1.0184	0.9984	0.9793	0.9601
140	1.0548	1.0359	1.0178	0.9979	0.9785	0.9596
150	1.0543	1.0353	1.017	0.9972	0.9778	0.9589
160	1.0537	1.0346	1.0161	0.9962	0.9771	0.958
170	1.053	1.0337	1.0152	0.9954	0.9763	0.9572
180	1.052	1.0327	1.0143	0.9947	0.9754	0.9564
190	1.0511	1.032	1.0133	0.9939	0.9745	0.9555
200	1.0502	1.031	1.0125	0.9929	0.9735	0.9546

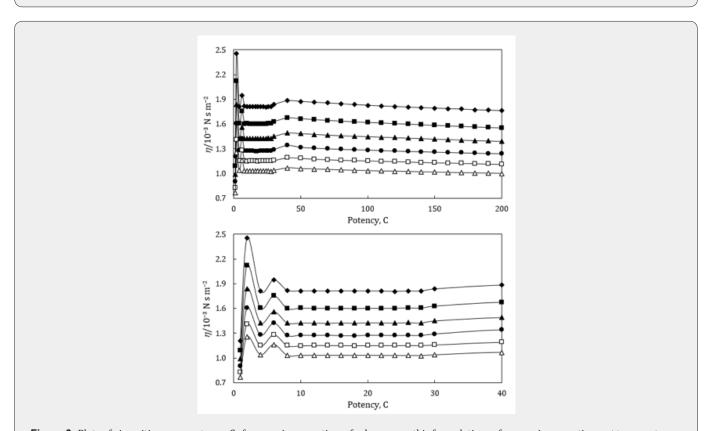




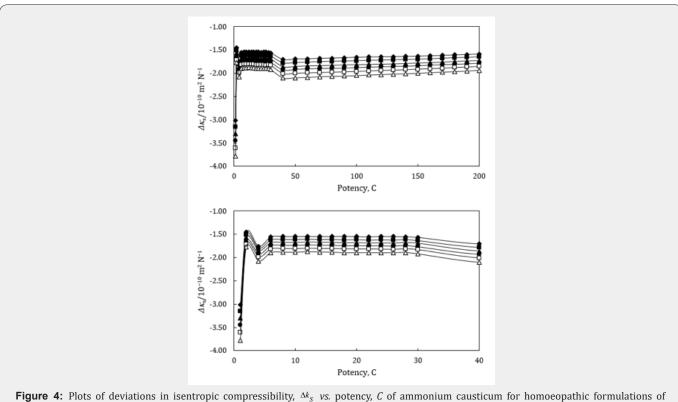


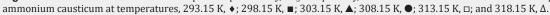


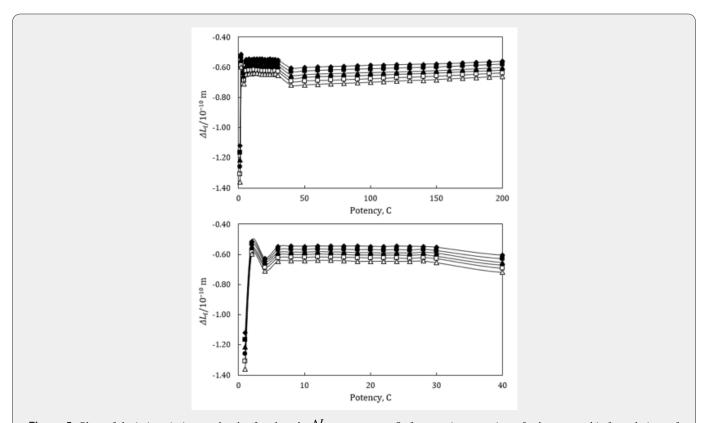
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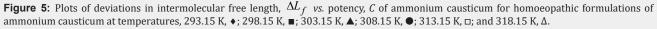


**Figure 3:** Plots of viscosities,  $\eta$  vs. potency, C of ammonium causticum for homoeopathic formulations of ammonium causticum at temperatures, 293.15 K,  $\blacklozenge$ ; 298.15 K,  $\blacklozenge$ ; 303.15 K,  $\blacklozenge$ ; 308.15 K,  $\blacklozenge$ ; 313.15 K,  $\Box$ ; and 318.15 K,  $\Delta$ .

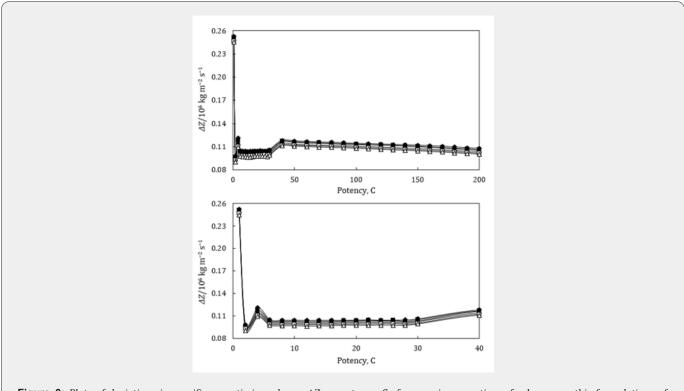


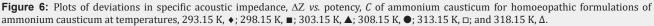


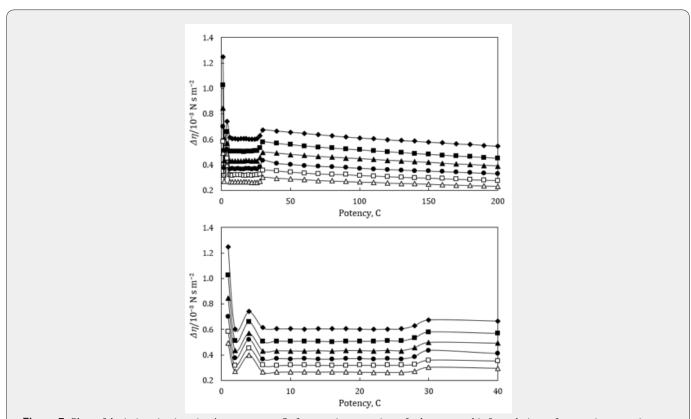


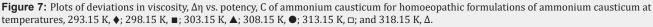


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## Discussion

A close examination of Tables 1-3 and Figure 1-3 indicates that the values of  $\rho$  and u and  $\eta$  of ammonium causticum in ethanol are greater than those of ethanol control for all the potencies (1C to 200C) at each investigated temperature and these values decrease with increase in temperature. The values of p and u are maximum at 1C and then decrease significantly in presence of ammonium causticum for simple successive dilution to the potency 2C and after that these values increase to maxima at 4C. Thereafter, these values decrease till 6C, then these values remain nearly constant up to 30C, and again increase till 40C and thereafter, these values decrease regularly up to 200C (Figure 1 and 2) with successive dilutions. The values of values of  $\eta$  are maximum at 1C and then decrease significantly in presence of ammonium causticum for simple successive dilution to the potency 2C and after that these values increase to maxima at 4C. Thereafter, these values decrease till 6C, then these values remain nearly constant up to 26C, and again increase till 30C and thereafter, these values decrease regularly up to 200C with successive dilutions. (Figure 3). The observed anomalous trends in  $\rho$ , u and  $\eta$  at certain potencies indicates that these potencies exhibit different solution structure from that of ethanol control.

A close perusal of Tables 4 and 5 indicate that the values of

 $\kappa_{\rm s}$  and  $L_{\rm f}$  for all the ammonium causticum potencies are less than those of ethanol controls at each investigated temperature and these values increase with increase in temperature, which indicates significant interaction between ammonium causticum and ethanol molecules. The values of  $\kappa_{\rm s}$  and  $L_{\rm f}$  are minimum at 1C and then increase significantly in presence of ammonium causticum for simple successive dilution to the potency 2C and after that these values decrease to minima at 4C. Thereafter, these values increase till 6C, then these values remain nearly constant up to 30C, and again decrease till 40C and thereafter, these values increase slightly regularly up to 200C (Figure 1 and 2) with successive dilutions (Tables 4 and 5). These variations

in  $\kappa_{\rm s}$  and  $L_{\rm f}$  are expressed in terms of deviations in isentropic compressibility,  $\Delta \kappa_{\rm s}$  and deviations in intermolecular free length,  $\Delta L_{\rm f}$  and are shown graphically in Figure 4 and 5. Figures 4 and 5 indicate that the values of  $\Delta \kappa_{\rm s}$  and  $\Delta L_{\rm f}$  are negative for all the ammonium causticum potencies are less than those of ethanol controls at each investigated temperature, indicating that the solutions are more compressible in presence of ammonium causticum than ethanol controls. This may be due to the formation of stronger interactions between ammonium causticum and ethanol-water molecules than the hydrogen bonding interactions present in ethanol control.

The  $\Delta \kappa_s$  and  $\Delta L_f$  are minimum at 1C and then increase significantly in presence of ammonium causticum for simple successive dilution to the potency 2C and after that these values decrease to minima at 4C. Thereafter, these values increase till 6C, then these values remain nearly constant up to 30C, and again decrease till 40C and thereafter, these values increase slightly regularly up to 200C (Figure 4 and 5) with successive dilutions (Tables 4 and 5). This indicates that the potencies 1C, 4C and 40C are less compressible and 2C, 6C to 30C are more compressible than the other potencies, indicating that the potencies 1C, 4C and 40C exhibit more compact solution structure and the potencies 2C, 6C to 30C exhibit less compact solution structure as compared

to other potencies and ethanol control. The minimum in  $\Delta \kappa_{s}$  and

 $\Delta L_{\rm f}$  values at potencies 1C, 4C and 40C indicate that these have most compact solution structure as compared to other potencies, hence, these potencies may show different behaviour in terms of efficacy when used in practice.

A close perusal of Table 6 indicates that the acoustic impedances, Z of potencies of ammonium causticum are more than those of ethanol control for all the potencies at each investigated temperature and the values decrease with increase in temperature, which indicates significant interaction between ammonium causticum and ethanol/water molecules. The values of Z are maximum at 1C and then decrease significantly in presence of ammonium causticum for simple successive dilution to the potency 2C and after that these values increase to maximum at 4C. Thereafter, these values decrease till 6C, then these values remain nearly constant up to 30C, and again increase till 40C and thereafter, these values decrease regularly up to 200C (Table 6) with successive dilutions. These variations in Z are expressed in terms of deviations in acoustic impedance,  $\Delta Z$  and are shown in Figure 6. Figure 6 indicates that values are positive, i.e., Z values for potencies of ammonium causticum are more than those of ethanol control. These values are maximum at 1C and then decrease significantly in presence of ammonium causticum for simple successive dilution to the potency 2C and after that these values increase to maximum at 4C. Thereafter, these values decrease till 6C, then these values remain nearly constant up to 30C, and again increase till 40C and thereafter, these values decrease regularly up to 200C (Figure 6). This indicates that the potencies 1C, 4C and 40C possess more compact solution structure and 2C, 6C to 30C possess less compact structure than the other potencies and ethanol control. The variations in values of Z and  $\Delta Z$  of these potencies may be due to interaction between ammonium causticum and ethanol/water molecules.

The variations in  $\eta$  are expressed in terms of deviations in viscosity,  $\Delta \eta$  and are shown in Figure 7. Figure 7 indicates that  $\Delta \eta$  values are positive, i.e.,  $\eta$  values for ammonium causticum are more than those of ethanol control. These  $\Delta \eta$  values are maximum at 1C and then decrease significantly in presence of ammonium causticum for simple successive dilution to the potency 2C and after that these values increase to maxima at 4C. Thereafter, these values decrease till 6C, then these values remain nearly constant up to 26C, and again increase till 30C and thereafter, these values decrease regularly up to 200C with successive dilutions (Figure 7).

The variations in values of  $\eta$  and  $\Delta \eta$  of these potencies may be due interaction between ammonium causticum and ethanol/ water molecules. It is observed that the variations observed in the values of measured properties,  $\rho$  and u and  $\eta$ ; and calculated

parameters,  $\kappa_{\rm s}$ ,  $L_{\rm f}$ ,  $\Delta \kappa_{\rm s}$ ,  $\Delta L_{\rm f}$ ,  $\Delta Z$  and  $\Delta \eta$  support each other. Similar anomalous trends in these properties have also been reported earlier for different potencies of ammonium aceticum [28] and acidum salicylicum [25].

#### Conclusion

The densities, ultrasonic speeds, and viscosities of ethanol control, 33 formulations of ammonium causticum in ethanol control are measured for potencies from 1C to 200C (with an interval of 2C up to 30C and then of 10C up to 200C) at six different temperatures and atmospheric pressure. From these experimental

data, various physicochemical parameters, viz.,  $\kappa_s$ ,  $L_f$ , Z,

 $\Delta \kappa_{\rm s}$ ,  $\Delta L_{\rm f}$ ,  $\Delta Z$  and  $\Delta \eta$  were calculated. The results have been qualitatively discussed in terms of interactions/physicochemical behaviour of these extremely dilute homoeopathic formulations of ammonium causticum in ethanol. This indicates that the potencies 1C, 4C and 40C possess more compact solution structure and 2C, 6C to 30C possess less compact structure as compared to other potencies and ethanol control. Hence, these potencies may have different behaviour in terms of efficacy when used in practice. It can be qualitatively concluded that even in extreme dilutions the molecules of ammonium causticum may be present in these homoeopathic formulations, however it needs to be confirmed from other techniques.

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