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Investigation of the physicochemical behaviour of *Acidum citricum* homoeopathic dilutions at temperatures from 293.15 K to 318.15 K by using volumetric, acoustic and viscometric methods

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Abstract

Background: The physicochemical properties of homoeopathic medicines have been gaining importance in elucidating the physicochemical behaviour (interactions) and, hence in establishing the presence of medicine at ultra-high dilutions. **Objective:** The present study is aimed to reconnoiter the existence of drug in ultra-diluted medicines and evidence regarding the physicochemical behaviour and prevailing interactions in the dilutions of *Acidum citricum*. **Materials and Methods:** The densities (ρ), ultrasonic speeds (u) and viscosities (η) of pure ethanol control (91% ethanol in water) and 33 dilutions of *Acidum citricum* of potencies across 1C to 200C at temperatures from 293.15 K to 318.15 K and at atmospheric pressure. **Results:** The measured data are used to compute the physicochemical parameters, viz., isentropic compressibility (κ_s), intermolecular free lengths (L_f), acoustic impedance (Z) relative association (R_A), relaxation time (τ), ultrasonic absorption (α/ρ^2) pseudo-Grüneisen parameter (Γ), deviations in isentropic compressibility, ($\Delta\kappa_s$), deviations in intermolecular free length, (ΔL_f), deviations in acoustic impedance (ΔZ), deviations in viscosity ($\Delta\eta$) and deviations in pseudo-Grüneisen parameter ($\Delta\Gamma$). These parameters specified diverse trends at certain potencies of these homoeopathic dilutions. **Conclusion:** The results are qualitatively interpreted by means of prevailing interactions of these *Acidum citricum* dilutions. It is observed that the interactions may take place among H^+ and citrate ions and the molecular aggregates of water-ethanol, *i.e.*, dissipative structures. The results specified that even in high-dilutions (50C, 110C, 150C and 160C) the molecules of medicine may still exist in these homoeopathic formulations.

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ORIGINAL ARTICLE

Investigation of the physicochemical behaviour of *Acidum citricum* homoeopathic dilutions at temperatures from 293.15 K to 318.15 K by using volumetric, acoustic and viscometric methods

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ABSTRACT

Background: The physicochemical properties of homoeopathic medicines have been gaining importance in elucidating the physicochemical behaviour (interactions) and, hence in establishing the presence of medicine at ultra-high dilutions. **Objective:** The present study is aimed to reconnoiter the existence of drug in ultra-diluted medicines and evidence regarding the physicochemical behaviour and prevailing interactions in the dilutions of *Acidum citricum*. **Materials and Methods:** The densities (ρ), ultrasonic speeds (u) and viscosities (η) of pure ethanol control (91% ethanol in water) and 33 dilutions of *Acidum citricum* of potencies across 1C to 200C at temperatures from 293.15 K to 318.15 K and at atmospheric pressure. **Results:** The measured data are used to compute the physicochemical parameters, viz., isentropic compressibility (κ_s), intermolecular free lengths (L_f), acoustic impedance (Z) relative association (R_A), relaxation time (τ), ultrasonic absorption (α/f^2) pseudo-Grüneisen parameter (Γ), deviations in isentropic compressibility, ($\Delta\kappa_s$), deviations in intermolecular free length, (ΔL_f), deviations in acoustic impedance (ΔZ), deviations in viscosity ($\Delta\eta$) and deviations in pseudo-Grüneisen parameter ($\Delta\Gamma$). These parameters specified diverse trends at certain potencies of these homoeopathic dilutions. **Conclusion:** The results are qualitatively interpreted by means of prevailing interactions of these *Acidum citricum* dilutions. It is observed that the interactions may take place among H^+ and citrate ions and the molecular aggregates of water-ethanol, i.e., dissipative structures. The results specified that even in high-dilutions (50C, 110C, 150C and 160C) the molecules of medicine may still exist in these homoeopathic formulations.

Keywords: *Acidum citricum*, Density, Homoeopathic medicines, Hydrogen bonding, Ultrasonic speed, Viscosity

Introduction

Homoeopathy is one of the most extensively practiced alternative medicine with holistic approach. The potency of a homoeopathic medicine increases with serial dilution followed by succussion (potentisation), which poses two main challenges to the homoeopathic community: (i) their preparation comprises ultra dilutions, which raises questions concern-

ing the existence of active medicine molecules at such ultra-dilutions, and (ii) effectiveness of Homoeopathy is well supported by the reported evidences; however, there are disputes regarding implausibility in their biological activity wherein the dilution of source

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drug exceeds Avagadro's limit, *i.e.*, the ultra-diluted medicine formulation can be considered like the solvent.^{1,2} There have been few researches to investigate the existence of drug in ultra-high dilutions¹⁻⁷ and their mechanism of action, but the query is still unanswered.

In addition to the past research on the physicochemical behaviour of ultra-diluted homoeopathic formulations,⁸⁻¹¹ the results of the physicochemical study on the behaviour of homoeopathic potencies of *Acidum citricum* are reported here. *Acidum citricum* is a remedy prepared from potentiation of citric acid. It is useful for the treatment of various health problems such as scurvy, chronic rheumatism and hemorrhages.¹² This investigation of the homoeopathic dilutions of *Acidum citricum* was conducted to understand their physicochemical behaviour comparison to the vehicle, *i.e.*, alcohol.

The physicochemical properties and their derived parameters of electrolytes/amino acids/carbohydrates/drugs in aqueous and mixed-aqueous solvents have been utilised in elucidating the existing interactions, which assist in understanding of solute-solvation and hydration behaviour of solute in these solvents.¹³⁻¹⁹ Since the homoeopathic medicines are ultra-diluted solutions, their physical properties, *viz.*, density, ultrasonic speed and viscosity can be measured at various potencies and temperature. The physicochemical parameters calculated using these measured data provide significant evidence concerning their physicochemical behaviour and prevailing interactions therein. There have been a few physicochemical studies on extremely diluted solutions of inorganic salts²⁰⁻²⁴ and homoeopathic medicines²⁵⁻²⁷ by using conductivity and pH-metric measurements reported in the literature. These studies furnished significant information on the physicochemical behaviour of these ultra-diluted solutions. To the best of our knowledge, very few physicochemical studies on homoeopathic formulations using volumetric, acoustic and viscometric have been reported in the literature.^{4,8-12,27-29}

In the present investigation, the densities (ρ), ultrasonic speeds (u) and viscosities (η) of pure ethanol control (91% ethanol in water) and 33 dilutions of *Acidum citricum* of potencies across 1C to 200C (having intervals of 2C up to 30C, and thereafter having intervals of 10C up to 200C) at 293.15, 298.15, 303.15, 308.15, 313.15 and 318.15 K and at atmospheric pressure. From these measured data, the isentropic compressibilities (κ_s), intermolecular free lengths (L_f), acoustic impedance (Z) relative association (R_A), relaxation time (τ), ultrasonic absorption (α/f^2) pseudo-Grüneisen parameter (Γ), deviations in isentropic compressibility, ($\Delta\kappa_s$), deviations in

intermolecular free length, (ΔL_f), deviations in acoustic impedance (ΔZ), deviations in viscosity ($\Delta\eta$) and deviations in pseudo-Grüneisen parameter ($\Delta\Gamma$) have been calculated. The potency and temperature dependence of these parameters are interpreted qualitatively in relation to interactions/physicochemical conduct of these *Acidum citricum* dilutions.

Materials and methods

Materials

The citric acid (s.d. fine chemicals, India, mass fraction purity > 0.99) was used for preparation of 33 potencies of *Acidum citricum*. The ethanol control (91% ethanol in water) and succussed ethanol control (91% ethanol in water) was prepared by using the ethanol (E. Merck, India, mass fraction purity > 0.995) and triple distilled water.

Preparation of medicines

Various potencies of *Acidum citricum* used in the investigation were prepared in the laboratory of Dyal Singh College, New Delhi, India, by using the method introduced by Dr. Hahnemann (1755-1843) using centesimal scale, as described in the Homoeopathic Pharmacopoeia of India.³⁰ The potencies across 1C to 200C (having intervals of 2C upto 30C, and thereafter having intervals of 10C up to 200C) have been prepared. The homoeopathic formulations of *Acidum citricum* were prepared by successive dilution of 1:100 followed by ten succussions for each potency. The ethanol control samples at each potency were prepared by applying an equivalent number of succussions as given for the respective potency. The succussions for preparing various potencies were performed by using a hammer type mini-potentizer (A.R.P. Industries, Sahibabad, Uttar Pradesh, India). The prepared potencies were stored in amber glass bottles and were kept in cool, dark place, away from direct sunlight, strong-smelling substances and mother tinctures, in properly stoppered amber bottles which were not fully filled.

Objectives

The aim of this study was to carry out the measurements of the densities, ultrasonic speeds and viscosities of ethanol control samples, and various potencies of *Acidum citricum*. These data were then used to compute several physicochemical parameters, which can be used to reconnoiter the existence of drug in ultra diluted medicines and provide evidence regarding

Table 1. The densities ($\rho/\text{kg m}^{-3}$) of ethanol control and *Acidum citricum* potencies (C) at temperatures (293.15–318.15) K and atmospheric pressure^a

Potency (C)	T/K					
	293.15	298.15	303.15	308.15	313.15	318.15
0	808.248	804.277	800.155	796.005	791.621	787.436
1	826.816	822.483	818.080	813.626	809.113	804.541
2	818.998	814.660	810.266	805.819	801.316	796.754
4	821.651	817.309	812.908	808.452	803.938	799.365
6	818.088	813.745	809.348	804.894	800.386	795.817
8	819.980	815.645	811.247	806.796	802.288	797.720
10	820.054	815.716	811.315	806.862	802.353	797.780
12	820.099	815.759	811.356	806.901	802.390	797.814
14	820.101	815.921	811.509	807.074	802.579	798.024
16	820.340	815.998	811.601	807.155	802.648	798.079
18	820.432	816.082	811.684	807.231	802.721	798.151
20	820.571	816.130	811.728	807.271	802.766	798.286
22	820.653	816.306	811.904	807.450	802.849	798.567
24	820.811	816.469	812.069	807.616	803.105	798.754
26	821.005	816.664	812.261	807.806	803.294	798.820
28	819.236	814.895	810.497	806.044	801.535	797.043
30	822.914	818.573	814.618	809.709	805.192	800.618
40	827.057	822.410	818.006	813.547	809.029	804.453
50	829.969	825.624	821.221	816.764	812.247	807.648
60	826.066	821.713	817.309	812.850	808.334	803.756
70	826.115	821.770	817.365	812.906	808.388	803.821
80	826.429	822.075	817.667	813.206	808.688	804.115
90	826.415	822.067	817.665	813.207	808.690	804.110
100	826.041	821.700	817.293	812.831	808.312	803.727
110	829.547	825.195	820.797	816.341	811.822	807.243
120	826.213	821.859	817.456	813.099	808.580	803.902
130	826.116	821.776	817.370	812.909	808.391	803.813
140	826.285	821.937	817.533	813.075	808.557	803.981
150	830.827	826.491	822.091	817.633	813.116	808.538
160	829.271	824.932	820.528	816.067	811.548	806.969
170	826.351	822.010	817.607	813.148	808.629	804.052
180	826.161	821.825	817.422	812.964	808.449	803.871
190	826.213	821.867	817.460	813.002	808.489	803.909
200	826.390	822.017	817.714	813.258	808.741	804.162

^aStandard uncertainties σ are $\sigma(T) = 0.01$ K, $\sigma(\rho) = 0.05$ kg m⁻³ and $\sigma(p) = 5.0$ kPa (at 68% confidence interval).

the physicochemical behaviour and prevailing interactions in ultra-dilutions of *Acidum citricum*.

Methods

Density and ultrasonic speed measurements

The densities and ultrasonic speeds of the samples were measured by using Density and Sound Analyzer

(DSA 5000M, Anton Paar, Austria). The oscillating U-tube principle was used in density measurements while the propagation time technique was utilised in the ultrasonic speed measurement. This instrument was equipped with a combination of density and ultrasonic cells, which provides the reproducibility of $\pm 1 \cdot 10^{-3}$ kg·m⁻³ and $\pm 1 \cdot 10^{-2}$ m·s⁻¹ for density and ultrasonic speed, respectively. The temperature

Table 1a. The densities, ρ /(kg m⁻³) of succussed ethanol control with succussions equivalent to each potency (C) at temperatures (293.15–318.15) K and atmospheric pressure^a

Potency (C)	T/K					
	293.15	298.15	303.15	308.15	313.15	318.15
0	808.248	804.277	800.155	796.005	791.621	787.436
1	825.443	821.101	816.759	812.417	808.075	803.733
2	825.451	821.111	816.771	812.431	808.091	803.751
4	825.446	821.090	816.734	812.378	808.022	803.666
6	825.436	821.082	816.728	812.374	808.020	803.666
8	825.441	821.115	816.789	812.463	808.137	803.811
10	825.460	821.115	816.770	812.425	808.080	803.735
12	825.456	821.109	816.762	812.415	808.068	803.721
14	825.454	821.099	816.744	812.389	808.034	803.679
16	825.448	821.107	816.766	812.425	808.084	803.743
18	825.461	821.102	816.743	812.384	808.025	803.666
20	825.455	821.113	816.771	812.429	808.087	803.745
22	825.451	821.095	816.739	812.383	808.027	803.671
24	825.439	821.085	816.731	812.377	808.023	803.669
26	825.461	821.115	816.769	812.423	808.077	803.731
28	825.444	821.089	816.734	812.379	808.024	803.669
30	825.494	821.147	816.800	812.453	808.106	803.759
40	825.453	821.110	816.767	812.424	808.081	803.738
50	825.449	821.108	816.767	812.426	808.085	803.744
60	825.444	821.085	816.726	812.367	808.008	803.649
70	825.446	821.106	816.766	812.426	808.086	803.746
80	825.451	821.095	816.739	812.383	808.027	803.671
90	825.456	821.101	816.746	812.391	808.036	803.681
100	825.455	821.104	816.753	812.402	808.051	803.700
110	825.448	821.089	816.730	812.371	808.012	803.653
120	825.453	821.111	816.769	812.427	808.085	803.743
130	825.462	821.106	816.750	812.394	808.038	803.682
140	825.455	821.101	816.747	812.393	808.039	803.685
150	825.449	821.103	816.757	812.411	808.065	803.719
160	825.441	821.086	816.731	812.376	808.021	803.666
170	825.463	821.116	816.769	812.422	808.075	803.728
180	825.454	821.111	816.768	812.425	808.082	803.739
190	825.464	821.118	816.772	812.426	808.080	803.734
200	825.451	821.102	816.753	812.404	808.055	803.706

^aStandard uncertainties σ are $\sigma(T) = 0.01$ K, $\sigma(\rho) = 0.05$ kg m⁻³ and $\sigma(p) = 5.0$ kPa (at 68% confidence interval).

of the density and ultrasonic cells was maintained within ± 0.01 K by a Peltier type thermostat fitted in the instrument. The calibration of the instrument was done with triply distilled degassed water and dry air at atmospheric pressure.^{10,11} The frequency of ultrasonic waves was kept at 3 MHz in the ultrasonic speed measurements. The standard uncertainties as-

sociated with the measurements of density, ultrasonic speed and temperature were within ± 0.05 kg·m⁻³, ± 0.5 m·s⁻¹ and ± 0.01 K, respectively.

Viscosity measurements

The viscosities of the samples were measured by using a microviscometer (Lovis 2000M, Anton Paar,

Table 2. The ultrasonic speeds ($u/m\ s^{-1}$) of ethanol control and *Acidum citricum* potencies (C) at temperatures (293.15–318.15) K and atmospheric pressure^a

Potency (C)	T/K					
	293.15	298.15	303.15	308.15	313.15	318.15
0	1195.93	1179.76	1163.54	1147.33	1131.38	1114.91
1	1254.83	1238.33	1221.70	1204.97	1188.22	1171.36
2	1241.71	1225.36	1208.56	1191.62	1174.68	1157.72
4	1249.20	1232.64	1215.79	1198.86	1181.91	1164.93
6	1240.66	1223.98	1207.06	1190.09	1173.09	1156.08
8	1245.33	1228.75	1212.00	1195.14	1178.31	1161.37
10	1244.90	1228.39	1211.47	1194.50	1177.73	1160.51
12	1244.75	1228.13	1211.25	1194.43	1177.53	1160.52
14	1245.32	1228.61	1211.57	1194.86	1178.14	1161.10
16	1245.77	1229.33	1212.47	1195.64	1178.82	1161.82
18	1246.43	1229.86	1213.01	1196.08	1179.10	1162.11
20	1246.49	1230.28	1213.35	1196.35	1179.42	1162.35
22	1246.55	1229.94	1213.02	1196.15	1179.39	1162.09
24	1246.76	1230.36	1213.45	1196.49	1179.72	1162.53
26	1247.27	1230.85	1213.96	1197.02	1180.05	1163.06
28	1240.65	1224.24	1207.33	1190.36	1173.37	1156.36
30	1252.25	1235.93	1218.55	1202.27	1185.35	1168.40
40	1262.07	1245.68	1228.92	1212.07	1195.21	1178.29
50	1270.25	1254.03	1237.42	1220.70	1203.91	1187.08
60	1260.03	1243.87	1227.10	1210.27	1193.38	1176.68
70	1261.09	1244.69	1228.17	1211.57	1194.47	1177.68
80	1261.08	1244.92	1228.24	1211.48	1194.59	1177.70
90	1261.09	1244.78	1228.02	1211.17	1194.30	1177.40
100	1260.20	1243.92	1227.19	1210.34	1193.47	1176.86
110	1269.56	1253.20	1236.51	1219.73	1202.89	1186.02
120	1261.99	1245.56	1228.82	1211.97	1194.97	1178.25
130	1260.33	1243.98	1227.18	1210.32	1193.45	1176.54
140	1260.64	1244.34	1227.62	1210.84	1193.97	1177.26
150	1272.15	1256.13	1239.64	1222.97	1206.21	1189.41
160	1268.35	1252.12	1235.41	1218.63	1201.80	1184.94
170	1261.02	1244.72	1227.95	1211.11	1194.23	1177.54
180	1260.48	1244.37	1227.69	1210.90	1194.04	1177.15
190	1260.71	1244.31	1227.51	1210.68	1193.77	1176.86
200	1260.45	1244.05	1227.31	1210.49	1193.62	1176.73

^aStandard uncertainties σ are $\sigma(T) = 0.01\ K$, $\sigma(u) = 0.5\ m\ s^{-1}$ and $\sigma(p) = 5.0\ kPa$ (at 68% confidence interval).

Austria) at temperatures, (293.15 – 318.15) K, and at atmospheric pressure. The instrument utilises rolling ball principle for the measurement of viscosity, having a calibrated glass capillary with a steel ball as supplied by the manufacturer. The calibration of capillary was performed by means of standard viscosity

fluids provided by the manufacturer. The temperature of the sample in capillary was maintained within $\pm 0.02\ K$ by means of a Peltier thermostat fitted in the instrument. The standard uncertainties in the measurements of viscosity measurements and temperature were within $\pm 0.5\%$ and $\pm 0.02\ K$, respectively.

Table 2a. The ultrasonic speeds ($u/m\ s^{-1}$) of succussed ethanol with succussions equivalent to each potency (C) at temperatures (293.15–318.15) K and atmospheric pressure^a

Potency (C)	T/K					
	293.15	298.15	303.15	308.15	313.15	318.15
0	1195.93	1179.76	1163.54	1147.33	1131.38	1114.91
1	1235.42	1218.51	1202.83	1186.72	1169.16	1152.54
2	1235.55	1218.22	1202.44	1186.58	1169.69	1152.75
4	1235.29	1218.08	1202.37	1186.62	1169.81	1152.94
6	1235.62	1218.23	1202.12	1186.40	1169.58	1152.68
8	1235.46	1218.66	1202.61	1186.04	1169.47	1152.86
10	1235.39	1218.53	1202.23	1186.23	1169.72	1152.52
12	1235.61	1218.66	1202.17	1186.27	1169.35	1152.39
14	1235.42	1218.32	1202.43	1186.45	1169.22	1152.64
16	1235.53	1218.27	1202.49	1186.65	1169.76	1152.84
18	1235.41	1218.22	1202.30	1186.82	1169.52	1152.42
20	1235.62	1218.63	1202.15	1186.49	1169.65	1152.72
22	1235.56	1218.58	1202.30	1186.69	1169.52	1152.54
24	1235.58	1218.43	1202.22	1186.57	1169.62	1152.68
26	1235.52	1218.50	1202.17	1186.42	1169.33	1152.74
28	1235.55	1218.12	1202.32	1186.46	1169.21	1152.19
30	1235.44	1218.57	1202.73	1186.62	1169.26	1152.44
40	1235.52	1218.25	1202.40	1186.48	1169.59	1152.70
50	1235.26	1218.12	1202.34	1186.52	1169.71	1152.84
60	1235.58	1218.27	1202.17	1186.44	1169.48	1152.58
70	1235.43	1218.60	1202.66	1186.14	1169.40	1152.76
80	1235.36	1218.45	1202.27	1186.33	1169.62	1152.42
90	1235.56	1218.57	1202.32	1186.37	1169.30	1152.49
100	1235.45	1218.30	1202.40	1186.48	1169.32	1152.44
110	1235.50	1218.22	1202.43	1186.45	1169.66	1152.74
120	1235.43	1218.35	1202.35	1186.72	1169.42	1152.52
130	1235.22	1218.67	1202.25	1186.59	1169.55	1152.62
140	1235.60	1218.68	1202.33	1186.49	1169.42	1152.64
150	1235.77	1218.53	1202.28	1186.53	1169.52	1152.38
160	1235.65	1218.56	1202.23	1186.46	1169.43	1152.54
170	1235.55	1218.22	1202.36	1186.40	1169.31	1152.39
180	1235.46	1218.38	1202.48	1186.35	1169.42	1152.46
190	1235.49	1218.37	1202.39	1186.55	1169.66	1152.73
200	1235.47	1218.32	1202.31	1186.62	1169.37	1152.42

^aStandard uncertainties σ are $\sigma(T) = 0.01\ K$, $\sigma(u) = 0.5\ m\ s^{-1}$ and $\sigma(p) = 5.0\ kPa$ (at 68% confidence interval).

Results

The measured values of densities (ρ), ultrasonic speeds (u) and viscosities (η) of the unsuccussed ethanol control, succussed ethanol controls and the *Acidum citricum* formulations as function of potency (in centesimal) at different temperatures are recorded

in Tables 1, 2 and 3, and are displayed graphically in Figs. 1, 2 and 3, respectively.

The isentropic compressibility (κ_s), intermolecular free length (L_f), acoustic impedance (Z), relative association (R_A) relaxation time (τ), ultrasonic absorption (α/f^2), and pseudo-Grüneisen parameter (Γ) of the *acidum citricum* potencies have been evaluated by

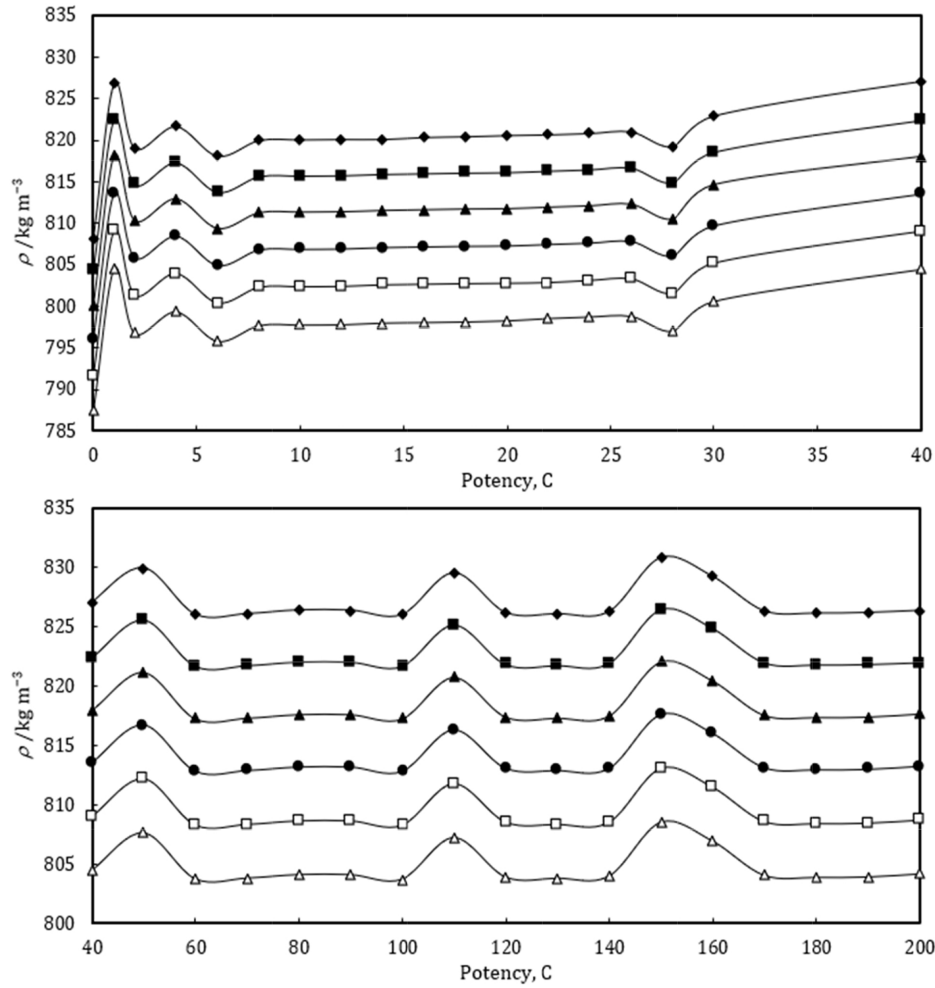


Fig. 1. Variations of densities, ρ vs. potency, C of *Acidum citricum* for dilutions of *Acidum citricum* at temperatures, 293.15 K, \blacklozenge ; 298.15 K, \blacksquare ; and 303.15 K, \blacktriangle ; 308.15 K, \bullet ; 313.15 K, \square ; and 318.15 K, \triangle

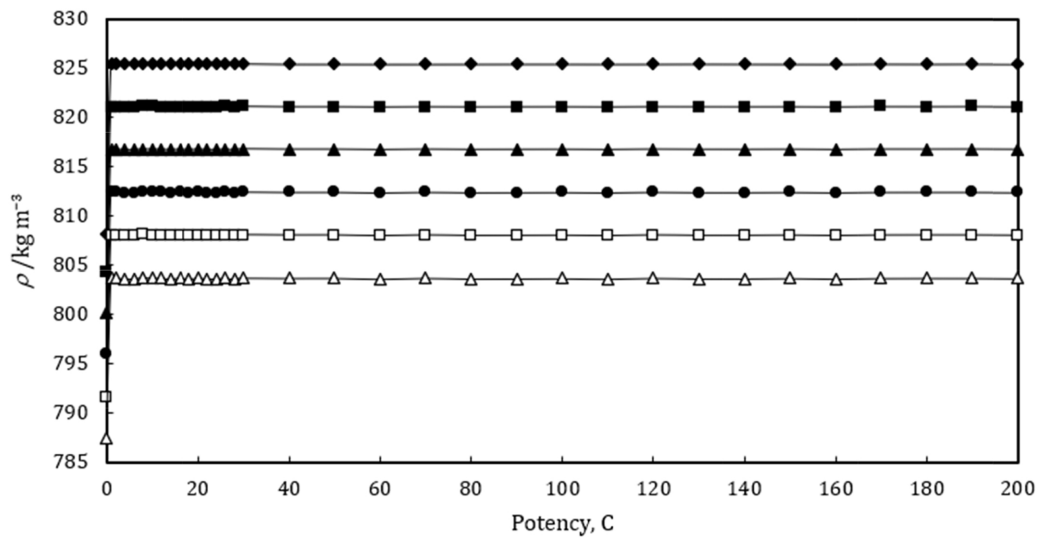


Fig. 1a. Variations of densities, ρ of succussed ethanol with succussions equivalent to each potency, C at temperatures, 293.15 K, \blacklozenge ; 298.15 K, \blacksquare ; and 303.15 K, \blacktriangle ; 308.15 K, \bullet ; 313.15 K, \square ; and 318.15 K, \triangle

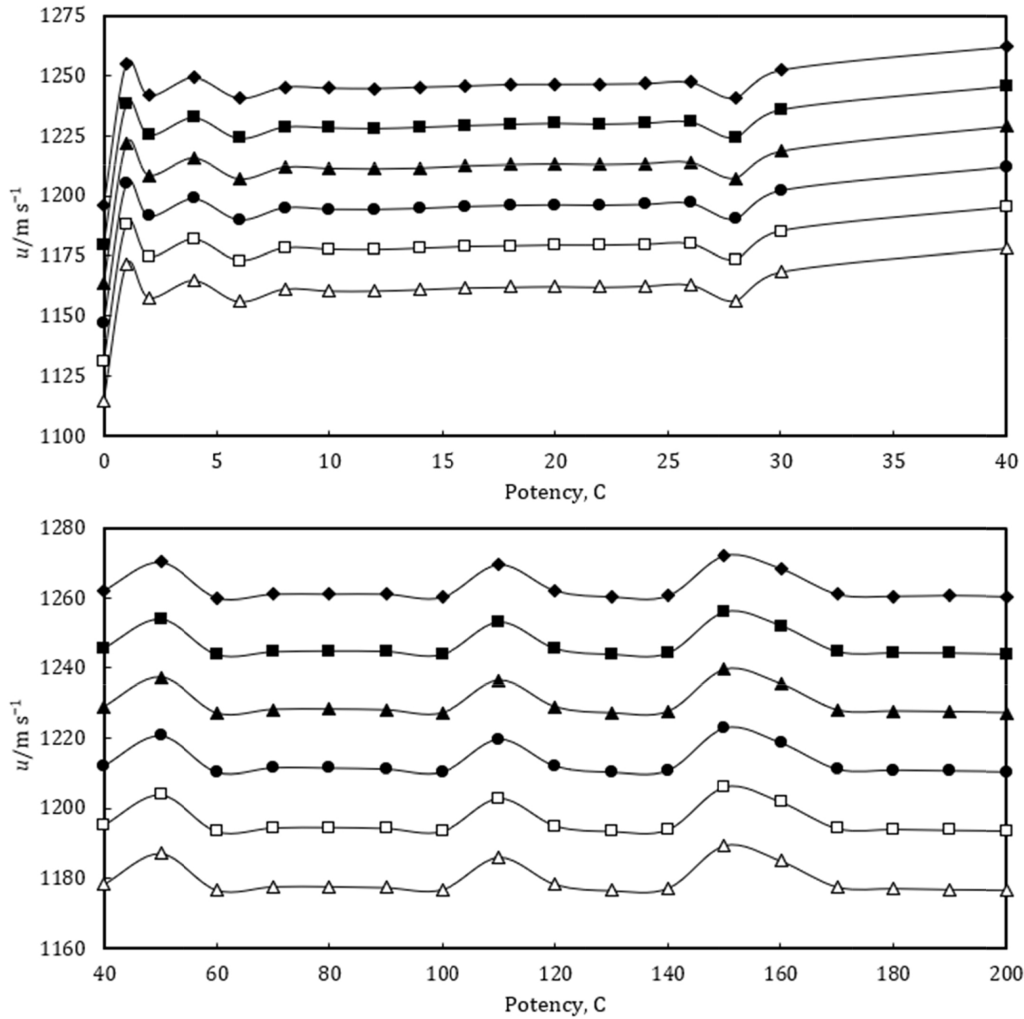


Fig. 2. Variations of ultrasonic speeds, u vs. potency, C of *Acidum citricum* for dilutions of *Acidum citricum* at temperatures, 293.15 K, \blacklozenge ; 298.15 K, \blacksquare ; and 303.15 K, \blacktriangle ; 308.15 K, \bullet ; 313.15 K, \square ; and 318.15 K, \triangle

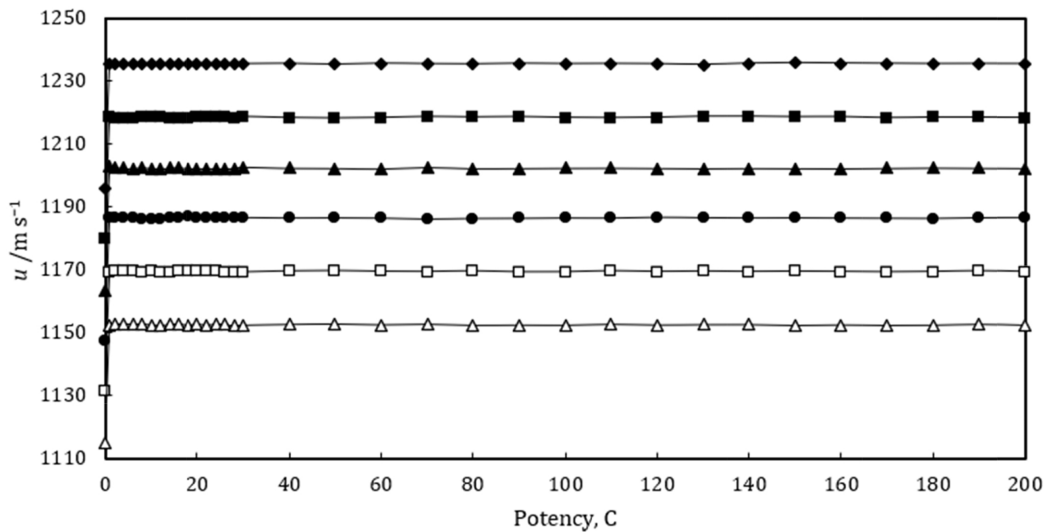


Fig. 2a. Variations of ultrasonic speeds, u of succussed ethanol control with succussions equivalent to each potency, C at temperatures, 293.15 K, \blacklozenge ; 298.15 K, \blacksquare ; and 303.15 K, \blacktriangle ; 308.15 K, \bullet ; 313.15 K, \square ; and 318.15 K, \triangle

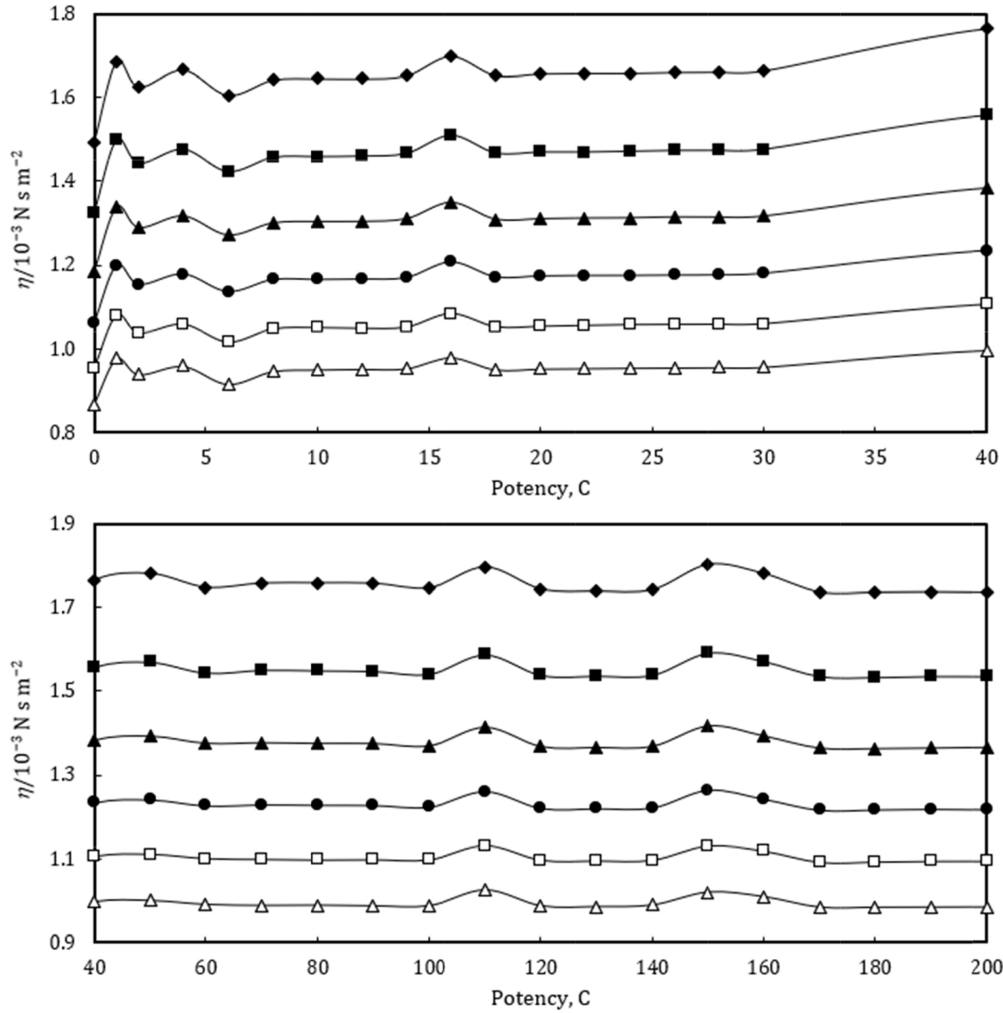


Fig. 3. Variations of viscosities, η vs. potency, C of *Acidum citricum* for dilutions of *Acidum citricum* at temperatures, 293.15 K, \blacklozenge ; 298.15 K, \blacksquare ; and 303.15 K, \blacktriangle ; 308.15 K, \bullet ; 313.15 K, \square ; and 318.15 K, \triangle

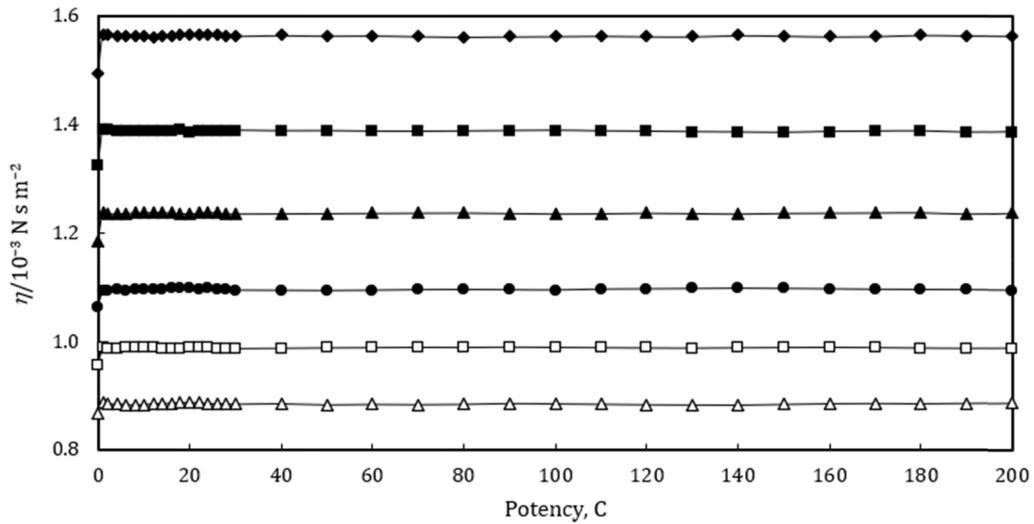


Fig. 3a. Variations of viscosities, η of succussed ethanol control with succussions equivalent to each potency, C at temperatures, 293.15 K, \blacklozenge ; 298.15 K, \blacksquare ; and 303.15 K, \blacktriangle ; 308.15 K, \bullet ; 313.15 K, \square ; and 318.15 K, \triangle

means of the relations^{25,31-36}

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

$$L_f = K'/u\rho^{1/2} \tag{2}$$

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

$$R_A = (\rho/\rho_0)(u_0/u)^{1/3} \tag{4}$$

$$\tau = (4/3)/\kappa_s\eta \tag{5}$$

$$(\alpha/f^2) = 8\pi^2\eta/3\rho u^3 \tag{6}$$

$$\Gamma = \alpha_p/\kappa_T \tag{7}$$

where K' is temperature dependent constant [= (93.875 + 0.375 T) × 10⁻⁸]; T is the temperature in Kelvin; ρ_0 and u_0 are the density and ultrasonic speed of the ethanol control, respectively; α_p is the

isobaric coefficient of thermal expansion and κ_T is the isothermal compressibility. The values of α_p and κ_T are obtained by means of the relations^{36,37}

$$\alpha_p = (-1/\rho)(\partial\rho/\partial T)_p \tag{8}$$

$$\kappa_T = (1.71 \times 10^{-3}) / (T^{4/9}u^2\rho^{4/3}) \tag{9}$$

The values of κ_s , L_f , Z , R_A , τ , (α/f^2) and Γ are given in Tables 4 to 10, respectively.

The deviations in κ_s , L_f , Z , η and Γ of ethanol control due to addition of *Acidum citricum* with dilution and succussion are signified by the deviation in the values of these parameters, i.e., the deviations in isentropic compressibility ($\Delta\kappa_s$), deviations in intermolecular free length, (ΔL_f), deviations in acoustic impedance (ΔZ), deviations in pseudo-Grüneisen parameter ($\Delta\Gamma$) and deviations in viscosity ($\Delta\eta$) have been evaluated by means of the

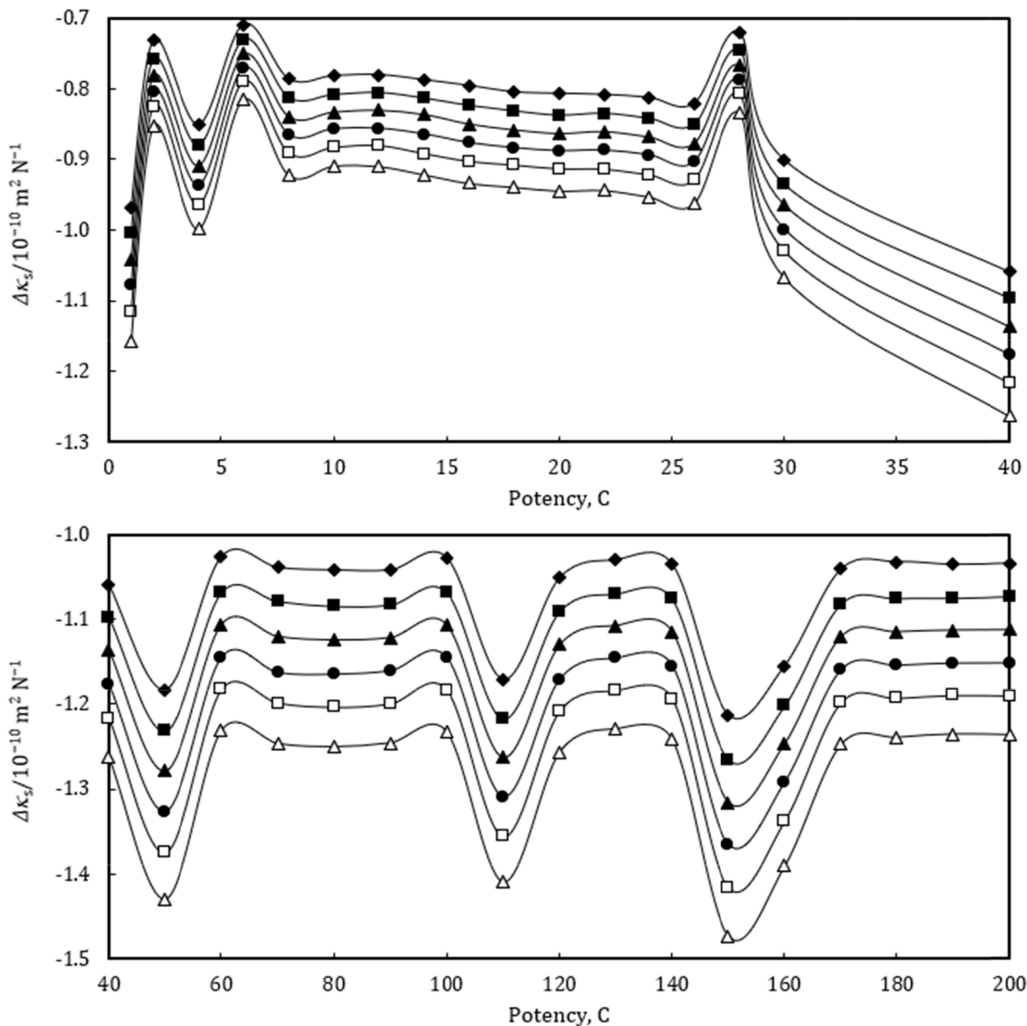


Fig. 4. Variations of deviations in isentropic compressibilities, $\Delta\kappa_s$ vs. potency, C of *Acidum citricum* for dilutions of *Acidum citricum* at temperatures, 293.15 K, \blacklozenge ; 298.15 K, \blacksquare ; and 303.15 K, \blacktriangle ; 308.15 K, \bullet ; 313.15 K, \square ; and 318.15 K, \triangle

Table 3. The viscosities ($\eta/10^{-3}$ N s m⁻²) of ethanol control and *Acidum citricum* potencies (C) at temperatures (293.15–318.15) K and atmospheric pressure^a

Potency (C)	T/K					
	293.15	298.15	303.15	308.15	313.15	318.15
0	1.4935	1.3239	1.1844	1.0616	0.9548	0.8677
1	1.6862	1.4984	1.3373	1.1998	1.0809	0.9764
2	1.6252	1.4426	1.2884	1.1535	1.0383	0.9389
4	1.6671	1.4766	1.3164	1.1788	1.0594	0.9588
6	1.6061	1.4229	1.2710	1.1365	1.0165	0.9158
8	1.6414	1.4557	1.3005	1.1623	1.0448	0.9463
10	1.6438	1.4587	1.3032	1.1665	1.0481	0.9503
12	1.6453	1.4599	1.3034	1.1667	1.0480	0.9502
14	1.6528	1.4665	1.3101	1.1703	1.0524	0.9532
16	1.6997	1.5101	1.3488	1.2076	1.0846	0.9792
18	1.6535	1.4675	1.3096	1.1727	1.0542	0.9524
20	1.6574	1.4708	1.3129	1.1754	1.0552	0.9525
22	1.6585	1.4717	1.3135	1.1750	1.0553	0.9534
24	1.6584	1.4726	1.3142	1.1751	1.0555	0.9544
26	1.6597	1.4725	1.3152	1.1759	1.0552	0.9550
28	1.6603	1.4737	1.3152	1.1765	1.0559	0.9557
30	1.6632	1.4761	1.3161	1.1803	1.0592	0.9566
40	1.7655	1.5576	1.3827	1.2343	1.1061	0.9968
50	1.7809	1.5698	1.3926	1.2411	1.1112	0.9994
60	1.7479	1.5439	1.3756	1.2268	1.0987	0.9881
70	1.7580	1.5510	1.3771	1.2283	1.0988	0.9878
80	1.7584	1.5497	1.3754	1.2284	1.0973	0.9884
90	1.7576	1.5481	1.3752	1.2280	1.0980	0.9888
100	1.7469	1.5415	1.3698	1.2241	1.0979	0.9872
110	1.7966	1.5876	1.4138	1.2612	1.1322	1.0248
120	1.7436	1.5385	1.3678	1.2207	1.0967	0.9867
130	1.7369	1.5349	1.3638	1.2182	1.0931	0.9835
140	1.7426	1.5392	1.3687	1.2222	1.0968	0.9859
150	1.8029	1.5921	1.4172	1.2647	1.1317	1.0199
160	1.7804	1.5737	1.3983	1.2477	1.1184	1.0086
170	1.7362	1.5357	1.3671	1.2203	1.0914	0.9834
180	1.7351	1.5336	1.3667	1.2213	1.0923	0.9834
190	1.7359	1.5355	1.3664	1.2217	1.0941	0.9837
200	1.7358	1.5357	1.3666	1.2214	1.0943	0.9842

^aStandard uncertainties σ are $\sigma(T) = 0.02$ K, $\sigma(\eta) = 0.5\%$ and $\sigma(p) = 1.0$ kPa (at 68% confidence interval).

relations^{25,28}

$$\Delta\kappa_s = \kappa_s - \kappa_s^o \tag{10}$$

$$\Delta L_f = L_f - L_f^o \tag{11}$$

$$\Delta Z = Z - Z^o \tag{12}$$

$$\Delta\eta = \eta - \eta^o \tag{13}$$

$$\Delta\Gamma = \Gamma - \Gamma^o \tag{14}$$

where the superscript ‘^o’ refers to the values for pure ethanol control (91% ethanol in water). The variations of $\Delta\kappa_s$, ΔL_f , ΔZ , $\Delta\eta$, τ , (α/f^2) and $\Delta\Gamma$ with

Table 3a. The viscosities, ($\eta/10^{-3}$ N s m $^{-2}$) of succussed ethanol control with succussions equivalent to each potency (C) at temperatures (293.15–318.15) K and atmospheric pressure^a

Potency (C)	T/K					
	293.15	298.15	303.15	308.15	313.15	318.15
0	1.4935	1.3239	1.1844	1.0616	0.9548	0.8677
1	1.5649	1.3898	1.2369	1.0924	0.9875	0.8868
2	1.5644	1.3891	1.2346	1.0938	0.9863	0.8841
4	1.5632	1.3873	1.2357	1.0950	0.9873	0.8838
6	1.5622	1.3880	1.2362	1.0941	0.9882	0.8832
8	1.5638	1.3866	1.2373	1.0957	0.9891	0.8825
10	1.5626	1.3872	1.2371	1.0953	0.9883	0.8829
12	1.5605	1.3881	1.2383	1.0960	0.9876	0.8847
14	1.5615	1.3886	1.2377	1.0952	0.9867	0.8851
16	1.5619	1.3882	1.2370	1.0971	0.9862	0.8858
18	1.5645	1.3888	1.2359	1.0967	0.9872	0.8863
20	1.5649	1.3858	1.2355	1.0974	0.9885	0.8867
22	1.5644	1.3873	1.2370	1.0961	0.9881	0.8859
24	1.5651	1.3886	1.2382	1.0976	0.9877	0.8852
26	1.5641	1.3877	1.2378	1.0964	0.9871	0.8845
28	1.5635	1.3873	1.2349	1.0943	0.9858	0.8841
30	1.5629	1.3882	1.2353	1.0939	0.9863	0.8838
40	1.5642	1.3873	1.2356	1.0935	0.9869	0.8846
50	1.5632	1.3875	1.2361	1.0931	0.9876	0.8827
60	1.5637	1.3866	1.2365	1.0937	0.9881	0.8835
70	1.5622	1.3862	1.2369	1.0947	0.9887	0.8830
80	1.5612	1.3869	1.2372	1.0951	0.9884	0.8836
90	1.5619	1.3876	1.2363	1.0946	0.9892	0.8851
100	1.5624	1.3882	1.2353	1.0941	0.9886	0.8844
110	1.5631	1.3871	1.2357	1.0958	0.9880	0.8837
120	1.5621	1.3865	1.2368	1.0963	0.9875	0.8829
130	1.5619	1.3856	1.2361	1.0968	0.9871	0.8825
140	1.5642	1.3851	1.2352	1.0974	0.9878	0.8822
150	1.5633	1.3844	1.2365	1.0969	0.9883	0.8835
160	1.5618	1.3861	1.2369	1.0963	0.9888	0.8847
170	1.5623	1.3867	1.2373	1.0955	0.9882	0.8852
180	1.5648	1.3872	1.2377	1.0951	0.9874	0.8846
190	1.5638	1.3848	1.2352	1.0947	0.9868	0.8854
200	1.5631	1.3858	1.2366	1.0941	0.9873	0.8861

^aStandard uncertainties σ are $\sigma(T) = 0.02$ K, $\sigma(\eta) = 0.5\%$ and $\sigma(p) = 1.0$ kPa (at 68% confidence interval).

potency, C of *Acidum citricum* and temperature are displayed graphically in Figs. 4 to 10, respectively.

Discussion

A close review of Tables 1, 2 and 3 and Tables 1a, 2a and 3a; and Figs. 1, 2 and 3 and Figs. 1a, 2a

and 3a specifies that the values of ρ , u and η of the succussed ethanol control and potencies (1C to 200C) of *Acidum citricum* are greater than those of unsuccussed ethanol control at each examined temperature and these values decrease with rise in temperature. The greater ρ , u and η values (Figs. 1a, 2a and 3a) for the succussed ethanol control than unsuccussed

Table 4. Isentropic compressibilities, $\kappa_s/(10^{-10} \text{ m}^2 \text{ N}^{-1})$ ethanol control and *Acidum citricum* potencies (C) at temperatures (293.15–318.15) K^a

Potency (C)	T/K					
	293.15	298.15	303.15	308.15	313.15	318.15
0	8.6506	8.9332	9.2313	9.5435	9.8688	10.2166
1	7.6811	7.9287	8.1898	8.4649	8.7538	9.0588
2	7.9191	8.1752	8.4496	8.7395	9.0439	9.3642
4	7.7992	8.0527	8.3223	8.6061	8.9045	9.2184
6	7.9414	8.2028	8.4802	8.7721	9.0790	9.4018
8	7.8637	8.1203	8.3915	8.6776	8.9774	9.2941
10	7.8684	8.1243	8.3982	8.6862	8.9855	9.3072
12	7.8699	8.1274	8.4008	8.6868	8.9882	9.3066
14	7.8627	8.1194	8.3948	8.6787	8.9767	9.2949
16	7.8547	8.1091	8.3814	8.6665	8.9656	9.2827
18	7.8455	8.1013	8.3731	8.6593	8.9605	9.2773
20	7.8434	8.0953	8.3679	8.6549	8.9552	9.2719
22	7.8419	8.0980	8.3707	8.6559	8.9547	9.2728
24	7.8377	8.0909	8.3630	8.6492	8.9468	9.2636
26	7.8295	8.0825	8.3540	8.6395	8.9397	9.2544
28	7.9304	8.1878	8.4644	8.7556	9.0617	9.3828
30	7.7493	7.9975	8.2672	8.5441	8.8391	9.1494
40	7.5910	7.8361	8.0946	8.3668	8.6526	8.9535
50	7.4672	7.7020	7.9525	8.2165	8.4942	8.7865
60	7.6247	7.8656	8.1256	8.3990	8.6866	8.9858
70	7.6114	7.8547	8.1109	8.3804	8.6702	8.9699
80	7.6087	7.8488	8.1069	8.3785	8.6653	8.9663
90	7.6087	7.8507	8.1099	8.3828	8.6694	8.9709
100	7.6229	7.8651	8.1245	8.3982	8.6856	8.9834
110	7.4792	7.7162	7.9684	8.2338	8.5131	8.8067
120	7.5997	7.8428	8.1014	8.3728	8.6609	8.9603
130	7.6206	7.8636	8.1239	8.3976	8.6850	8.9873
140	7.6153	7.8575	8.1165	8.3887	8.6757	8.9745
150	7.4373	7.6682	7.9157	8.1773	8.4528	8.7425
160	7.4959	7.7320	7.9852	8.2515	8.5314	8.8257
170	7.6101	7.8520	8.1114	8.3842	8.6711	8.9694
180	7.6184	7.8582	8.1166	8.3890	8.6758	8.9774
190	7.6151	7.8585	8.1186	8.3917	8.6793	8.9814
200	7.6166	7.8604	8.1188	8.3917	8.6788	8.9805

^aStandard uncertainties σ are $\sigma(T) = 0.02 \text{ K}$, $\sigma(\kappa_s) = 2.5 \times 10^{-13} \text{ m}^2 \text{ N}^{-1}$.

ethanol (91% ethanol in water) control (0 potency) for each potency indicate more compact packing of the molecules due to succussion process, which leads to increase in these values. However, these remain nearly constant with increasing the number of succussions with increase in potency. The values of ρ , u and η are maximum at 1C and then decrease on addition of *Acidum citricum* with dilution till the potency

2C and then these values increase to a maximum at 4C. Then, these values again decrease till 6C, then increase up to potency 8C and thereafter these values increase slightly up to 26C, and again decrease up to 28C and thereafter, these values increase regularly till 50C (Figs. 1, 2 and 3) with successive dilutions. After exhibiting a maximum at 50C these values decrease to 60C and then remain nearly constant till 200C,

Table 5. Intermolecular free lengths, $L_f/(10^{-10} \text{ m})$ ethanol control and *Acidum citricum* potencies (C) at temperatures (293.15–318.15) K^a

Potency (C)	T/K					
	293.15	298.15	303.15	308.15	313.15	318.15
0	5.9943	6.1475	6.3062	6.4699	6.6381	6.8140
1	5.6484	5.7916	5.9398	6.0933	6.2519	6.4163
2	5.7353	5.8809	6.0333	6.1913	6.3546	6.5235
4	5.6917	5.8367	5.9877	6.1439	6.3055	6.4726
6	5.7433	5.8908	6.0442	6.2029	6.3669	6.5366
8	5.7152	5.8611	6.0125	6.1694	6.3312	6.4991
10	5.7169	5.8626	6.0149	6.1724	6.3341	6.5037
12	5.7174	5.8637	6.0158	6.1726	6.3350	6.5035
14	5.7148	5.8608	6.0137	6.1698	6.3310	6.4994
16	5.7119	5.8571	6.0089	6.1654	6.3271	6.4951
18	5.7086	5.8543	6.0059	6.1629	6.3253	6.4932
20	5.7078	5.8521	6.0041	6.1613	6.3234	6.4913
22	5.7073	5.8531	6.0050	6.1617	6.3232	6.4916
24	5.7058	5.8505	6.0023	6.1593	6.3204	6.4884
26	5.7027	5.8475	5.9991	6.1558	6.3179	6.4852
28	5.7394	5.8854	6.0386	6.1970	6.3609	6.5300
30	5.6735	5.8166	5.9678	6.1217	6.2823	6.4483
40	5.6152	5.7576	5.9052	6.0579	6.2156	6.3789
50	5.5693	5.7082	5.8531	6.0032	6.1585	6.3191
60	5.6277	5.7685	5.9165	6.0695	6.2279	6.3904
70	5.6228	5.7645	5.9111	6.0628	6.2220	6.3847
80	5.6218	5.7623	5.9097	6.0621	6.2202	6.3834
90	5.6218	5.7630	5.9108	6.0637	6.2217	6.3851
100	5.6270	5.7683	5.9161	6.0692	6.2275	6.3895
110	5.5737	5.7134	5.8590	6.0096	6.1653	6.3264
120	5.6184	5.7601	5.9077	6.0601	6.2186	6.3813
130	5.6262	5.7677	5.9159	6.0690	6.2273	6.3909
140	5.6242	5.7655	5.9132	6.0658	6.2239	6.3864
150	5.5581	5.6956	5.8396	5.9889	6.1435	6.3033
160	5.5799	5.7193	5.8651	6.0160	6.1720	6.3332
170	5.6223	5.7635	5.9113	6.0642	6.2223	6.3846
180	5.6253	5.7657	5.9132	6.0659	6.2240	6.3874
190	5.6241	5.7659	5.9139	6.0669	6.2252	6.3888
200	5.6247	5.7666	5.9140	6.0669	6.2250	6.3885

^aStandard uncertainties σ are $\sigma(T) = 0.02 \text{ K}$, $\sigma(L_f) = 2.5 \times 10^{-13} \text{ m}$.

except exhibiting maximums at potencies 110C, 150C to 160 C (Figs. 1, 2 and 3). Additionally, the values of η exhibit a maximum at 16C (Fig. 3), which is not observed in case of ρ and u values (Figs. 1 and 2). The observed abnormal trends in ρ , u and η values at the potencies, viz., 1C, 4C, 30C to 50C, 110C, 150C and 160C exhibiting larger values than the other potencies. This specifies that the solution structure of these

potencies is different as compared to other potencies and the ethanol control, it may be due interaction between *Acidum citricum* and ethanol-water molecules; and the succession process is also responsible for the higher values.

A close scrutiny of Tables 5 and 6 specify that the κ_s and L_f values for succussed ethanol controls and *Acidum citricum* potencies are less than those of

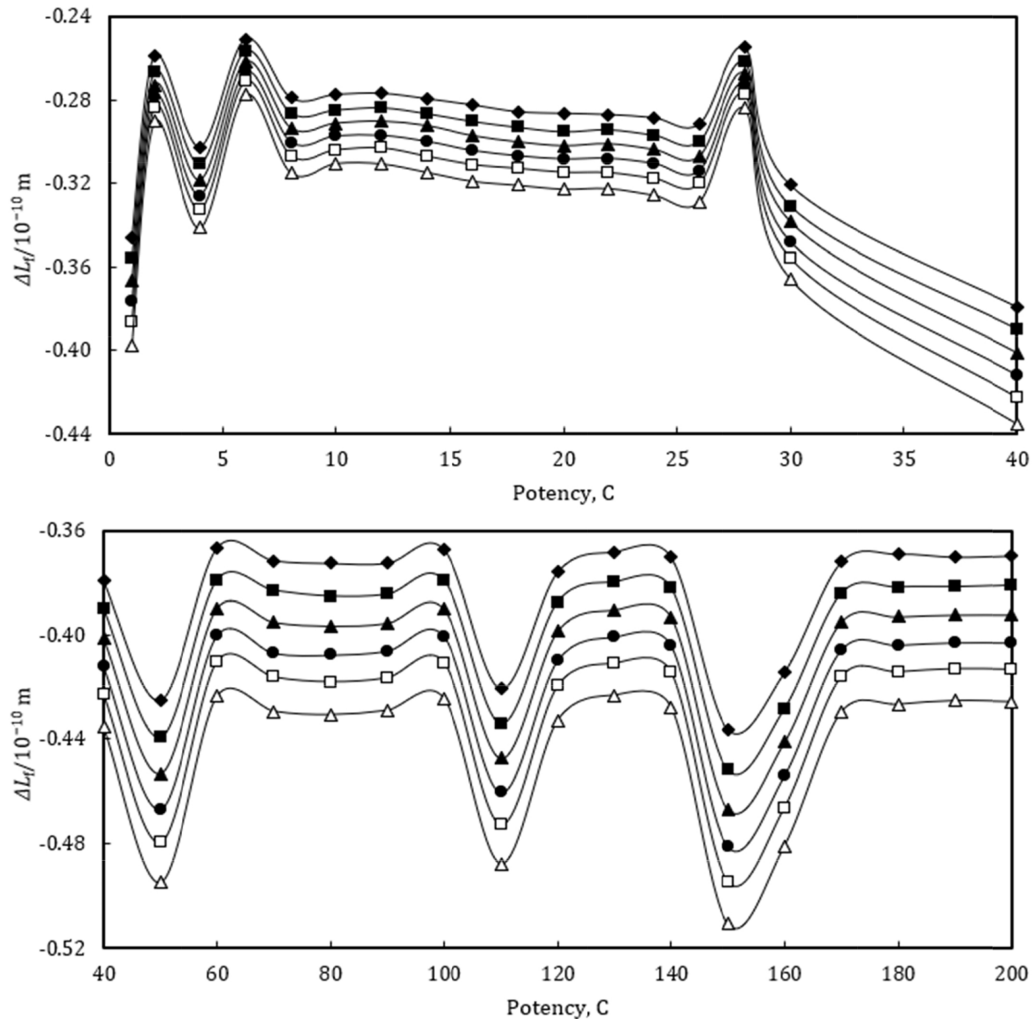


Fig. 5. Variations of deviations in intermolecular free length, ΔL_f vs. potency, C of *Acidum citricum* for dilutions of *Acidum citricum* at temperatures, 293.15 K, \blacklozenge ; 298.15 K, \blacksquare ; and 303.15 K, \blacktriangle ; 308.15 K, \bullet ; 313.15 K, \square ; and 318.15 K, \triangle

unsuccussed ethanol control for the studied potencies at each investigated temperature and these values increase with rise in temperature, which specifies substantial interaction among *Acidum citricum* and ethanol/water molecules and the impact of succussion process. The values of κ_s and L_f are minimum at 1C and then increase in presence of *Acidum citricum* with dilution till the potency 2C and after that these values decrease to minimum at 4C. Thereafter, these values increase up to 6C, then decrease to minimum at 8C and thereafter these values decrease slightly up to 26C, and again increase till 28C and thereafter, these values decrease regularly up to 50C (Tables 5 and 6) with successive dilutions. After exhibiting minimum at 50C these values increase to 60C and then remain nearly constant till 200C, except exhibiting minimums at potencies 110C, 150C to 160 C (Tables 5 and 6). Figs. 4 and 5 indicate that the values of $\Delta\kappa_s$ and ΔL_f are negative and these values are mini-

mum at 1C and then increase in presence of *Acidum citricum* with dilution till the potency 2C and after that these values decrease to minimum at 4C. Thereafter, these values increase till 6C, then decrease to minimum at 8C and thereafter these values decrease slightly up to 26C, and again increase till 28C and thereafter, these values decrease regularly till 50C (Figs. 4 and 5) with dilution. After exhibiting minimum at 50C these values increase to 60C and then remain nearly constant till 200C, except exhibiting minimums at potencies 110C, 150C to 160 C (Figs. 4 and 5). This indicates that at potencies 1C, 4C, 30C to 50C, 110C, 150C and 160C are less compressible and 2C, 6C and 28C are more compressible than the other potencies, indicating that the potencies 1C, 4C, 30C to 50C, 110C, 150C and 160C exhibit more compact solution structure and the potencies 2C, 6C and 28C exhibit less compact solution structure as compared to other potencies. The minimum in $\Delta\kappa_s$ and ΔL_f

Table 6. Specific acoustic impedances, $Z/(10^5 \text{ kg m}^{-2} \text{ s}^{-1})$ ethanol control and *Acidum citricum* potencies (C) at temperatures (293.15–318.15) K^a

Potency (C)	T/K					
	293.15	298.15	303.15	308.15	313.15	318.15
0	9.6661	9.4885	9.3101	9.1328	8.9562	8.7792
1	10.3751	10.1851	9.9945	9.8039	9.6140	9.4241
2	10.1696	9.9825	9.7926	9.6023	9.4129	9.2242
4	10.2641	10.0745	9.8833	9.6922	9.5018	9.3120
6	10.1497	9.9601	9.7693	9.5790	9.3892	9.2003
8	10.2115	10.0222	9.8323	9.6423	9.4534	9.2645
10	10.2089	10.0202	9.8288	9.6380	9.4496	9.2583
12	10.2082	10.0186	9.8275	9.6379	9.4484	9.2588
14	10.2129	10.0245	9.8320	9.6434	9.4555	9.2659
16	10.2195	10.0313	9.8404	9.6507	9.4618	9.2722
18	10.2261	10.0367	9.8458	9.6551	9.4649	9.2754
20	10.2283	10.0407	9.8491	9.6578	9.4680	9.2789
22	10.2298	10.0401	9.8486	9.6583	9.4687	9.2801
24	10.2335	10.0455	9.8541	9.6630	9.4744	9.2858
26	10.2401	10.0519	9.8605	9.6696	9.4793	9.2908
28	10.1639	9.9763	9.7854	9.5948	9.4050	9.2167
30	10.3049	10.1170	9.9265	9.7349	9.5443	9.3544
40	10.4380	10.2446	10.0526	9.8608	9.6696	9.4788
50	10.5427	10.3536	10.1620	9.9702	9.7787	9.5874
60	10.4087	10.2210	10.0292	9.8377	9.6465	9.4576
70	10.4181	10.2285	10.0386	9.8489	9.6560	9.4664
80	10.4219	10.2342	10.0429	9.8518	9.6605	9.4701
90	10.4218	10.2329	10.0411	9.8493	9.6582	9.4676
100	10.4098	10.2213	10.0297	9.8380	9.6470	9.4587
110	10.5316	10.3413	10.1492	9.9572	9.7653	9.5741
120	10.4267	10.2367	10.0451	9.8545	9.6623	9.4720
130	10.4118	10.2227	10.0306	9.8388	9.6477	9.4572
140	10.4165	10.2277	10.0362	9.8450	9.6539	9.4649
150	10.5694	10.3818	10.1910	9.9994	9.8079	9.6168
160	10.5181	10.3291	10.1369	9.9448	9.7532	9.5621
170	10.4205	10.2317	10.0398	9.8481	9.6569	9.4680
180	10.4136	10.2265	10.0354	9.8442	9.6532	9.4628
190	10.4161	10.2266	10.0344	9.8429	9.6515	9.4609
200	10.4162	10.2263	10.0359	9.8444	9.6533	9.4628

^aStandard uncertainties σ are $\sigma(T) = 0.02 \text{ K}$, $\sigma(Z) = 2.5 \times 10^3 \text{ kg m}^{-2} \text{ s}^{-1}$.

values at potencies 1C, 4C, 50C, 110C, 150C and 160C indicate that these have most compressed solution structure, hence, these potencies show diverse behaviour which may be attributed to interaction among *Acidum citricum* and ethanol-water molecules; and the succussion process is also responsible for the lower values.

A close perusal of Table 6 indicates that the specific acoustic impedances, Z for succussed ethanol controls and potencies of *Acidum citricum* are greater than those of unsuccussed ethanol control for the studied potencies at each investigated temperature and the values decrease with rise in temperature, which indicates significant interaction between *Acidum citricum*

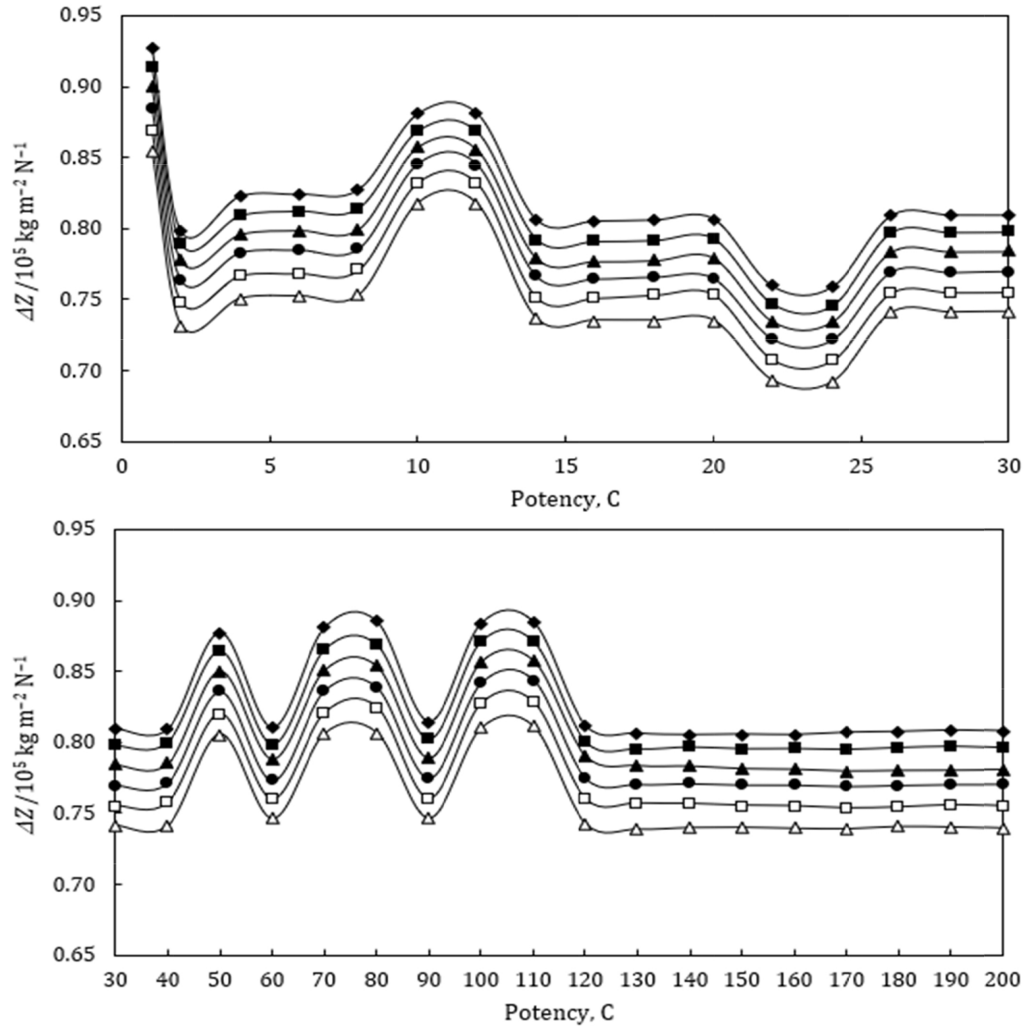


Fig. 6. Variations of deviations in acoustic impedance, ΔZ vs. potency, C of *Acidum citricum* for dilutions of *Acidum citricum* at temperatures, 293.15 K, \blacklozenge ; 298.15 K, \blacksquare ; and 303.15 K, \blacktriangle ; 308.15 K, \bullet ; 313.15 K, \square ; and 318.15 K, \triangle

and ethanol molecules. Fig. 6 indicates that ΔZ values are positive, i.e., Z values for *Acidum citricum* are more than those of ethanol control. The ΔZ values are maximum at 1C and then decrease in presence of *Acidum citricum* with dilution up to the potency 2C and after that these values increase to maximum at 4C. Thereafter, these values decrease till 6C, then increase to maximum at 8C and thereafter these values increase slightly up to 26C, and again decrease till 28C and thereafter, these values increase regularly up to 50C (Fig. 6) with successive dilutions. After exhibiting maximum at 50C these values decrease to 60C and then remain nearly constant till 200C, except exhibiting maximums at potencies 110C, 150C to 160C (Fig. 6). This indicates that at potencies 1C, 4C, 50C, 110C, 150C and 160C offer more resistance to the dissemination of sound waves through the solution due to more compressed structure than the other poten-

cies. The observed trends of Z and ΔZ values of these potencies may be due interaction between *Acidum citricum* and ethanol-water molecules; and the succussion process is also responsible for the greater values.

Fig. 7 indicates that $\Delta\eta$ values are positive, i.e., η values for *Acidum citricum* are greater than those of ethanol control. These $\Delta\eta$ values are maximum for potency 1C and then decrease up to the potency 4C and thereafter the values increase till potency 8C and then remain nearly constant on more dilution till 14C, thereafter these values increase and exhibit a maximum at 16C and thereafter from 18C the values remain nearly constant up to 30C and then these values increase regularly till potency 50C (Fig. 7) with successive dilutions. After exhibiting maximum at 50C these values decrease to 60C and then remain nearly constant till 200C, except exhibiting maximums at potencies 110C, 150C to 160C (Fig. 7). The

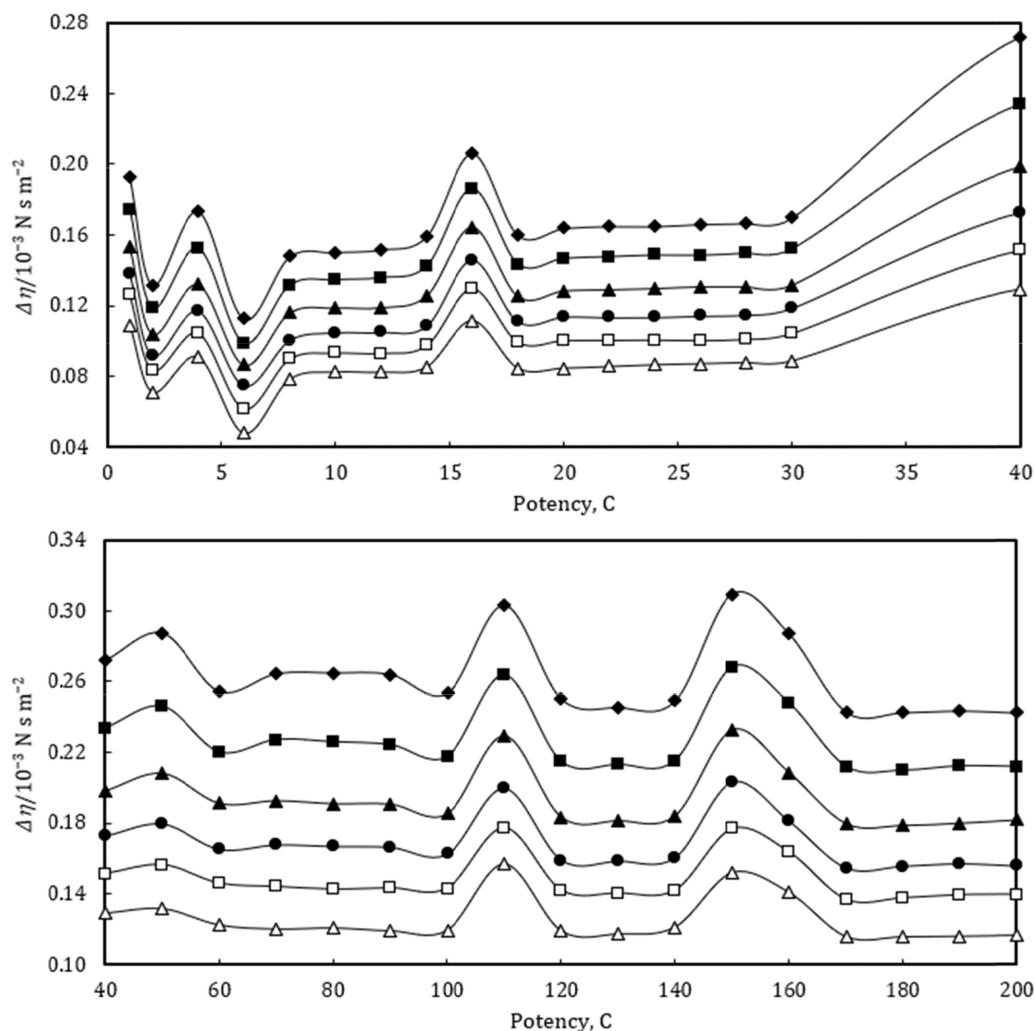


Fig. 7. Variations of deviations in viscosity, $\Delta\eta$ vs. potency, C of *Acidum citricum* for dilutions of *Acidum citricum* at temperatures, 293.15 K, \blacklozenge ; 298.15 K, \blacksquare ; and 303.15 K, \blacktriangle ; 308.15 K, \bullet ; 313.15 K, \square ; and 318.15 K, \triangle

maximums in values of $\Delta\eta$ at 1C, 4C, 16C, 50C, 110C, 150C and 160C potencies may be due interaction between *Acidum citricum* and ethanol-water molecules; and the succession process is also responsible for the greater values. It is observed that the observed trends in the values of ρ and u and η ; and calculated values, κ_s , L_f , $\Delta\kappa_s$, ΔL_f , ΔZ and $\Delta\eta$ are in agreement with each other.

A close inspection of Table 7 specifies that the values of R_A for 1C, 4C, 30C to 50C, 110C, 150C and 160C potencies of *Acidum citricum* are more than 1 and exhibit maximums at these potencies, other potencies show varying values. The values of R_A decrease with rise in temperature. The changes in values of R_A of solution in presence of *Acidum citricum* are due to diverse extents of breaking/formation of hydrogen-bonded associates in ethanol controls and their interaction with *Acidum citricum* with successive

dilutions and successions. The values of R_A decrease with rise in temperature that may due breaking of associations/interactions amid the component molecules.

A close scrutiny of Table 8 and Fig. 8 specifies that the relaxation time, τ for *Acidum citricum* potencies is greater than those of ethanol controls for all the potencies at each investigated temperature and these values decrease with rise in temperature. The τ value is the time in which the structural deformation caused by propagation of ultrasonic wave is restored in the medium through translational motion, which point to substantial interaction among *Acidum citricum* and ethanol-water molecules. These τ values decrease from 1C to 2C and then increase till potency 4C and after that values decrease till potency 6C and then increase till 8C and remain nearly constant on more dilution from 8C to 14C, thereafter these

Table 7. The relative associations, R_A for *Acidum citricum* potencies (C) at temperatures (293.15–318.15) K^a

Potency (C)	T/K					
	293.15	298.15	303.15	308.15	313.15	318.15
1	1.0067	1.0063	1.0059	1.0056	1.0055	1.0050
2	1.0007	1.0002	0.9999	0.9996	0.9997	0.9992
4	1.0019	1.0015	1.0012	1.0009	1.0009	1.0004
6	0.9999	0.9994	0.9992	0.9989	0.9989	0.9985
8	1.0009	1.0005	1.0002	0.9999	0.9998	0.9994
10	1.0011	1.0007	1.0004	1.0001	1.0001	0.9997
12	1.0012	1.0008	1.0005	1.0002	1.0002	0.9997
14	1.0011	1.0009	1.0006	1.0003	1.0002	0.9998
16	1.0012	1.0007	1.0005	1.0002	1.0001	0.9997
18	1.0012	1.0007	1.0004	1.0001	1.0002	0.9997
20	1.0013	1.0007	1.0004	1.0001	1.0001	0.9998
22	1.0014	1.0010	1.0007	1.0004	1.0002	1.0002
24	1.0016	1.0010	1.0008	1.0005	1.0005	1.0003
26	1.0017	1.0012	1.0009	1.0006	1.0006	1.0003
28	1.0013	1.0008	1.0005	1.0003	1.0003	1.0000
30	1.0026	1.0021	1.0025	1.0015	1.0015	1.0010
40	1.0051	1.0042	1.0038	1.0035	1.0035	1.0030
50	1.0064	1.0059	1.0055	1.0051	1.0050	1.0044
60	1.0044	1.0038	1.0035	1.0031	1.0031	1.0025
70	1.0042	1.0037	1.0033	1.0029	1.0029	1.0023
80	1.0046	1.0040	1.0036	1.0032	1.0032	1.0027
90	1.0046	1.0040	1.0037	1.0033	1.0033	1.0028
100	1.0043	1.0038	1.0034	1.0031	1.0031	1.0025
110	1.0061	1.0056	1.0052	1.0048	1.0048	1.0042
120	1.0041	1.0035	1.0032	1.0030	1.0030	1.0023
130	1.0044	1.0039	1.0035	1.0032	1.0032	1.0027
140	1.0045	1.0040	1.0036	1.0033	1.0032	1.0027
150	1.0070	1.0064	1.0059	1.0055	1.0055	1.0049
160	1.0061	1.0055	1.0052	1.0048	1.0047	1.0042
170	1.0045	1.0040	1.0036	1.0033	1.0032	1.0027
180	1.0044	1.0038	1.0035	1.0031	1.0031	1.0026
190	1.0044	1.0039	1.0036	1.0032	1.0032	1.0027
200	1.0047	1.0041	1.0039	1.0036	1.0036	1.0030

^aStandard uncertainties σ are $\sigma(T) = 0.02$ K, $\sigma(R_A) = 0.0001$.

values increase and exhibit a maximum at 16C and thereafter from 18C the values remain nearly constant till 30C and thereafter these values increase regularly up to 50C (Fig. 8) with successive dilutions. After exhibiting maximum at 50C these values decrease to 60C and then remain nearly constant till 200C, except exhibiting maximums at potencies 110C, 150C to 160 C (Fig. 8). This indicates that at potencies 1C, 4C, 16C, 50C, 110C, 150C and

160C possess more compressed solution structure as compared to other potencies. The maximum in the values of τ for 1C, 4C, 10C, 12C, 70C and 140C potencies indicate that the structural deformation by propagation of ultrasonic wave is restored slowly, which may be due substantial interaction between *Acidum citricum* and water-ethanol molecules; and the succession process is also responsible for the greater values.

Table 8. The relaxation time, τ (10^{-12} s) of ethanol control and *Acidum citricum* potencies (C) at temperatures (293.15–318.15) K^a

Potency (C)	T/K					
	293.15	298.15	303.15	308.15	313.15	318.15
0	1.7226	1.5769	1.4578	1.3509	1.2564	1.1820
1	1.7269	1.5840	1.4603	1.3542	1.2616	1.1793
2	1.7160	1.5725	1.4515	1.3441	1.2520	1.1723
4	1.7336	1.5854	1.4607	1.3527	1.2578	1.1785
6	1.7006	1.5562	1.4371	1.3293	1.2305	1.1480
8	1.7210	1.5761	1.4551	1.3448	1.2506	1.1727
10	1.7246	1.5801	1.4593	1.3510	1.2557	1.1793
12	1.7264	1.5820	1.4599	1.3513	1.2559	1.1791
14	1.7327	1.5876	1.4664	1.3542	1.2596	1.1813
16	1.7801	1.6327	1.5073	1.3954	1.2965	1.2120
18	1.7297	1.5852	1.4620	1.3540	1.2595	1.1781
20	1.7333	1.5875	1.4648	1.3564	1.2599	1.1775
22	1.7341	1.5890	1.4660	1.3561	1.2600	1.1788
24	1.7331	1.5886	1.4654	1.3552	1.2591	1.1788
26	1.7326	1.5869	1.4650	1.3546	1.2578	1.1784
28	1.7556	1.6088	1.4843	1.3735	1.2758	1.1956
30	1.7185	1.5740	1.4507	1.3446	1.2483	1.1670
40	1.7869	1.6274	1.4923	1.3770	1.2761	1.1900
50	1.7731	1.6121	1.4766	1.3597	1.2585	1.1708
60	1.7770	1.6192	1.4903	1.3738	1.2725	1.1839
70	1.7841	1.6243	1.4893	1.3725	1.2702	1.1814
80	1.7839	1.6218	1.4867	1.3723	1.2678	1.1816
90	1.7831	1.6205	1.4870	1.3725	1.2692	1.1827
100	1.7755	1.6165	1.4839	1.3707	1.2715	1.1825
110	1.7916	1.6334	1.5021	1.3846	1.2851	1.2033
120	1.7668	1.6088	1.4775	1.3628	1.2665	1.1788
130	1.7648	1.6093	1.4773	1.3640	1.2658	1.1785
140	1.7694	1.6126	1.4812	1.3670	1.2687	1.1797
150	1.7878	1.6278	1.4957	1.3789	1.2755	1.1889
160	1.7794	1.6224	1.4888	1.3727	1.2722	1.1869
170	1.7617	1.6078	1.4785	1.3642	1.2618	1.1761
180	1.7625	1.6068	1.4791	1.3661	1.2635	1.1771
190	1.7625	1.6089	1.4791	1.3670	1.2661	1.1780
200	1.7628	1.6095	1.4793	1.3666	1.2663	1.1785

^aStandard uncertainties σ are $\sigma(T) = 0.02$ K, $\sigma(\tau) = 2 \times 10^{-14}$ s.

The loss of energy of ultrasonic waves by the concerned medium is called ultrasonic absorption or attenuation (α/f^2). The (α/f^2) values for *Acidum citricum* potencies are greater than those of ethanol controls for all the potencies at each investigated temperature and these values decrease with rise in temperature (Table 9 and Fig. 9). As expected, the trends of ultrasonic absorption resemble with the re-

laxation time (Fig. 8). The viscosity appears to be the main factor accountable for ultrasonic absorption in these homoeopathic dilutions. The (α/f^2) values decrease from 1C to 2C and then increase till potency 4C and after that values decrease till potency 6C and then increase up to potency 8C and remain nearly constant on more dilution from 8C to 14C, then these values increase and exhibit a maximum at 16C and afterwards

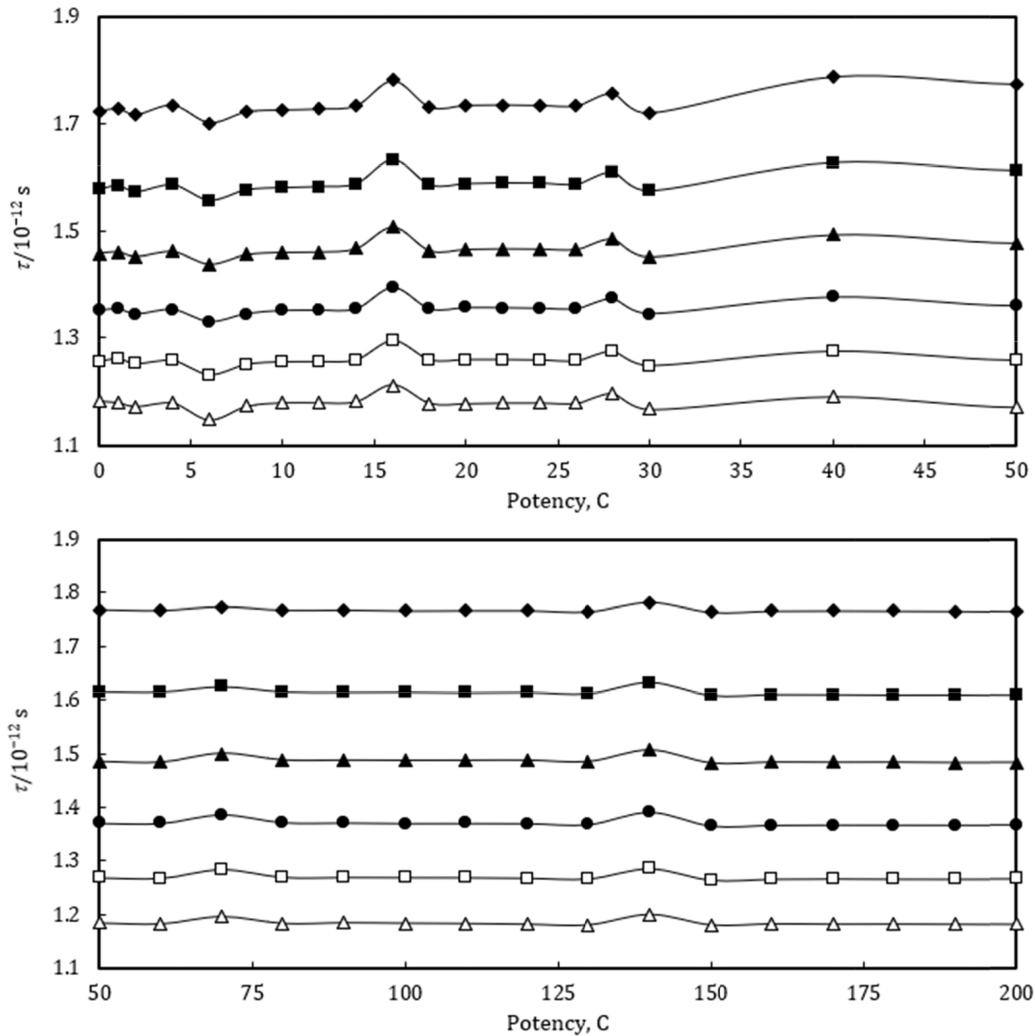


Fig. 8. Variations of relaxation time, τ vs. potency, C of *Acidum citricum* for dilutions of *Acidum citricum* at temperatures, 293.15 K, \blacklozenge ; 298.15 K, \blacksquare ; and 303.15 K, \blacktriangle ; 308.15 K, \bullet ; 313.15 K, \square ; and 318.15 K, \triangle

from 18C the values remain almost constant up to potency 30C and then these values increase regularly up to 50C (Fig. 9) with successive dilutions. After exhibiting a maximum at 50C these values decrease to 60C and then remain nearly constant till 200C, except exhibiting maximums at potencies 110C, 150C to 160 C (Fig. 9). The enhancement in (α/f^2) values for 1C, 4C, 16C, 50C, 110C, 150C and 160C potencies reflect a more ordered structure and significant interactions among the constituents may be due the presence of physical interaction because of hydrogen bonding; and the succession process is also responsible for the greater values.

A close perusal of Table 10 and Fig. 10 specifies that the pseudo-Grüneisen parameters, Γ for *Acidum citricum* potencies is greater than those of ethanol controls for the studied potencies at each investigated temperature and these values increase with

rise in temperature, which indicates substantial interaction between *Acidum citricum* and ethanol-water molecules. Fig. 10 indicates that $\Delta\Gamma$ values are positive, i.e., $\Delta\Gamma$ values for *Acidum citricum* potencies are more than those of ethanol control. These $\Delta\Gamma$ values are maximum for potency 1C and then decrease up to potency 4C and after that values increase up to potency 8C and then remain nearly constant up to potency 14C, afterwards these values increase and exhibit a maximum at 16C and thereafter from 18C the values remain nearly constant till 30C and thereafter these values increase regularly up to 50C (Fig. 10) with successive dilutions. After exhibiting a maximum at 50C these values decrease to 60C and then remain nearly constant till 200C, except exhibiting maximums at potencies 110C, 150C to 160 C (Fig. 10). The maximum in the values of $\Delta\Gamma$ for 1C, 4C, 26C, 50C, 110C and 150C to 160C potencies may

Table 9. The ultrasonic absorption, $(\alpha/f^2)/(10^{-11} \text{ Np s}^{-2} \text{ m}^{-1})$ of ethanol control *Acidum citricum* potencies (C) at temperatures (293.15–318.15) K^a

Potency (C)	T/K					
	293.15	298.15	303.15	308.15	313.15	318.15
0	3.3969	3.1095	2.8747	2.6638	2.4775	2.3308
1	3.4053	3.1236	2.8796	2.6703	2.4878	2.3256
2	3.3839	3.1008	2.8623	2.6505	2.4689	2.3116
4	3.4185	3.1263	2.8804	2.6673	2.4803	2.3239
6	3.3535	3.0688	2.8339	2.6212	2.4265	2.2638
8	3.3937	3.1079	2.8693	2.6518	2.4661	2.3124
10	3.4007	3.1159	2.8776	2.6640	2.4761	2.3255
12	3.4044	3.1196	2.8789	2.6647	2.4766	2.3251
14	3.4168	3.1306	2.8916	2.6704	2.4839	2.3295
16	3.5102	3.2196	2.9723	2.7517	2.5567	2.3899
18	3.4108	3.1258	2.8830	2.6699	2.4836	2.3231
20	3.4179	3.1305	2.8885	2.6747	2.4845	2.3220
22	3.4195	3.1335	2.8908	2.6741	2.4846	2.3244
24	3.4175	3.1326	2.8897	2.6723	2.4829	2.3245
26	3.4166	3.1292	2.8888	2.6711	2.4802	2.3237
28	3.4618	3.1725	2.9270	2.7083	2.5157	2.3577
30	3.3887	3.1038	2.8607	2.6515	2.4616	2.3012
40	3.5237	3.2091	2.9427	2.7153	2.5163	2.3466
50	3.4964	3.1789	2.9118	2.6811	2.4817	2.3088
60	3.5040	3.1928	2.9388	2.7091	2.5093	2.3345
70	3.5181	3.2031	2.9367	2.7064	2.5048	2.3296
80	3.5177	3.1980	2.9317	2.7060	2.5000	2.3301
90	3.5161	3.1955	2.9323	2.7065	2.5028	2.3322
100	3.5012	3.1877	2.9261	2.7029	2.5072	2.3317
110	3.5329	3.2209	2.9620	2.7303	2.5342	2.3729
120	3.4839	3.1725	2.9135	2.6873	2.4973	2.3245
130	3.4801	3.1734	2.9130	2.6897	2.4961	2.3240
140	3.4891	3.1798	2.9208	2.6957	2.5018	2.3263
150	3.5254	3.2099	2.9495	2.7191	2.5151	2.3443
160	3.5089	3.1992	2.9357	2.7069	2.5087	2.3404
170	3.4739	3.1704	2.9156	2.6900	2.4882	2.3191
180	3.4755	3.1686	2.9166	2.6938	2.4916	2.3212
190	3.4756	3.1726	2.9167	2.6955	2.4967	2.3229
200	3.4761	3.1738	2.9172	2.6949	2.4970	2.3239

^aStandard uncertainties σ are $\sigma(T) = 0.02 \text{ K}$, $\sigma(\alpha/f^2) = 2 \times 10^{-14} \text{ Np s}^{-2} \text{ m}^{-1}$.

be due substantial interaction among *Acidum citricum* and water-ethanol molecules; and the succussion process is also responsible for the greater values.

It has been observed from the analysis of the studied physicochemical parameters, viz., κ_s , L_f , Γ , Z , R_A , $\Delta\kappa_s$, ΔL_f , ΔZ , $\Delta\eta$, τ , (α/f^2) and $\Delta\Gamma$ that all the potencies show more compressed solution structure in comparison to pure ethanol control; and the

potencies 1C, 4C, 16C, 50C, 110C, 150C and 160C exhibit more compressed solution structure than the other studied potencies. The differences in the physicochemical properties of the ethanol control (91% ethanol in water) and the succussed ethanol controls corresponding to each potency clearly indicate more compact packing of the molecules due to the succussion process, which leads to an increase in these

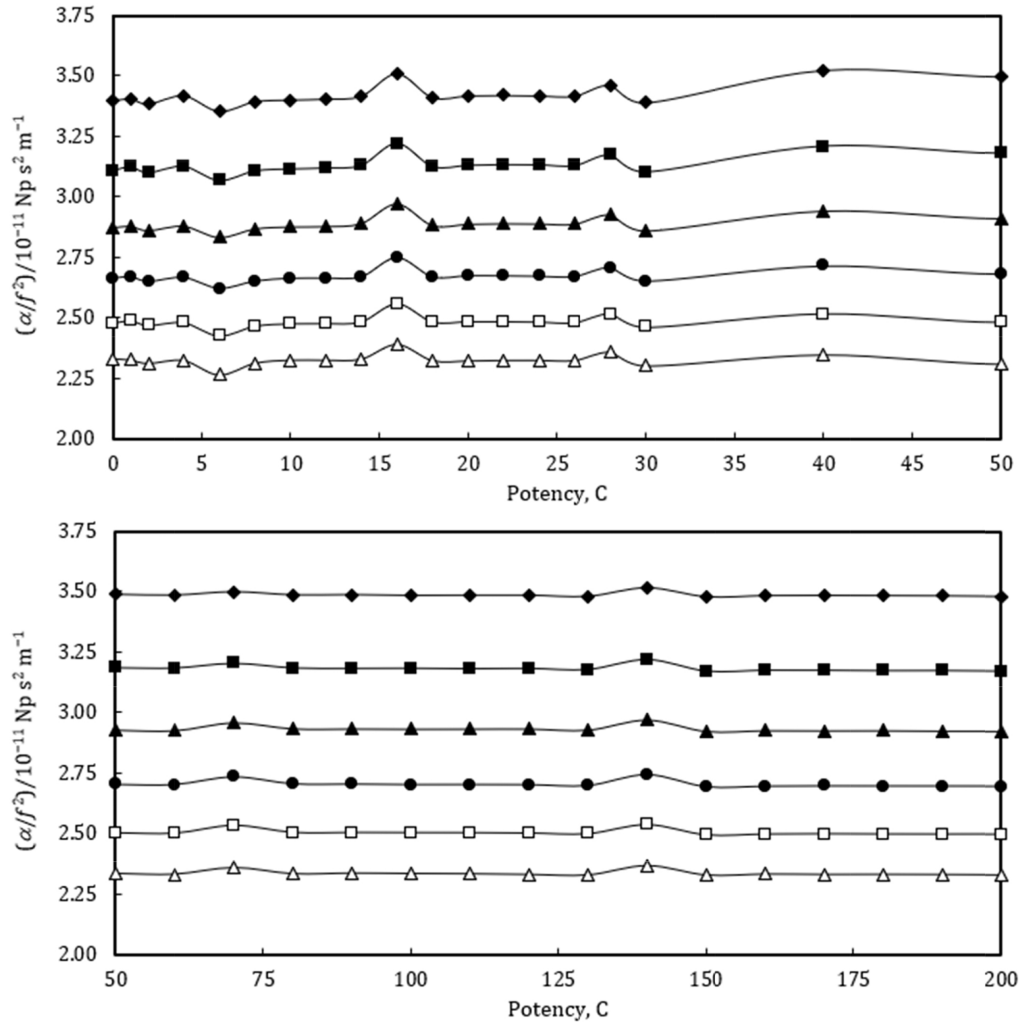


Fig. 9. Variations of ultrasonic absorption, (α/f^2) vs. potency, C of *Acidum citricum* for dilutions of *Acidum citricum* at temperatures, 293.15 K, \blacklozenge ; 298.15 K, \blacksquare ; and 303.15 K, \blacktriangle ; 308.15 K, \bullet ; 313.15 K, \square ; and 318.15 K, \triangle

values. The differences between the physicochemical properties of these *Acidum citricum* dilutions, succussed ethanol control and unsuccussed ethanol control (91% ethanol in water) clearly specify that both the succussion process as well as the presence of medicine which consequences in substantial structural changes in solution for the studies potencies and it is more pronounced in some potencies.

The outcomes can be qualitatively discussed in terms of interactions prevailing in these *Acidum citricum* dilutions in ethanol-water controls. The main factors which may be affecting the solution structure are the nature of solute, the presence of medicine molecules and the potentization process. It is a recognised fact that hydrogen bonding is one of the utmost significant weak interactions amongst the molecules in solution leading to the formation of well-defined molecular aggregates, called as

dissipative structures.^{4,20} It has been reported³⁸ that the potentisation process permanently modifies the properties of the solution. The succussion process excites the creation of dissipative structures, which are exaggerated by presence of ethanol and medicine molecules³⁸ (citric acid dissociated into H^+ and citrate ions). The results may be understood by considering the interactions that can take place between H^+ and citrate ions and the molecular aggregates of water-ethanol molecules, *i.e.*, dissipative structures.¹⁸ The hydrogen bonding in ethanol-water will be significantly affected by the presence of H^+ and citrate ions in solution and it can be assumed that the effect of medicine molecules is likely to alter after each successive dilution and succussion on moving from one potency to another next potency. A qualitative comparison between various potencies can be considered due to the nature of driving force

Table 10. The pseudo-Grüneisen parameter, $\Gamma/(10^6)$ of ethanol control and *Acidum citricum* potencies (C) at temperatures (293.15–318.15) K^a

Potency (C)	T/K	293.15	298.15	303.15	308.15	313.15	318.15
0		8.1269	7.9552	7.7821	7.6087	7.4381	7.2613
1		9.6171	9.4200	9.2202	9.0184	8.8160	8.6119
2		9.3728	9.1802	8.9803	8.7779	8.5753	8.3724
4		9.5148	9.3176	9.1154	8.9117	8.7074	8.5026
6		9.3648	9.1671	8.9654	8.7625	8.5590	8.3554
8		9.4388	9.2421	9.0422	8.8404	8.6387	8.4353
10		9.4380	9.2423	9.0398	8.8362	8.6354	8.4279
12		9.4404	9.2430	9.0410	8.8396	8.6367	8.4322
14		9.3800	9.1832	8.9802	8.7819	8.5831	8.3796
16		9.4456	9.2509	9.0494	8.8479	8.6463	8.4420
18		9.4644	9.2675	9.0659	8.8626	8.6584	8.4540
20		9.4679	9.2760	9.0730	8.8687	8.6651	8.4598
22		9.4254	9.2288	9.0270	8.8255	8.6250	8.4180
24		9.4034	9.2104	9.0092	8.8069	8.6072	8.4020
26		9.4520	9.2578	9.0560	8.8530	8.6494	8.4457
28		9.3457	9.1525	8.9513	8.7488	8.5459	8.3429
30		9.5989	9.4043	9.1946	8.9977	8.7926	8.5870
40		9.8344	9.6348	9.4299	9.2232	9.0160	8.8078
50		9.8835	9.6884	9.4865	9.2824	9.0768	8.8704
60		9.7069	9.5141	9.3113	9.1071	8.9017	8.6990
70		9.7190	9.5226	9.3235	9.1228	8.9141	8.7101
80		9.7274	9.5344	9.3327	9.1294	8.9237	8.7179
90		9.7226	9.5275	9.3247	9.1201	8.9149	8.7091
100		9.7125	9.5179	9.3156	9.1110	8.9058	8.7043
110		9.8649	9.6679	9.4650	9.2602	9.0542	8.8475
120		9.7130	9.5163	9.3143	9.1104	8.9037	8.7006
130		9.7108	9.5151	9.3119	9.1072	8.9021	8.6963
140		9.7154	9.5205	9.3184	9.1149	8.9098	8.7068
150		9.9060	9.7140	9.5139	9.3104	9.1052	8.8991
160		9.8468	9.6519	9.4488	9.2442	9.0384	8.8320
170		9.7202	9.5253	9.3224	9.1180	8.9127	8.7100
180		9.7074	9.5155	9.3141	9.1106	8.9057	8.7002
190		9.7159	9.5194	9.3161	9.1119	8.9062	8.7003
200		9.6697	9.4740	9.2730	9.0698	8.8656	8.6610

^aStandard uncertainties σ are $\sigma(T) = 0.02$ K, $\sigma(\Gamma) = 2.5 \times 10^3$.

that leads to formation of aggregates (due to solvation of these ions by polar ethanol/water dipoles) between H⁺ and citrate ions and dissipative structures of ethanol-water molecules.^{19,39} This driving force is supplied by the succussion process in which a vast amount of mechanical energy (~404.3 Newton-meter by 10 strokes)⁴⁰ is transferred. This transference of energy by successive dilution and succussion process

is accountable for diverse/anomalous behaviour of *Acidum citricum* of different potencies. It has also been reported in literature^{5,7} that the same medicine of different potency displays diverse behaviour due to vehicle-molecule structure (ethanol-water aggregates) generated by potentiation process.

The findings presented in this study are well supported by other investigators^{1,41,42} who, by using

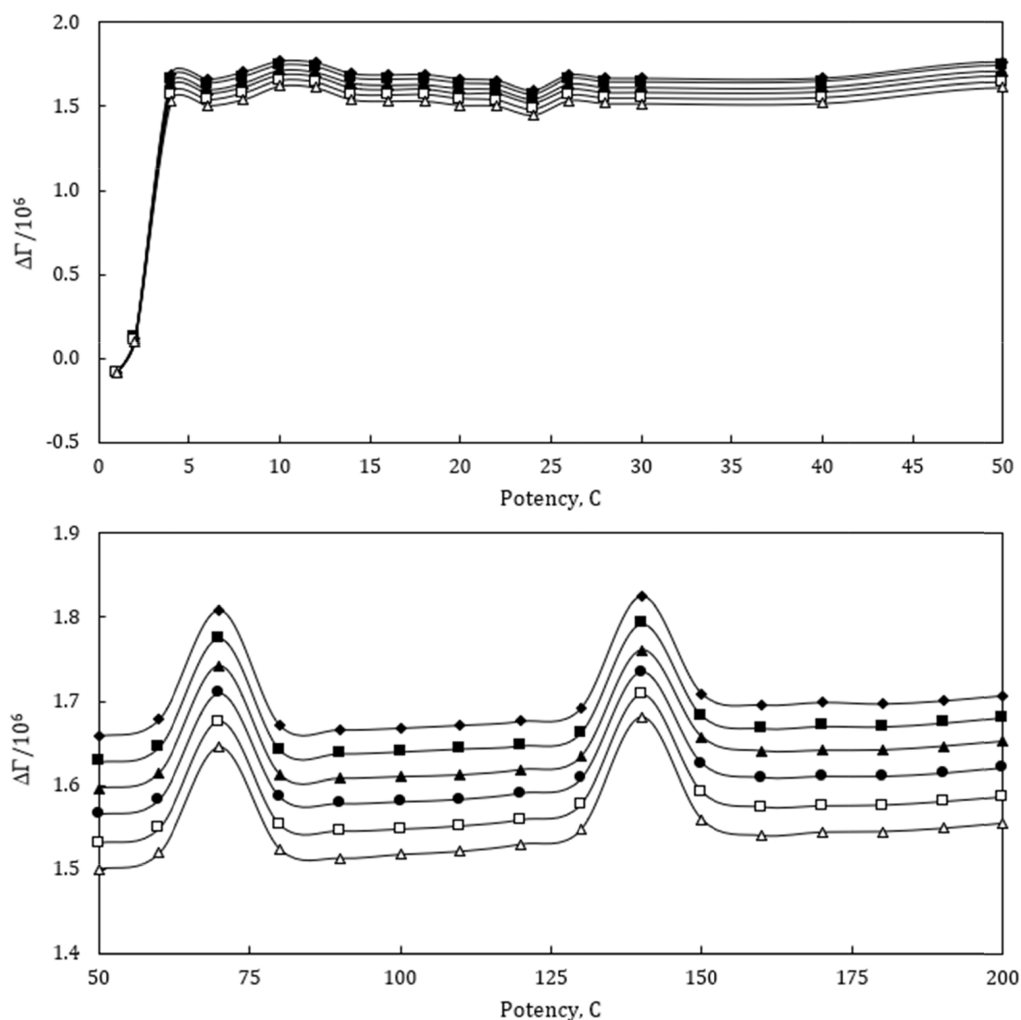


Fig. 10. Plots of deviations in pseudo-Grüneisen parameter, $\Delta\Gamma$ vs. potency, C of *Acidum citricum* for dilutions of *Acidum citricum* at temperatures, 293.15 K, \blacklozenge ; 298.15 K, \blacksquare ; and 303.15 K, \blacktriangle ; 308.15 K, \bullet ; 313.15 K, \square ; and 318.15 K, \triangle

various techniques, suggested a similar conclusion regarding the presence of medicine at high dilutions and found the potentization process affecting the behaviour of the homeopathic potencies. Bellare *et al.*¹ have also demonstrated the presence of nanoparticles of the starting materials and their aggregates even at extremely high dilutions using advanced techniques (TEM, SAED, and ICP-AES). They have revealed that despite large differences in the degree of dilution from 6C to 200C, no major differences in the nature of the particles (shape and size) of the starting material and their absolute concentrations (in pg/ml) were observed. Shah *et al.*⁴¹ used biophysical techniques such as UV and visible spectroscopy, fluorimetry, and Raman spectroscopy methods and observed that the highly potentised homeopathic preparations, that is, diluted beyond the Avogadro limit, exhibit physicochemical properties different from the control of un-potentised samples. The observed differences in

the physicochemical properties of potentised *Sulphur* in ethanol clearly indicated the presence of sulphur caused some modifications in solution. They also concluded that the presence of sulphur as well as the process of potentisation are responsible for the variation of the physicochemical properties of the potentised preparations. Cartright⁴² investigated the nature of homeopathic medicines using an experimental protocol in which solvatochromic dyes are used as molecular probes of serially diluted and agitated solutions and electronic spectroscopy detected the changes in the absorbance of this class of dyes across the visible spectrum in the presence of homeopathic potencies.

This study is one the series of studies undertaken understand physico-chemical properties of homeopathic medicine of different sources of homeopathic medicine, such as elements, organic, inorganic using the volumetric, acoustic and viscometric methods.⁹⁻¹¹ The variability in the results of these

studies needs to be closely analysed to find pattern of signals among closely related groups of drugs. Further biological study with potencies with different properties of the same medicine will help in the utilization of such data in quality control of medicines and effective potency for different medicines for clinical use.

Conclusion

The potencies 1C, 4C, 26C, 50C, 110C and 150C to 160C exhibit more compact solution structure as compared to other potencies and ethanol control. It is found that the interactions can exist between H⁺ and citrate ions and the molecular aggregates of water-ethanol, i.e., dissipative structures. Hence, these potencies might have diverse behaviour in terms of properties and efficacy when utilised in practice. It can be qualitatively concluded that even in high dilutions the molecules of *Acidum citricum* might be present in these homeopathic formulations, however it requires to be established from other more accurate spectroscopic and other techniques, like FTIR, Raman, NMR, DLS, or MD simulations.

Author contribution

Anil Kumar Nain: Concepts, Design, Definition of intellectual content, Literature search, Experimental studies, Data acquisition, Data analysis, Manuscript preparation, Manuscript editing, Manuscript review.

Neha Chaudhary: Design, Literature search, Experimental studies, Data acquisition, Data analysis, Manuscript preparation, Manuscript editing, Manuscript review.

Raj Kumar Manchanda: Concepts, Design, Definition of intellectual content, Literature search, Manuscript editing, Manuscript review.

Debadatta Nayak: Concepts, Design, Definition of intellectual content, Literature search, Manuscript editing, Manuscript review.

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Conflicts of interest

No conflict of interest to disclose.

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Étude du comportement physicochimique des dilutions homéopathiques d'*Acidum citricum* à des températures comprises entre 293,15 K et 318,15 K par des méthodes volumétriques, acoustiques et viscosimétriques.

Contexte : Les propriétés physicochimiques des médicaments homéopathiques gagnent en importance pour élucider leur comportement physicochimique (interactions) et, par conséquent, pour établir la présence de médicaments à des dilutions ultra-élevées. **Objectif :** La présente étude vise à identifier la présence de médicaments dans les médicaments ultra-dilués et à recueillir des preuves concernant le comportement physicochimique et les interactions prédominantes dans les dilutions d'*Acidum citricum*. **Matériel et méthodes :** Les densités (ρ), les vitesses ultrasoniques (u) et les viscosités (η) du témoin d'éthanol pur (91 % d'éthanol dans l'eau) et de 33 dilutions d'*Acidum citricum* de puissances comprises entre 1 °C et 200 °C à des températures de 293,15 K à 318,15 K et à pression atmosphérique. **Résultats :** Les données mesurées sont utilisées pour calculer les paramètres physicochimiques, à savoir la compressibilité isentropique (K_s), les longueurs libres intermoléculaires (L_f), l'impédance acoustique (Z), l'association relative (RA), le temps de relaxation (τ), l'absorption ultrasonore (a/f^2), le paramètre pseudo-Grüneisen (γ), les écarts de compressibilité isentropique (ΔK_s), les écarts de longueur libre intermoléculaire (ΔL_f), les écarts d'impédance acoustique (ΔZ), les écarts de viscosité ($\Delta \eta$) et les écarts du paramètre pseudo-Grüneisen ($\Delta \gamma$). Ces paramètres ont spécifié diverses tendances à certaines puissances de ces dilutions homéopathiques. **Conclusion :** Les résultats sont interprétés qualitativement au moyen des interactions prédominantes de ces dilutions d'*Acidum citricum*. Il est observé que les interactions peuvent avoir lieu entre les ions H^+ et citrate et les agrégats moléculaires d'eau-éthanol, c'est-à-dire des structures dissipatives. Les résultats ont montré que même à des dilutions élevées (50C, 110C, 150C et 160C), les molécules du médicament peuvent encore exister dans ces formulations homéopathiques.

Untersuchung des physikochemischen Verhaltens homöopathischer *Acidum citricum*-Verdünnungen bei Temperaturen von 293,15 K bis 318,15 K mittels volumetrischer, akustischer und viskosimetrischer Methoden

Hintergrund: Die physikochemischen Eigenschaften homöopathischer Arzneimittel gewinnen zunehmend an Bedeutung für die Aufklärung des physikochemischen Verhaltens (Wechselwirkungen) und damit für den Nachweis von Arzneimitteln in ultrahohen Verdünnungen.

Ziel: Ziel der vorliegenden Studie ist es, das Vorhandensein von Arzneimitteln in ultraverdünnten Arzneimitteln zu untersuchen und Hinweise auf das physikochemischen Verhalten und die vorherrschenden Wechselwirkungen in den Verdünnungen von *Acidum citricum* zu finden. **Material und Methoden:** Die Dichten (ρ), Ultraschallgeschwindigkeiten (u) und Viskositäten (η) der reinen Ethanolkontrolle (91 % Ethanol in Wasser) und 33 Verdünnungen von *Acidum citricum* mit Potenzen von 1 °C bis 200 °C bei Temperaturen von 293,15 K bis 318,15 K und bei Atmosphärendruck. **Ergebnisse:** Die Messdaten werden verwendet, um die physikochemischen Parameter zu berechnen, nämlich isentrope Kompressibilität (K_s), intermolekulare freie Längen (L_f), akustische Impedanz (Z), relative Assoziation (RA), Relaxationszeit (τ), Ultraschallabsorption (a/f^2), Pseudo-Grüneisen-Parameter (γ), Abweichungen der isentropen Kompressibilität (ΔK_s), Abweichungen der intermolekularen freien Länge (ΔL_f), Abweichungen der

akustischen Impedanz (AZ), Abweichungen der Viskosität (An) und Abweichungen des Pseudo-Grüneisen-Parameters (AF). Diese Parameter zeigten unterschiedliche Trends bei bestimmten Potenzen dieser homöopathischen Verdünnungen. **Schlussfolgerung:** Die Ergebnisse wurden qualitativ anhand der vorherrschenden Wechselwirkungen dieser *Acidum citricum*-Verdünnungen interpretiert. Es wurde beobachtet, dass Wechselwirkungen zwischen H*- und Citrationen und den Molekülaggregaten von Wasser und Ethanol, d. h. dissipativen Strukturen, stattfinden können. Die Ergebnisse zeigten, dass selbst in hohen Verdünnungen (50 °C, 110 °C, 150 °C und 160 °C) die Arzneimittelmoleküle in diesen homöopathischen Formulierungen noch vorhanden sein können.

293.15 K से 318.15 K के तापमान पर आयतनमितीय, ध्वनिक और श्यानमितीय विधियों द्वारा एसिडम साइट्रिकम होम्योपैथिक तनुकरणों का भौतिक-रासायनिकी अध्ययन

पृष्ठभूमि: होम्योपैथिक औषधियों के भौतिक-रासायनिक गुण आजकल विशेष महत्व प्राप्त कर रहे हैं क्योंकि वे उनके भौतिक-रासायनिक व्यवहार को स्पष्ट करने और अति-उच्च तनुकरणों में भी औषधि की उपस्थिति स्थापित करने में और सहायक होते हैं। **उद्देश्य:** इस अध्ययन का मुख्य उद्देश्य अति-तनुकृत औषधियों में एसिडम साइट्रिकम के अस्तित्व की संभावनाओं पता लगाना तथा इसके विभिन्न के तनुकरणों में भौतिक-रासायनिक व्यवहार और संभावित अंतःक्रियाओं से संबंधित साक्ष्य एकत्रित करना था। **सामग्री और विधियाँ:** अध्ययन में एसिडम साइट्रिकम के 1C से 200C तक की पोटेसी वाले कुल 33 तनुकरणों तथा शुद्ध इथेनॉल नियंत्रण (पानी में 91% इथेनॉल) का उपयोग किया गया। 293.15 K से 318.15 K के तापमान तथा वायुमंडलीय दबाव इन नमूनों की घनत्व (p), अल्ट्रासोनिक गति (u) और श्यानता (7) का मापन किया गया। **परिणाम:** प्राप्त डेटा का उपयोग विभिन्न भौतिक-रासायनिक मापदंडों की गणना हेतु किया गया। जैसे आइसेंट्रॉपिक संपीड़नशीलता (Ks), अंतर-आणविक मुक्त लंबाई (Lf), ध्वनिक प्रतिबाधा (Z) सापेक्ष संघ (RA), विश्राम समय (7), अल्ट्रासोनिक अवशोषण (a / f²) छद्म-गुनेसेन पैरामीटर (r), आइसेंट्रॉपिक संपीड़नशीलता में विचलन, (AKS), अंतर-आणविक मुक्त लंबाई में विचलन, (AL), ध्वनिक प्रतिबाधा में विचलन (AZ), चिपचिपाहट में विचलन (An) और छद्म-गुनेसेन पैरामीटर (AF) में विचलन। इन मापदंडों ने कुछ विशेष पोर्टेसीज़ पर भिन्न-भिन्न प्रवृत्तियों को होम्योपैथिक कमजोरियों उजागर किया। यह देखा गया कि H* और सीट्रेट आयनों और जल-एथेनॉल के आणविक समुच्चयों, यानी अपघट्य संरचनाओं के बीच परस्पर क्रियाएँ हो सकती हैं। परिणामों से पता चला है कि उच्च तनुकरणों (50°C, 110°C, 150°C और 160°C) में भी, इन होम्योपैथिक योगों में औषधि के अणु मौजूद रह सकते हैं।

Investigación del comportamiento fisicoquímico de diluciones homeopáticas de *Acidum citricum* a temperaturas de 293,15 K a 318,15 K mediante métodos volumétricos, acústicos y viscosimétricos.

Antecedentes: Las propiedades fisicoquímicas de los medicamentos homeopáticos han cobrado importancia para dilucidar su comportamiento fisicoquímico (interacciones) y, por consiguiente, para determinar la presencia del medicamento en diluciones ultraaltas. **Objetivo:** El presente estudio tiene como objetivo determinar la presencia del fármaco en medicamentos ultradiluidos y obtener evidencia sobre el

comportamiento fisicoquímico y las interacciones predominantes en las diluciones de *Acidum citricum*. **Material y métodos:** Se analizaron las densidades (ρ), velocidades ultrasónicas (u) y viscosidades (η) del control de etanol puro (91 % de etanol en agua) y 33 diluciones de *Acidum citricum* con potencias de 1 °C a 20 °C a temperaturas de 293,15 K a 318,15 K y a presión atmosférica. **Resultados:** Los datos medidos se utilizan para calcular los parámetros fisicoquímicos, a saber, compresibilidad isentrópica (K_s), longitudes libres intermoleculares (L_f), impedancia acústica (Z), asociación relativa (RA), tiempo de relajación (τ), absorción ultrasónica (a/f^2), parámetro pseudo-Grüneisen (r), desviaciones en la compresibilidad isentrópica (AKS), desviaciones en la longitud libre intermolecular (AL), desviaciones en la impedancia acústica (AZ), desviaciones en la viscosidad (A_n) y desviaciones en el parámetro pseudo-Grüneisen (AF). Estos parámetros especificaron diversas tendencias a ciertas potencias de estas diluciones homeopáticas. **Conclusión:** Los resultados se interpretan cualitativamente mediante las interacciones predominantes de estas diluciones de *Acidum citricum*. Se observa que las interacciones pueden tener lugar entre los iones H^* y citrato y los agregados moleculares de agua-etanol, es decir, estructuras disipativas. Los resultados especificaron que incluso en altas diluciones (50C, 110C, 150C y 160C) las moléculas del medicamento aún pueden existir en estas formulaciones homeopáticas.

使用体积法、声学法和粘度法研究柠檬酸 (*Acidum citricum*) 的物理化学行为

顺势疗法稀释液在 293.15 K 至 318.15 K 温度下的物理化学行为

背景: 顺势疗法药物的物理化学特性在阐明其物理化学行为 (相互作用) 以及确定超高稀释度药物存在方面日益重要。 **目的:** 本研究旨在探究超稀释药物中药物的存在, 并提供有关柠檬酸稀释液中物理化学行为和主要相互作用的证据。 **材料与方法:** 在大气压下, 在 293.15 K 至 318.15 K 的温度条件下, 测量纯乙醇对照 (91% 乙醇水溶液) 和 33 种柠檬酸稀释液 (效力范围为 1C 至 200C) 的密度 (ρ)、超声波速度 (u) 和粘度 (η)。 **结果:** 测量数据用于计算物理化学参数, 即等熵压缩率 (K_s)、分子间自由长度 (L_f)、声阻抗 (Z)、相对缔合 (RA)、弛豫时间 (τ)、超声波吸收 (a / f^2)、伪格吕奈森参数 (r)、等熵压缩率偏差 (AKS)、分子间自由长度偏差 (AL)、声阻抗偏差 (AZ)、粘度偏差 (A_n) 和伪格吕奈森参数偏差 (AF)。 这些参数在这些顺势疗法稀释液的不同效力下指定了不同的趋势。 **结论:** 通过这些柠檬酸稀释液中普遍存在的相互作用对结果进行了定性解释。 观察到相互作用可能发生在 H^* 和柠檬酸根离子以及水-乙醇分子聚集体 (即耗散结构) 之间。 结果表明, 即使在高稀释度 (50C、110C、150C 和 160C) 下, 药物分子仍可能存在于这些顺势疗法配方中。