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Physicochemical behaviour of homoeopathic dilutions of ammonium carbonicum at ambient temperatures: Volumetric, ultrasonic and viscometric study

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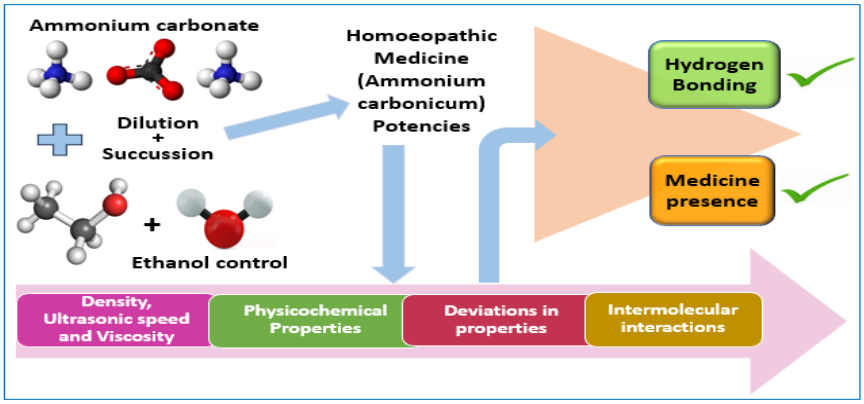
ABSTRACT



The physicochemical behaviour of homoeopathic dilutions of ammonium carbonicum has been studied from the measurements of the densities, ρ, ultrasonic speeds, u and viscosities, η of pure ethanol control and 23 formulations of potencies of ammonium carbonicum ranging from 1C to 100C (with intervals of 2C up to 30C and thereafter with intervals of 10C up to 100C) at six different temperatures and atmospheric pressure. Various physicochemical properties, *viz.*, the isentropic compressibilities, κ_s, intermolecular free length, L_f, pseudo-Grüneisen parameter, Γ, deviations in isentropic compressibility, $\Delta\kappa_s$, deviations in intermolecular free length, ΔL_f, deviation in pseudo-Grüneisen parameter, $\Delta\Gamma$ and deviations in viscosity, $\Delta\eta$ have been calculated. The variations of these properties with potency show strange behaviour at certain potencies. The results have been qualitatively discussed in terms of interactions of these ammonium carbonicum homoeopathic formulations. The results show that even in high dilutions (≈ 100C) the molecules of ammonium carbonicum may be present in these homoeopathic formulations.

Keywords: Density, Ultrasonic speed, Viscosity, Homoeopathic medicines, Ammonium carbonicum, Extremely diluted solutions.

INTRODUCTION

Homeopathy is known for its holistic healing approach, but it remains controversial due to two main reasons. Firstly, the preparation of homeopathic medicines involves ultra-high dilutions, raising questions about the presence of active principles even at such extreme levels. The homoeopathic medicines are prepared using the blend of two processes, *viz.*, a dilution of 1:100 followed by succussion and these "extremely diluted solutions" show anomalous behaviour in medicinal efficacy. Secondly, the efficacy of homoeopathic medicines is well supported by research evidences; however, there are debates regarding questionability in biological activity of these medicines wherein the source drug is diluted beyond Avagadro's limit, *i.e.*, the ultra-diluted medicine formulation might be similar to the solvent. There have been a few research studies relating to explore the presence of drug in ultra-diluted medicines[1–6] and its mechanism of action, but the question still exists unreciprocated.

The measurements of physicochemical properties and derived parameters of aqueous and mixed-aqueous solutions of

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electrolytes, amino acids, carbohydrates, drugs, *etc.* have been supportive in characterizing the prevailing interactions, which are subsequently useful in understanding of solute-solvation/hydration behaviour of solute.[7–12] The homoeopathic formulations are dilute solutions, therefore, their physical properties, *viz.*, density, ultrasonic speed, viscosity, refractive index, *etc.* can be measured simply at varying potency (concentration) and temperature. The physicochemical properties derived from these experimental data can deliver valuable info regarding of physicochemical behaviour and mechanism of action of these homoeopathic medicines. There have been few physicochemical studies on extremely diluted solutions of inorganic salts[13–15] and homoeopathic medicines[16–20] by using physicochemical methods. These studies provided interesting and convincing information on the behaviour of these extremely diluted solutions. To the best of our knowledge, very rare physicochemical studies on homoeopathic medicines using volumetric, acoustic and viscometric have been reported in the literature.[21,22] In continuation to earlier research on the physicochemical behaviour of extremely diluted homoeopathic formulations,[23–25] here we report the results of our study on the physicochemical properties of homoeopathic dilutions of ammonium carbonicum.

The homeopathic remedy ammonium carbonicum is derived from ammonium carbonate. In its crude state, carbonate of ammonia is inert. When it undergoes potentization (process of preparing homeopathic medicines that extracts medicinal properties from a crude substance), it transforms into an important homeopathic medicine named ammonium carbonicum. It is used as a stimulant, produces perspiration and is also a cough expectorant. It aids to increase the alkalinity of blood and reduces coagulation tendencies. It is very beneficial against nasal catarrh.

In the present study, the densities, ρ, ultrasonic speeds, u and viscosities, η of pure ethanol control (91% ethanol in water) and 23 formulations of ammonium carbonicum with potencies from 1C to 100C (with intervals of 2C till 30C, and thereafter with intervals of 10C till 100C) at 293.15, 298.15, 303.15, 308.15, 313.15 and 318.15 K and at atmospheric pressure. From these experimental data, the isentropic compressibilities, κ_s, intermolecular free length, L_f, pseudo-Grüneisen parameter, Γ, deviations in isentropic compressibility, $\Delta\kappa_s$, deviations in intermolecular free length, ΔL_f, deviation in pseudo-Grüneisen parameter, $\Delta\Gamma$ and deviations in viscosity, $\Delta\eta$ have been calculated. The variations of these parameters with potency and temperature are qualitatively discussed in terms of interactions/ physicochemical behaviour of these ammonium carbonicum homoeopathic formulations.

EXPERIMENTAL

Ethanol control and homoeopathic formulations of various potencies of ammonium carbonicum used in the study were procured from Dr. Wilmer Schwabe India Pvt. Limited, India, prepared in accordance with Homoeopathic Pharmacopoia of India.[26] 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). This two-in-one instrument is equipped with both density and ultrasonic cells, with reproducibility of $\pm1 \times 10^{-3}$ kg·m^{-3} and $\pm1 \times 10^{-2}$ m·s^{-1} for density and ultrasonic speed, respectively. The temperature for both cells was kept constant by using built in Peltier thermostat within ±0.01 K. The calibration of instrument was done by using triply-distilled, degassed water and with dry air at atmospheric pressure and 293.15 K. The operating frequency used for ultrasonic speed measurements was 3 MHz. 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. The standard uncertainties related to the measurements of density, ultrasonic speed and temperature were found within ±0.05 kg·m^{-3}, ±0.5 m·s^{-1} and ±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 ±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 relative standard uncertainty in viscosity measurements was found to be within ±0.5 %.

RESULTS

The experimental values of densities, ρ, ultrasonic speeds, u and viscosities, η of homoeopathic formulations of ammonium carbonicum as function of potency, C (in centesimal) at different temperatures are listed in Tables 1–3, and are presented graphically in Figures 1–3, respectively.

Table 1: The densities, ρ/(kg m^{-3}) of ethanol control (0 potency, 91% ethanol in water) and 23 formulations of ammonium carbonicum in ethanol control, as function of potency, C of ammonium carbonicum (in centesimal) at the temperatures (293.15–318.15) K and atmospheric pressure

Potency (C)	T/K					
	293.15	298.15	303.15	308.15	313.15	318.15
0	789.460	785.212	780.820	776.420	771.781	767.380
1	1001.819	1000.546	999.048	997.346	995.457	993.392
2	988.269	987.106	985.712	984.097	982.279	980.274
4	830.539	826.183	821.774	817.319	812.806	808.226
6	827.808	823.454	819.049	814.592	810.075	805.494
8	828.092	823.737	819.331	814.872	810.356	805.776
10	827.784	823.428	819.024	814.565	810.049	805.470
12	827.722	823.362	818.954	814.498	809.979	805.397
14	827.894	823.535	819.128	814.666	810.148	805.571
16	827.911	823.555	819.144	814.986	810.744	805.890
18	827.805	823.451	819.043	814.586	810.068	805.489
20	827.800	823.444	819.040	814.582	810.065	805.486
22	827.914	823.559	819.153	814.694	810.176	805.596
24	827.726	823.371	818.960	814.503	809.988	805.409
26	827.748	823.390	818.987	814.529	810.013	805.434
28	827.731	823.376	818.970	814.509	809.993	805.414
30	827.717	823.364	818.957	814.498	809.980	805.395
40	827.781	823.425	819.021	814.988	810.110	805.710
50	828.232	823.878	819.472	815.014	810.496	805.916
60	828.151	823.794	819.392	814.930	810.417	805.838
70	851.925	847.516	843.071	838.582	834.050	829.465

80	827.736	823.382	818.975	814.516	809.998	805.417
90	828.113	823.757	819.352	814.894	810.377	805.796
100	827.770	823.409	819.009	814.550	810.035	805.455

Table 2: The ultrasonic speeds, u/(m s^{-1}) of ethanol control (0 potency, 91% ethanol in water) and 23 formulations of ammonium carbonicum in ethanol control, as function of potency, C of ammonium carbonicum (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	1165.93	1149.76	1133.54	1117.33	1101.38	1084.91
1	1542.66	1532.95	1523.76	1513.02	1501.74	1489.09
2	1531.19	1522.70	1512.70	1503.13	1493.12	1481.15
4	1271.15	1254.58	1237.89	1221.15	1204.36	1187.54
6	1264.29	1247.65	1230.87	1214.07	1197.23	1180.34
8	1265.12	1248.43	1231.66	1214.85	1198.02	1181.15
10	1264.20	1247.56	1230.78	1213.97	1197.13	1180.24
12	1264.03	1247.37	1230.59	1213.78	1196.94	1180.06
14	1264.68	1248.10	1231.35	1214.55	1197.71	1180.82
16	1264.52	1247.86	1231.09	1215.15	1198.32	1181.55
18	1264.28	1247.65	1230.89	1214.10	1197.28	1180.40
20	1264.24	1247.57	1230.80	1214.00	1197.16	1180.27
22	1264.52	1247.87	1231.10	1214.30	1197.46	1180.57
24	1264.03	1247.37	1230.61	1213.80	1196.95	1180.05
26	1264.11	1247.44	1230.67	1213.86	1197.01	1180.13
28	1264.05	1247.39	1230.61	1213.81	1196.97	1180.08
30	1264.03	1247.37	1230.60	1213.79	1196.94	1180.05
40	1264.19	1247.52	1230.83	1214.44	1197.11	1181.02
50	1265.39	1248.73	1231.98	1215.17	1198.34	1181.46
60	1265.15	1248.49	1231.73	1214.93	1198.11	1181.24
70	1279.67	1263.14	1246.43	1229.67	1212.86	1195.99
80	1264.07	1247.41	1230.64	1213.83	1196.98	1180.10
90	1265.22	1248.56	1231.80	1215.00	1198.16	1181.29
100	1264.16	1247.49	1230.72	1213.91	1197.06	1180.17

The values of the isentropic compressibility, κ_s, and intermolecular free length, L_f and pseudo-Grüneisen parameter, Γ have been calculated by using the following relations[27–30]

$$\kappa_s = 1/u^2\rho \quad (1)$$

$$L_f = K'/u\rho^{1/2} \quad (2)$$

$$\Gamma = \left[(\kappa_s/\kappa_T) - 1\right]/\alpha_p T \quad (3)$$

where K' is temperature dependent constant [= (93.875 + 0.375T) × 10^{-8}]; T is the absolute temperature; α_p is the isobaric expansivity and k_T is the isothermal compressibility. The values of αp and k_T are calculated using the relations[30–34]

$$\alpha_p = (-1/\rho)(\partial\rho/\partial T)_p \quad (4)$$

$$k_T = (1.71\times10^{-3})/(T^{4/9}u^2\rho^{4/3}) \quad (5)$$

The values of κ_s, L_f and Γ have been given in Tables 4–6, respectively.

Table 3: The viscosities, η/(10^{-3} N s m^{-2}) of ethanol control (0 potency, 91% ethanol in water) and 23 formulations of ammonium carbonicum in ethanol control, as function of potency, C of ammonium carbonicum (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	0.9940	0.9055	0.8316	0.7642
1	2.0239	1.8339	1.6820	1.5521	1.4321	1.3246
2	2.0587	1.8791	1.7223	1.5865	1.4671	1.3686
4	1.8483	1.6671	1.5134	1.3535	1.2286	1.1109
6	1.7437	1.5764	1.4038	1.2556	1.1284	1.0162
8	1.7575	1.5982	1.4074	1.2565	1.1230	1.0003
10	1.8122	1.6475	1.4601	1.2988	1.1728	1.0463
12	1.7801	1.5934	1.4173	1.2639	1.1353	1.0257
14	1.7816	1.5868	1.4165	1.2633	1.1325	1.0267
16	1.7821	1.5870	1.4161	1.2611	1.1350	1.0250
18	1.7837	1.5881	1.4153	1.2651	1.1346	1.0253
20	1.7858	1.5873	1.4162	1.2645	1.1316	1.0261
22	1.7840	1.5890	1.4169	1.2653	1.1317	1.0256
24	1.7866	1.5894	1.4179	1.2666	1.1328	1.0259
26	1.8169	1.6137	1.4426	1.2960	1.1651	1.0546
28	1.8189	1.6146	1.4425	1.2959	1.1668	1.0564
30	1.8014	1.5915	1.4233	1.2752	1.1453	1.0351
40	1.7984	1.5928	1.4246	1.2751	1.1443	1.0344
50	1.7971	1.5928	1.4243	1.2756	1.1426	1.0345
60	1.8016	1.5956	1.4242	1.2809	1.1465	1.0347
70	1.8675	1.6626	1.4868	1.3379	1.2012	1.0838
80	1.8016	1.6006	1.4262	1.2819	1.1525	1.0347
90	1.8003	1.5974	1.4230	1.2762	1.1462	1.0305
100	1.8011	1.5944	1.4211	1.2753	1.1451	1.0303

Table 4: Isentropic compressibilities, κ_s/(10^{-10} m^2 N^{-1}) of ethanol control (0 potency, 91% ethanol in water) and 23 formulations of ammonium carbonicum in ethanol controls, as function of potency, C of ammonium carbonicum (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	9.3180	9.6338	9.9673	10.3167	10.6815	11.0714
1	4.1944	4.2531	4.3110	4.3799	4.4544	4.5398
2	4.3159	4.3693	4.4335	4.4975	4.5664	4.6500
4	7.4515	7.6900	7.9412	8.2048	8.4820	8.7735
6	7.5575	7.8014	8.0587	8.3286	8.6123	8.9109
8	7.5450	7.7890	8.0456	8.3151	8.5980	8.8956
10	7.5588	7.8028	8.0601	8.3303	8.6140	8.9127
12	7.5614	7.8058	8.0633	8.3335	8.6175	8.9162
14	7.5520	7.7951	8.0517	8.3213	8.6046	8.9028
16	7.5538	7.7979	8.0549	8.3098	8.5896	8.8883
18	7.5576	7.8015	8.0585	8.3283	8.6117	8.9101
20	7.5582	7.8025	8.0597	8.3297	8.6134	8.9121
22	7.5538	7.7977	8.0547	8.3244	8.6079	8.9063
24	7.5613	7.8057	8.0630	8.3332	8.6173	8.9163
26	7.5602	7.8047	8.0619	8.3321	8.6161	8.9148
28	7.5611	7.8054	8.0629	8.3330	8.6169	8.9158
30	7.5614	7.8058	8.0632	8.3334	8.6175	8.9164
40	7.5589	7.8033	8.0595	8.3195	8.6137	8.8983
50	7.5405	7.7839	8.0400	8.3092	8.5919	8.8894
60	7.5441	7.7877	8.0441	8.3134	8.5960	8.8936
70	7.1681	7.3952	7.6348	7.8864	8.1505	8.4284
80	7.5608	7.8051	8.0625	8.3327	8.6167	8.9154
90	7.5436	7.7872	8.0436	8.3128	8.5957	8.8933
100	7.5594	7.8039	8.0611	8.3312	8.6152	8.9139

Table 5: Intermolecular free lengths, L_f/(10^{-10} m) of ethanol control (0 potency, 91% ethanol in water) and 23 formulations of ammonium carbonicum in ethanol control, as function of potency, C of ammonium carbonicum (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	6.2213	6.3840	6.5527	6.7268	6.9060	7.0933
1	4.1740	4.2418	4.3095	4.3830	4.4597	4.5422
2	4.2340	4.2993	4.3703	4.4415	4.5154	4.5970
4	5.5634	5.7037	5.8489	5.9990	6.1541	6.3144
6	5.6028	5.7449	5.8921	6.0440	6.2011	6.3637
8	5.5982	5.7403	5.8873	6.0391	6.1960	6.3582
10	5.6033	5.7454	5.8926	6.0446	6.2018	6.3643

12	5.6043	5.7465	5.8938	6.0458	6.2030	6.3656
14	5.6008	5.7425	5.8895	6.0414	6.1984	6.3608
16	5.6014	5.7436	5.8907	6.0372	6.1930	6.3556
18	5.6029	5.7449	5.8920	6.0439	6.2009	6.3634
20	5.6031	5.7453	5.8924	6.0444	6.2015	6.3641
22	5.6014	5.7435	5.8906	6.0425	6.1996	6.3621
24	5.6042	5.7465	5.8936	6.0457	6.2029	6.3656
26	5.6038	5.7461	5.8933	6.0453	6.2025	6.3651
28	5.6041	5.7464	5.8936	6.0456	6.2028	6.3654
30	5.6043	5.7465	5.8937	6.0458	6.2030	6.3657
40	5.6033	5.7456	5.8924	6.0407	6.2016	6.3592
50	5.5965	5.7385	5.8853	6.0370	6.1938	6.3560
60	5.5978	5.7398	5.8867	6.0385	6.1953	6.3575
70	5.4566	5.5933	5.7350	5.8814	6.0326	6.1890
80	5.6040	5.7463	5.8934	6.0455	6.2027	6.3653
90	5.5977	5.7397	5.8865	6.0383	6.1952	6.3574
100	5.6035	5.7458	5.8929	6.0450	6.2022	6.3648

Table 6: The pseudo-Grüneisen parameter, Γ for ethanol control (0 potency, 91% ethanol in water) and 23 formulations of ammonium carbonicum in ethanol control, as function of potency, *C* of ammonium carbonicum (in centesimal) at the temperatures (293.15–318.15) K

Potency (C)	*T*/K					
	293.15	298.15	303.15	308.15	313.15	318.15
0	2.0509	2.0172	1.9837	1.9510	1.9183	1.8870
1	7.3939	7.3123	7.2306	7.1489	7.0674	6.9861
2	7.6536	7.5701	7.4864	7.4024	7.3185	7.2346
4	2.1796	2.1441	2.1093	2.0751	2.0415	2.0084
6	2.1698	2.1345	2.0998	2.0657	2.0322	1.9992
8	2.1706	2.1352	2.1005	2.0664	2.0329	1.9999
10	2.1697	2.1344	2.0997	2.0656	2.0321	1.9991
12	2.1686	2.1333	2.0986	2.0645	2.0310	1.9981
14	2.1691	2.1338	2.0991	2.0650	2.0315	1.9985
16	2.1696	2.1343	2.0996	2.0657	2.0325	1.9992
18	2.1716	2.1363	2.1015	2.0674	2.0339	2.0009
20	2.1698	2.1344	2.0997	2.0657	2.0321	1.9992
22	2.1697	2.1344	2.0997	2.0656	2.0321	1.9991
24	2.1692	2.1338	2.0991	2.0650	2.0316	1.9986
26	2.1697	2.1343	2.0996	2.0656	2.0321	1.9991
28	2.1691	2.1338	2.0991	2.0650	2.0315	1.9986
30	2.1687	2.1334	2.0987	2.0646	2.0311	1.9982
40	2.1676	2.1323	2.0976	2.0650	2.0303	1.9980
50	2.1710	2.1357	2.1010	2.0669	2.0333	2.0004
60	2.1711	2.1357	2.1010	2.0669	2.0334	2.0004
70	2.2399	2.2036	2.1681	2.1332	2.0990	2.0654
80	2.1693	2.1340	2.0993	2.0652	2.0317	1.9987
90	2.1710	2.1356	2.1009	2.0668	2.0333	2.0003
100	2.1694	2.1340	2.0993	2.0653	2.0318	1.9988

The deviations in κ_s, L_f and η of ethanol due to addition of ammonium carbonicum with dilution and succussion are represented by deviation values of these properties. The deviations in isentropic compressibility, $\Delta\kappa_s$, deviations in intermolecular free length, ΔL_f, deviations in pseudo-Grüneisen parameter, $\Delta\Gamma$ and deviations in viscosity, $\Delta\eta$ have been calculated by using the following standard relations

$$\Delta\kappa_s = \kappa_s - \kappa_s^o \quad (6)$$

$$\Delta L_f = L_f - L_f^o \quad (7)$$

$$\Delta\Gamma = \Gamma - \Gamma^\circ \quad (8)$$

$$\Delta\eta = \eta - \eta^o \quad (9)$$

where the superscript 'o' represents the values for pure ethanol control (91% ethanol in water). The variations of $\Delta\kappa_s$, ΔL_f , $\Delta\Gamma$ and $\Delta\eta$ with potency, *C* of ammonium carbonicum and temperature are presented graphically in Figures. 4–7, respectively.

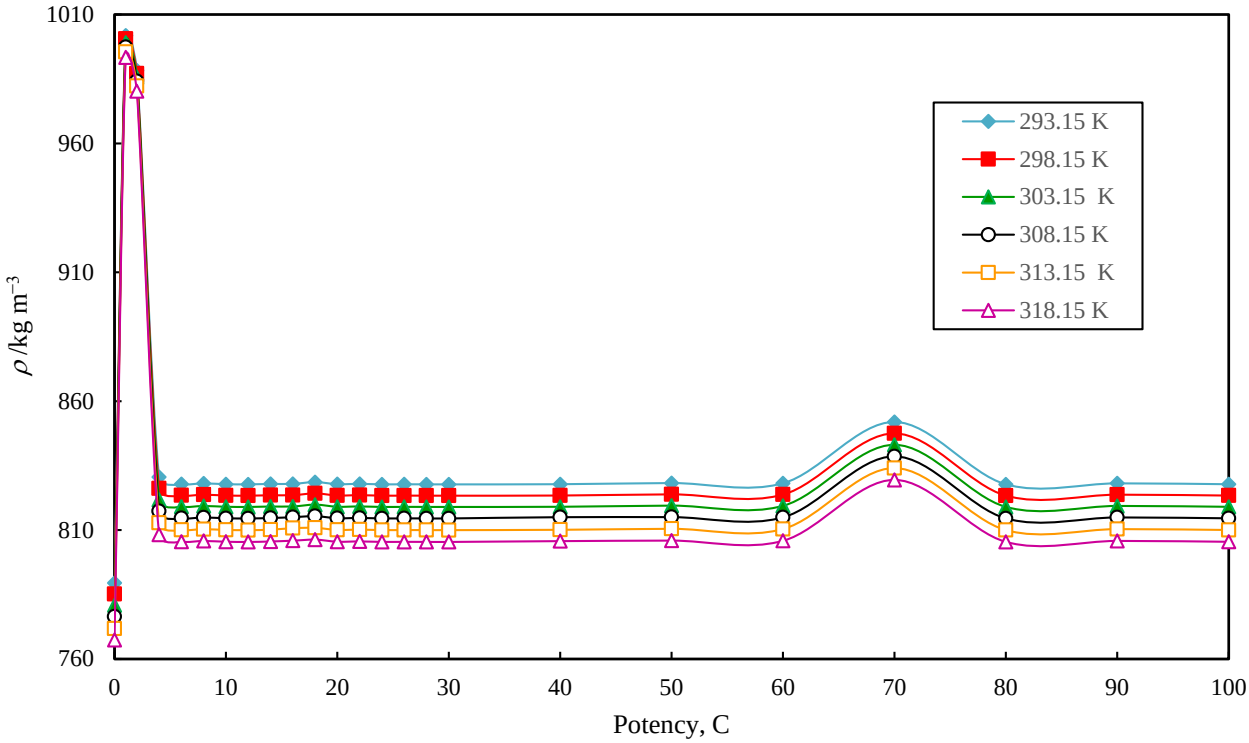

Figure 1. Plots of densities, ρ *vs.* potency, *C* of ammonium carbonicum for homoeopathic formulations of ammonium carbonicum at different temperatures

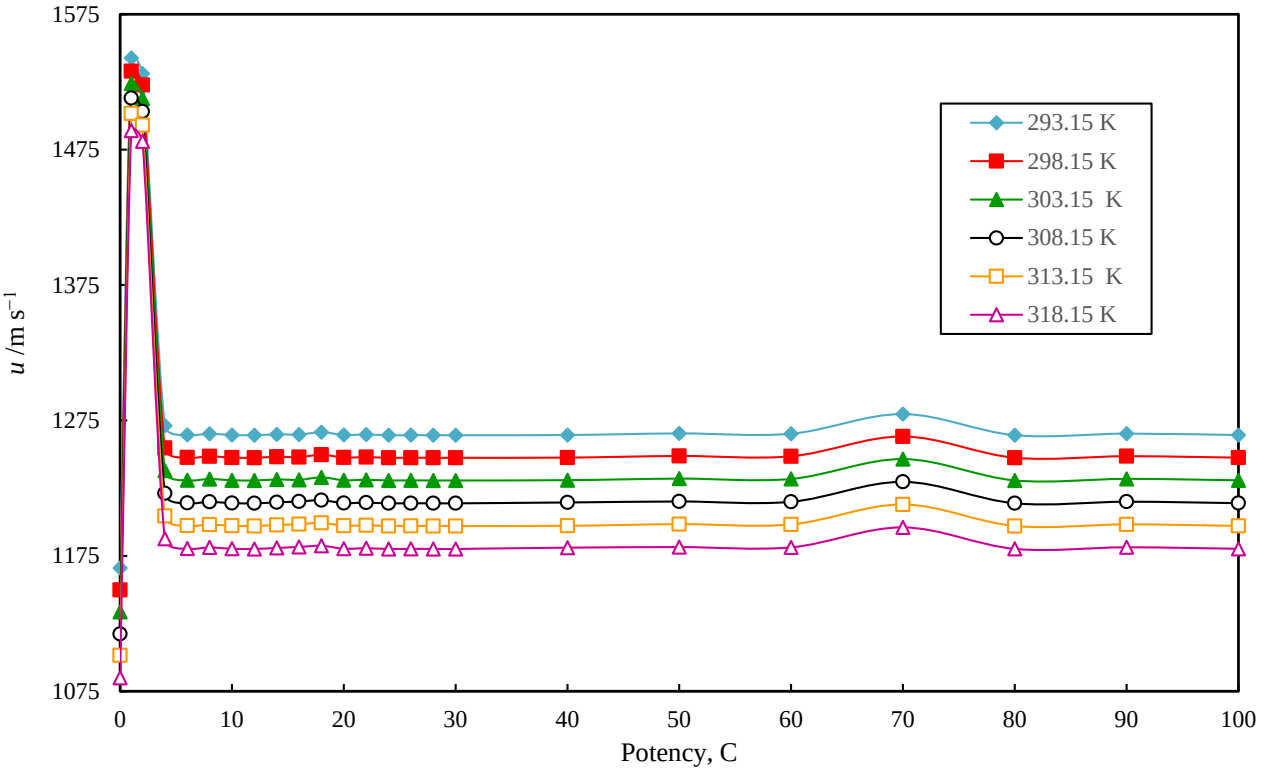

Figure 2. Plots of ultrasonic speeds, *u vs.* potency, *C* of ammonium carbonicum for homoeopathic formulations of ammonium carbonicum at different temperatures.

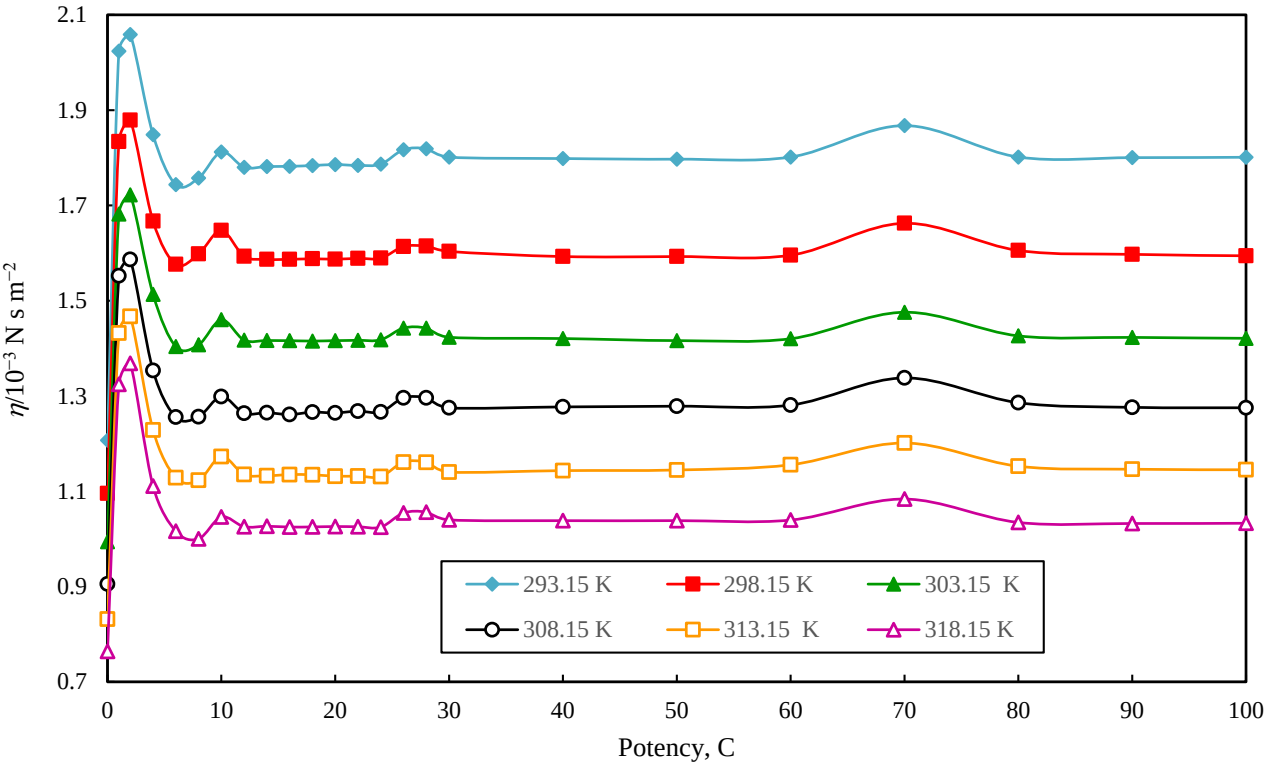

Figure 3. Plots of viscosities, *η vs.* potency, *C* of ammonium carbonicum for homoeopathic formulations of ammonium carbonicum at different temperatures.

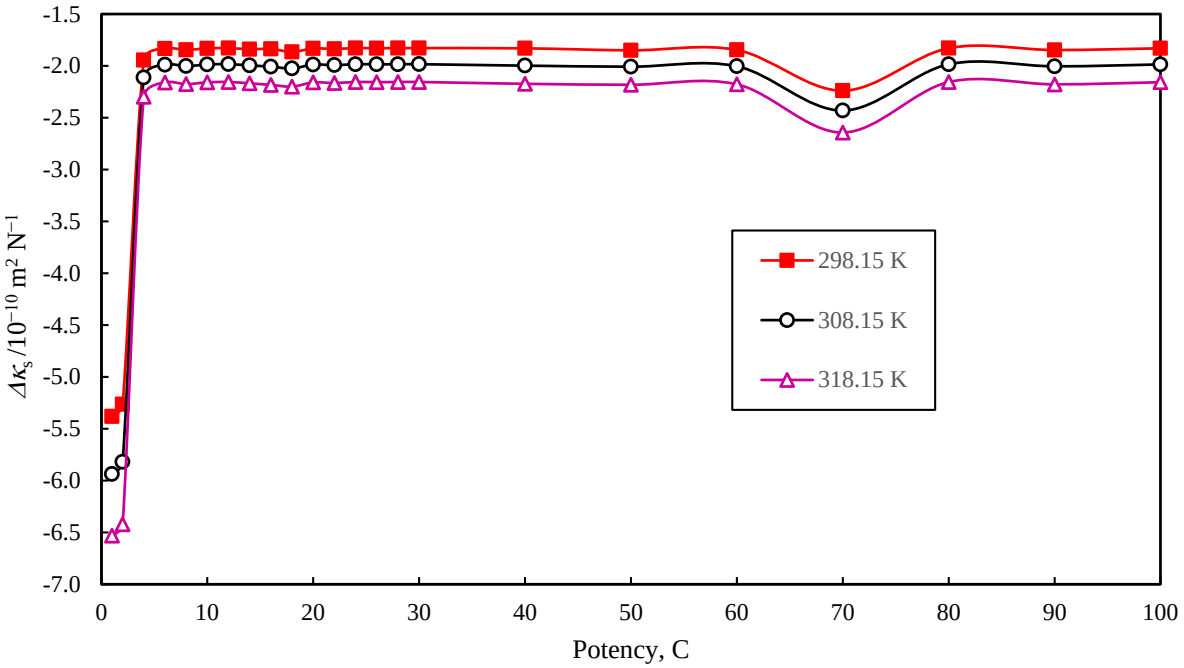

Figure 4. Plots of deviations in isentropic compressibilities, $\Delta\kappa_s$ *vs.* potency, *C* of ammonium carbonicum for homoeopathic formulations of ammonium carbonicum at different temperatures.

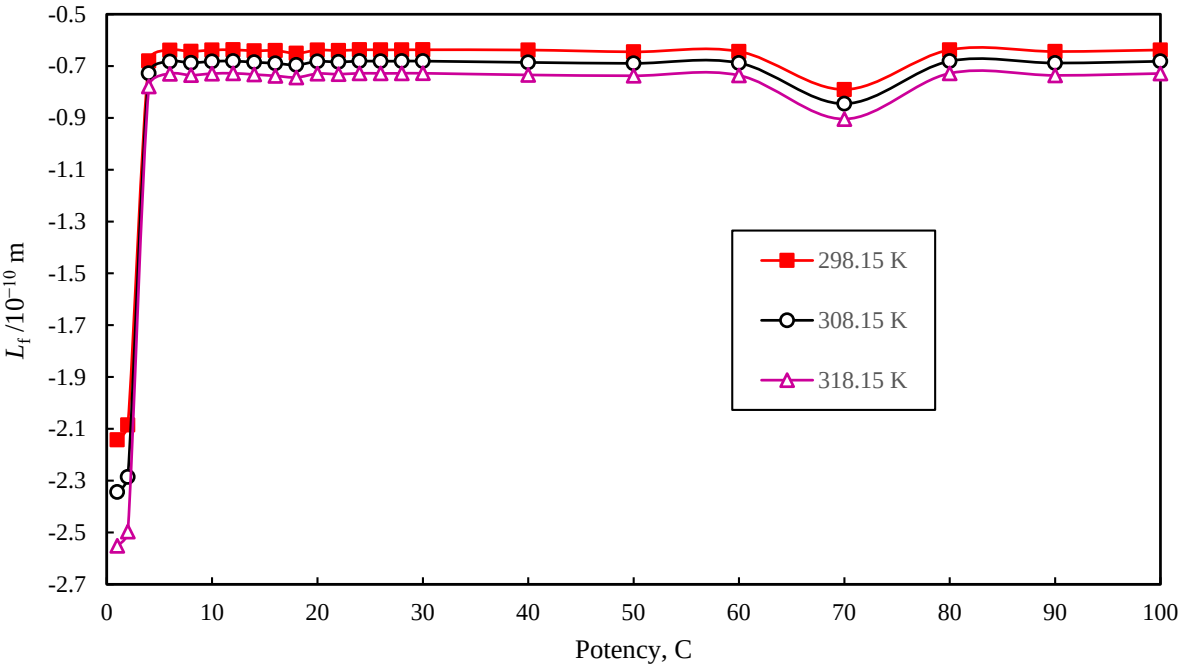

Figure 5. Plots of deviations in intermolecular free length, ΔL_f *vs.* potency, *C* of ammonium carbonicum for homoeopathic formulations of ammonium carbonicum at different temperatures.

DISCUSSION

A close perusal of Tables 1–3 and Figures 1–3 indicates that the values of ρ and u and η of ammonium carbonicum in ethanol are more than those of ethanol control for all the potencies (1C to 100C) at each investigated temperature and these values decrease with increase in temperature. The values of ρ and u are maximum at 1C and then decrease significantly in presence of ammonium carbonicum for simple successive dilution to the potency 4C and after that these values remain nearly constant up to potency 60C

and then increase and exhibit a maximum at 70C and thereafter 80C the values remain nearly constant up to 100C (Figures 1 and 2). The values of η are maximum at 1C decrease significantly in presence of ammonium carbonicum for simple successive dilutions from potency 8C and then increase and exhibit a maximum at 10C and thereafter 12C remain nearly constant up to potency 24C and then increase and exhibit a maximum at 26C and then after 30C the values remain nearly constant up to 60C and then increase and exhibit a maximum at 70C and thereafter 80C the values remain nearly constant up to 100C (Figure 3). The observed odd trends in ρ and u at certain potencies, *viz.*, 1C, 4C and 70C; and η at certain potencies, *viz.*, 1C, 8C, 10C, 26C, 28C and 70C indicate that these potencies exhibit different solution structure as compared to other potencies and ethanol control.

A close perusal of Tables 4 and 5 indicate that the values of κ_s and L_f for ammonium carbonicum potencies are less than those of ethanol controls for all the potencies at each investigated temperature and these values increase with increase in temperature, which indicates significant interaction between ammonium carbonicum and ethanol-water molecules. The values of κ_s and L_f are minimum for potency 1C and then increase significantly in presence of ammonium carbonicum for simple successive dilutions up to potency 4C and after that value remain nearly constant up to 60C and then decrease and exhibit a minimum at 70C and thereafter the potency 80C the values remain nearly constant till 100C potency (Tables 4 and 5). These variations in κ_s and L_f are expressed in terms of deviations in isentropic compressibility, $\Delta\kappa_s$ and deviations in intermolecular free length, ΔL_f and are shown graphically in Figures 4 and 5. Figures 4 and 5 indicate that the values of $\Delta\kappa_s$ and ΔL_f are negative and these values are minimum for potency 1C and then increase till potency 4C and after that value remain nearly constant up to 60C and then increase and exhibit a minimum at 70C and thereafter the potency 80C the values remain nearly constant till 100C potency. This indicates that at potencies 1C to 4C and 70C are less compressible than the other potencies, indicating that the potencies 1C to 4C and 70C exhibit more compact solution structure as compared to other potencies and ethanol control. The minimum in $\Delta\kappa_s$ and ΔL_f values at potencies 1C to 4C and 70C indicate that these have most compact solution structure as compared to other potencies, hence, these potencies might have diverse behaviour in terms of properties and efficacy when used in practice.

A close perusal of Figure 6 indicates that the pseudo-Grüneisen parameters, Γ for ammonium carbonicum potencies is more than those of ethanol controls for all the potencies at each investigated temperature and these values increase with increase in temperature, which indicates substantial interaction between ammonium carbonicum and ethanol-water molecules. The variations in Γ is expressed in terms of deviations in pseudo-Grüneisen parameter, $\Delta\Gamma$ and are shown in Figure 6. Figure 6 indicates that $\Delta\Gamma$ values are positive, *i.e.*, $\Delta\Gamma$ values for ammonium carbonicum are more than those of ethanol control. These $\Delta\Gamma$ values are maximum for potency 2C and decrease significantly in presence of ammonium carbonicum for simple successive dilution to potency 4C and thereafter it remains nearly

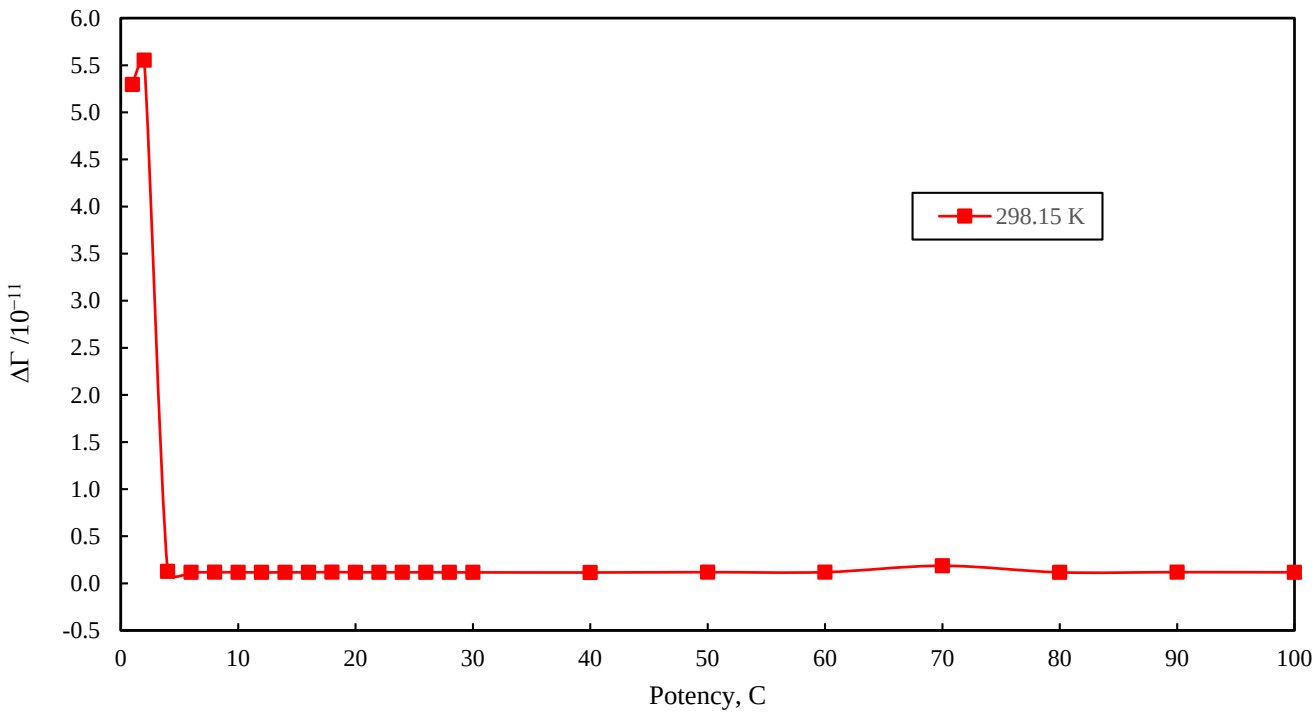

Figure 6. Plots of deviations in acoustic impedance, ΔZ *vs.* potency, *C* of ammonium carbonicum for homoeopathic formulations of ammonium carbonicum at temperatures, 298.15 K.

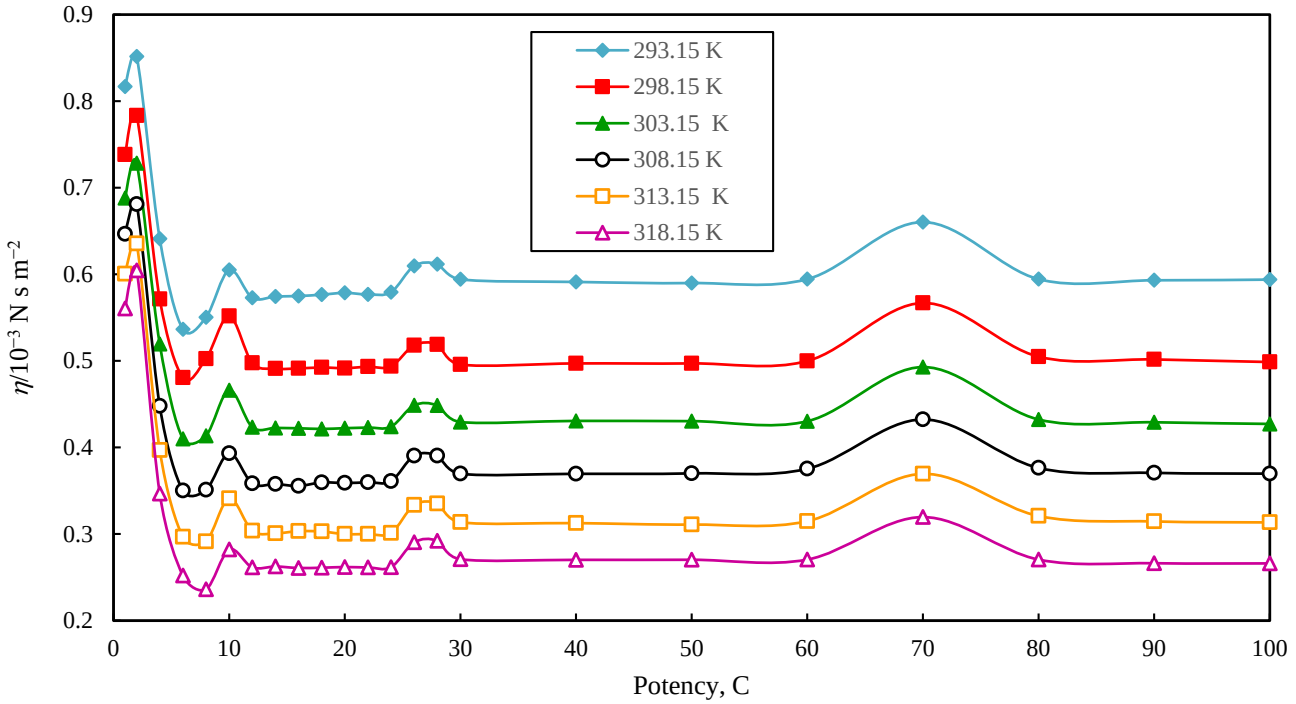

Figure 7. Plots of deviations in viscosity, $\Delta\eta$ *vs.* potency, *C* of ammonium carbonicum for homoeopathic formulations of ammonium carbonicum at different temperatures.

constant up to potency 60C and then again increase and exhibit a maximum at 70C and then after 80C the values remain nearly constant up to 100C. The variations in values of Γ and ΔΓ of these potencies may be due to interaction between ammonium carbonicum and water-ethanol molecules. It is observed that the variations observed in the values of measured properties, ρ and u and η ; and calculated parameters, κ_s, L_f, $\Delta\kappa_s$, ΔL_f and ΔΓ support each other.

A close perusal of Figure. 3 indicates that the viscosities, η of potencies of ammonium carbonicum 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 substantial interaction between ammonium carbonicum and ethanol molecules. These variations in η are expressed in terms of deviations in acoustic impedance, $\Delta\eta$ and are shown in Figure 7. Figure 7 indicates that $\Delta\eta$ values are positive, *i.e.*, η values for ammonium carbonicum are more than those of ethanol control. These $\Delta\eta$ values are maximum for potency 1C and decrease significantly in presence of ammonium carbonicum for simple successive dilutions from potency 8C and then increase and exhibit a maximum at 10C and thereafter 12C remain nearly constant up to potency 24C and then again increase and exhibit a maximum at 26C and then after 30C the values remain nearly constant up to 60C and then after 80C the values remain nearly constant up to 100C. The variations in values of η and $\Delta\eta$ of these potencies may be due interaction between ammonium carbonicum and ethanol molecules.

It has been observed from the analysis of the studied physicochemical parameters, *viz.*, κ_s, L_f, Γ, $\Delta\kappa_s$, ΔL_f, ΔΓ and $\Delta\eta$ that all the potencies exhibit more compact solution structure as compared to pure ethanol control; and the potencies 1C to 4C and 70C exhibit more compact solution structure than the other potencies. The difference in the physicochemical properties of these formulations of ammonium carbonicum in ethanol control (91% ethanol in water) clearly indicate that the presence of medicine results in significant structural modifications in solution for all the potencies and found more dominant in certain potencies. The results can be qualitatively discussed in terms of interactions/physicochemical behaviour of these extremely dilute homoeopathic formulations of ammonium carbonicum in ethanol controls. The key factors affecting the solution structure are nature of solute, degree of dilution, presence of ammonium carbonicum and succussion.

It is well known that hydrogen bonding is one of the most important weak interactions between molecules in solutions leading to the formation of well-defined molecular aggregates, also termed as supermolecular water structure, *i.e.*, a self-consistent interpretation due to formation of dissipative structures.[3,17] It had been reported that successive dilution with succussion permanently alters the physicochemical properties of the solution.[35] The succussion process triggers the formation of dissipative structures and these dissipative structures are affected by presence of ethanol and medicine molecules[35] (ammonium carbonate dissociated into NH_4^+ and CO_3^{2-} ions in the present study).

The results can be interpreted by considering the interactions that can take place between NH_4^+ and CO_3^{2-} ions and the hypothesized molecular aggregates of water-ethanol molecules, *i.e.*, dissipative structures.[16] A qualitative comparison between various potencies can be considered due to the nature of driving

force that leads to formation of complexes between NH_4^+ and CO_3^{2-} ions and dissipative structures of water molecules.[19,36] This driving force is provided by the succussion process used in preparing different potencies of homoeopathic medicine and in which a large amount of mechanical energy (~404.3 Newton-meter for 10 strokes)[37] is transferred. This transfer of energy due to successive dilution and succussion is responsible for different/anomalous behaviour of ammonium carbonicum of different potencies. It has also been observed by others[5,38] that same medicine of different potency and different medicines of same potency show different behaviour due to vehicle-molecule structure (ethanol-water system) generated by potentization process of homoeopathy.

These physicochemical data of homoeopathic medicines can be used as reference to standardize/test the purity/accuracy of potencies of commercially available homeopathic medicines. Further studies on the variation of these physicochemical properties with temperature/time may reflect the effect of temperature/time on the shelf life of these homoeopathic medicines. At present the homoeopathic medicines are commercially used without any expiry date.

CONCLUSION

The densities, ultrasonic speeds and viscosities of ethanol control, 22 formulations of ammonium carbonicum in ethanol control are measured for potencies from 1C to 100C (with an interval of 2C up to 30C and then of 10C up to 100C) at six different temperatures and atmospheric pressure. Using these experimental data, various physicochemical parameters, *viz.*, κ_s, L_f, Γ, $\Delta\kappa_s$, ΔL_f , $\Delta\Gamma$ and $\Delta\eta$ have been calculated. The results have been qualitatively discussed in terms of interactions/ physicochemical behaviour of these extremely dilute homoeopathic formulations of ammonium carbonicum in ethanol. It is found that the interactions can take place between NH_4^+ and CO_3^{2-} ions and the molecular aggregates of water-ethanol molecules, *i.e.*, dissipative structures. The potencies 1C to 4C and 70C exhibit more compact solution structure as compared to other potencies and ethanol control. Hence, these potencies may have different behaviour in terms of properties and efficacy when used in practice. It can be qualitatively concluded that even in extreme dilutions (70C) the molecules of ammonium carbonicum may be present in these homoeopathic formulations, however it needs to be confirmed from other more precise spectroscopic techniques.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interest.

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