



## Physico-Chemical Analysis on Some Polymers in Aqueous Surfactant Solution at Different Temperatures: Acoustical, UV and FTIR Studies

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The ultrasonic velocity, density and viscosity in mixtures of sorbitan sesquioleate and poly(vinyl pyrrolidone) (PVP), poly(vinyl alcohol) (PVA) and poly(ethylene glycol) (PEG) in different concentration ranges are measured at different temperatures (303, 313 and 323 K). From the experimental data, other related acoustical parameters such as adiabatic compressibility ( $\beta$ ), intermolecular free length ( $L_f$ ), internal pressure ( $\pi_i$ ), Rao's constant ( $R_a$ ), relaxation time ( $\tau$ ), acoustical impedance ( $Z_a$ ), absorption coefficient ( $\alpha/f^2$ ), free volume ( $V_f$ ), cohesive energy ( $C_E$ ) and solvation number ( $S_n$ ) have been evaluated. In aqueous solutions, surfactant molecule starts to aggregate and form micelle in concentration called as critical micelle concentration (CMC). It is one of the most important physical parameters of the surfactants. These results were interpreted in terms of surfactant-polymer complex reactions. The combinations of surfactant with polymers improve the desired properties of the product and the formation of complex was confirmed by UV and FTIR studies.

**Keywords:** Sorbitan sesquioleate, Polymers, Molecular interaction, Ultrasonic study.

### INTRODUCTION

The field of ultrasonics has grown enormously in scientific studies and has become a subject of active interest during the recent years. Ultrasonic technique has been found to be more accurate and comprehensive in understanding molecular interactions than the other methods [1]. Nonionic surfactant sorbitan sesquioleate is an emulsifier in cosmetic creams, pharmaceutical creams and ointments. An emulsifier is a thickening agent added to creams and ointments to thicken them and keep them stable [2]. Acoustical studies in polymers have been the subject of research in the previous years and carried out pioneering work on polymer solution using ultrasonic technique [3]. Polymers are used as binders for the formulation of pharmaceutical tablets, for moistening various personal care products, as food additives and adhesives [4].

The interactions between surfactant molecules and polymers in aqueous solutions are of importance to many applications. It is mainly predicted that the critical micelle concentration (CMC) is the most important factor in studies dealing with micellization of surfactants. In general, adsorption charac-

teristics at solid-liquid interfaces, the solubilization capacities in water for thinly soluble molecules and liquid-liquid interfacial tensions are altered by the related existence of surfactant and polymer molecules [5-8]. When the temperature value is increased, the value of critical micelle concentration is raised [9].

Various data on molecular interactions from ultrasonic velocity and its derived parameters shows the acoustical properties of surfactant with polymer solutions. Water soluble polymers with increased hydrophobicity, which might progress the soil eliminating capability of the polymer by interrelating with non-ionic surfactants by various mechanisms [10]. In the present study an attempt has been made to investigate the behaviour of sorbitan sesquioleate with aqueous solutions of PVP, PVA and PEG at different concentration and different temperatures 303, 313 and 323 K. The effect of sorbitan sesquioleate with PVP, PVA and PEG solution gives more information about complex reactions.

### EXPERIMENTAL

Sorbitan sesquioleate with PVP, PVA and PEG were obtained from Bio-corporals, Chennai. The experimental solutions were

prepared by adding a known weight of polymers (PVP, PVA and PEG) with different concentration of sorbitan sesquiolate to a fixed volume of water and attained a clear solution by stirring solution. Stock solution was prepared by using double distilled water. Since polymer solutions are very viscous, low concentrated solution is chosen for the study.

The ultrasonic velocity measurements in the sorbitan sesquiolate with PVP, PVA and PEG solutions were made in the ultrasonic interferometer (model F81, Mittal Enterprises, New Delhi, India) at a fixed single frequency of 2 MHz and at different temperatures (303, 313 and 323 K). The temperature was maintained constant by circulating water from a thermostatically controlled ( $\pm 0.1$  K) water bath. The values of densities at different temperatures were measured using specific gravity bottle by standard procedure and the viscosity was measured using Ostwald's viscometer with an accuracy of  $\pm 0.2$  % calibrated with double distilled water.

**Thermodynamic parameters:** By the use of empirical Jacobson's relations, various thermodynamic parameters [adiabatic compressibility ( $\beta$ ), intermolecular free length ( $L_f$ ), internal pressure ( $\pi_i$ ), acoustic impedance ( $Z_a$ ), cohesive energy ( $C_E$ ), Rao's constant ( $R$ ) and solvation number ( $S_n$ )] were calculated [11].

$$\text{Adiabatic compressibility } (\beta) = 1/U^2 \rho \quad (1)$$

where  $\rho$  is the density and  $U$  is the ultrasonic velocity of the medium using the Newton-Laplace equation.

$$\text{Intermolecular free length } (L_f) = K_T \beta^{1/2} \quad (2)$$

where  $K_T$  is the temperature dependent constant known as Jacobson's constant ( $K_T = 2.131 \times 10^{-6}$ ),  $\beta$  is the adiabatic compressibility

$$\text{Internal pressure } (\pi_i) = bRT [K \eta/u]^{1/2} \rho^{2/3} / M^{7/6} \quad (3)$$

(where,  $b$  stands for cubic packing)  $T$ -absolute temperature in Kelvin, where  $M_{\text{eff}}$  is the effective molecular weight of the mixture ( $M_{\text{eff}} = \sum m_i x_i$ , where  $m_i$  and  $x_i$  are the molecular weight and mole fraction of individual constituents, respectively  $K$  is a temperature independent constant which is equal to  $4.281 \times 10^9$  for all liquids,  $R$  is the universal gas constant,  $\eta$ -viscosity of the solution) [12].

$$\text{Rao's constant } (R_a) = (M/\rho) (u)^{1/3} \quad (4)$$

$$\text{Relaxation time } (\tau) = 4/3\beta \eta \quad (5)$$

$$\text{Acoustic impedance } (Z_a) = \rho \quad (6)$$

$$\text{Absorption coefficient } (\alpha/f^2) = (8\pi^2\eta/3\rho u^2) \quad (7)$$

$$\text{Free volume } (V_f) = (M_{\text{eff}} u / K \eta)^{3/2} \quad (8)$$

$$\text{Cohesive energy } (C_E) = V_f \pi_i \quad (9)$$

$$\text{Solvation number } (S_n) = \frac{M_2}{M_1} \left[ 1 - \left( \frac{\beta}{\beta_0} \right) \right] \left[ \frac{100-x}{x} \right] \quad (10)$$

where  $M_1$  is the molecular weight of the solvent and  $M_2$  is the molecular weight of the solute,  $\beta$  is the adiabatic compressibility of solution and  $\beta_0$  is the adiabatic compressibility of solvent.

## RESULTS AND DISCUSSION

The aqueous property of ultrasonic velocity, density and viscosity of measured parameters and acoustical parameters sorbitan sesquiolate + PVP, sorbitan sesquiolate + PVA and sorbitan sesquiolate + PEG at temperature 303, 313 and 323 K are measured (Table-1). The acoustical parameters such as adiabatic compressibility ( $\beta$ ), intermolecular free length ( $L_f$ ), internal pressure ( $\pi_i$ ), Rao's constant ( $R_a$ ), relaxation time ( $\tau$ ), acoustic impedance ( $Z_a$ ), absorption coefficient ( $\alpha/f^2$ ), free volume ( $V_f$ ), cohesive energy ( $C_E$ ) and solvation number ( $S_n$ ) have been computed.

Ultrasonic velocity of the binary mixture of non-ionic surfactant sorbitan sesquiolate with polymers increase with an increasing value of concentration of sorbitan sesquiolate. Initially the velocity increased and attains a maximum value at a particular concentration (critical micelle concentration (CMC)) and then the velocity value decreased. At CMC collection of monomers to form groups known as micelles take place hence at CMC ultrasonic velocity is maximum [13]. The increase in velocity is due to the intermolecular hydrogen bonding between the molecules. The variation of ultrasonic velocity with different concentration and temperature as shown in Fig. 1.

In the case of sorbitan sesquiolate with PVA mixture the CMC value occurs at 3:1 ratio. In other two mixtures of sorbitan sesquiolate + PVP and sorbitan sesquiolate + PEG the CMC value occurs at 4:1 ratio. Because in the PVA mixture, the concentration is very high due to its high molecular weight that is 89000 g/mol. So for PVA mixture CMC occurs quickly. Owing to the ultrasonic velocity mechanism should yield the increase in density with increase in concentration.

The viscosity of the binary mixture increases with increase in concentration. When a solute is added to a solvent, then the solvent becomes more viscous, particularly in the case of solid solute. So that, the mixture became thick liquid, less fluidity and hence with high viscosity. The increase in viscosity with concentration is showed inter chain forces over solvent-solvent forces and increases the relaxation time with concentration.

Adiabatic compressibility decreases with increase of concentration of non-ionic surfactant sorbitan sesquiolate, which shows in Fig. 2. This can be explained in terms of the electro-

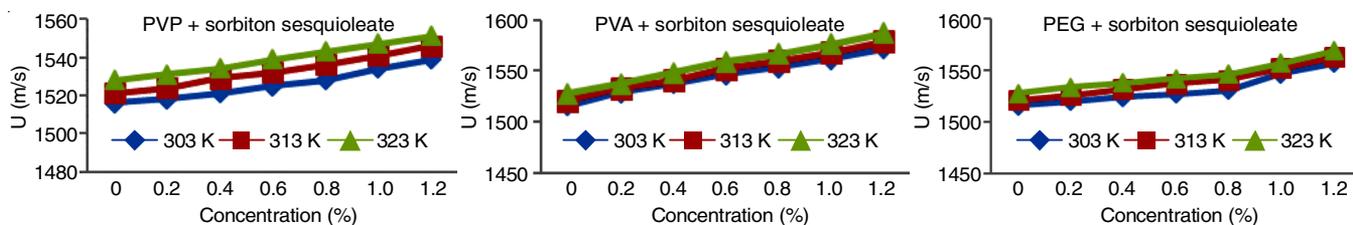


Fig. 1. Ultrasonic velocity vs. concentration of aqueous solution of some polymers + sorbitan sesquiolate at different temperatures

TABLE-1  
ULTRASONIC VELOCITY AND RELATED ACOUSTICAL PARAMETERS FOR SORBITAN  
SESQUIOLEATE IN SOME AQUEOUS POLYMERS AT DIFFERENT TEMPERATURES

Temp. (K)	Conc. (%)	U (m s <sup>-1</sup> )	$\rho$ (kg m <sup>-3</sup> )	$\eta \times 10^{-3}$ (Ns m <sup>-2</sup> )	$\beta \times 10^{-10}$ (N <sup>-1</sup> m <sup>2</sup> )	L <sub>r</sub> (Å)	$\pi_i \times 10^6$ (Pascal)	R	$\alpha f^2 \times 10^{-15}$ (Np m <sup>-1</sup> s <sup>2</sup> )	$V_f \times 10^{-12}$ (m <sup>3</sup> mol <sup>-1</sup> )	Cohesive energy $\times 10^{-6}$	$\tau \times 10^{-12}$ s	$Z_w \times 10^6$ (kg m <sup>-2</sup> s <sup>2</sup> )	S <sub>n</sub>
Sorbitan sesquioleate + PVP														
303	1:1	1521	1097	1.936	3.940	0.396	0.211	0.923	13.186	14.977	3.165	1.016	1.668	812
	2:1	1525	1109	1.984	3.877	0.392	0.216	0.909	13.262	15.463	3.350	1.025	1.691	1246
	3:1	1528	1118	2.147	3.830	0.390	0.227	0.897	14.153	17.308	3.944	1.096	1.708	1365
	4:1	1534	1126	2.372	3.774	0.386	0.241	0.886	15.343	20.044	4.847	1.193	1.727	1814
	5:1	1531	1132	2.586	3.768	0.387	0.255	0.876	16.737	22.556	5.759	1.299	1.733	1846
313	1:1	1529	1092	1.845	3.917	0.394	0.211	0.929	12.427	14.044	2.975	0.963	1.669	1102
	2:1	1532	1102	1.936	3.866	0.392	0.219	0.916	12.846	15.008	3.296	0.997	1.688	1337
	3:1	1536	1110	2.048	3.818	0.389	0.228	0.905	13.386	16.252	3.709	1.042	1.704	1450
	4:1	1541	1121	2.291	3.756	0.386	0.244	0.892	14.683	19.157	4.678	1.147	1.727	1940
	5:1	1536	1129	2.378	3.754	0.386	0.252	0.879	15.281	19.987	5.088	1.190	1.734	1947
323	1:1	1534	1088	1.658	3.905	0.394	0.206	0.933	11.099	12.022	2.482	0.863	1.668	1148
	2:1	1539	1097	1.719	3.848	0.391	0.212	0.921	11.302	12.643	2.686	0.881	1.688	1438
	3:1	1543	1106	1.827	3.797	0.388	0.221	0.909	11.822	13.787	3.053	0.924	1.706	1557
	4:1	1547	1114	1.975	3.750	0.386	0.232	0.898	12.590	15.423	3.587	0.987	1.723	1925
	5:1	1541	1121	2.133	3.756	0.386	0.244	0.887	13.671	17.062	4.177	1.068	1.727	1868
Sorbitan sesquioleate + PVA														
303	1:1	1538	1119	2.132	3.777	0.387	0.223	0.909	13.7694	17.617	3.933	1.073	1.721	3110
	2:1	1547	1131	2.347	3.694	0.383	0.236	0.896	14.7369	20.346	4.819	1.155	1.749	3165
	3:1	1554	1154	2.583	3.588	0.377	0.253	0.874	15.6817	23.442	5.931	1.235	1.793	3374
	4:1	1550	1169	2.792	3.560	0.376	0.267	0.857	16.863	26.014	6.958	1.325	1.811	3577
	5:1	1546	1178	2.842	3.551	0.376	0.273	0.845	17.1664	26.381	7.214	1.345	1.821	3627
313	1:1	1541	1112	1.942	3.786	0.388	0.219	0.915	12.5477	15.360	3.364	0.980	1.713	2886
	2:1	1552	1121	2.161	3.703	0.383	0.233	0.905	13.5582	18.063	4.209	1.066	1.739	3000
	3:1	1560	1139	2.326	3.607	0.378	0.245	0.887	14.143	20.148	4.944	1.118	1.776	3160
	4:1	1556	1148	2.573	3.597	0.378	0.261	0.874	15.6422	23.147	6.055	1.233	1.786	3221
	5:1	1552	1159	2.724	3.582	0.377	0.273	0.860	16.5301	24.899	6.799	1.300	1.798	3328
323	1:1	1548	1096	1.789	3.807	0.389	0.214	0.930	11.5695	13.674	2.931	0.908	1.696	2562
	2:1	1559	1115	1.938	3.690	0.383	0.226	0.911	12.0606	15.444	3.497	0.953	1.738	3106
	3:1	1567	1127	2.136	3.613	0.379	0.240	0.897	12.9509	17.850	4.292	1.028	1.766	3088
	4:1	1561	1138	2.375	3.606	0.378	0.249	0.882	14.4258	20.627	5.145	1.141	1.776	3130
	5:1	1557	1146	2.529	3.599	0.378	0.260	0.870	15.3719	22.382	5.835	1.213	1.784	3166
Sorbitan sesquioleate + PEG														
303	1:1	1524	1092	1.968	3.942	0.396	0.212	0.927	13.386	15.376	3.266	1.034	1.664	865
	2:1	1527	1096	2.046	3.913	0.394	0.218	0.919	13.785	16.209	3.539	1.067	1.673	947
	3:1	1531	1101	2.255	3.874	0.392	0.231	0.911	15.005	18.669	4.315	1.164	1.685	1060
	4:1	1547	1107	2.296	3.774	0.387	0.234	0.904	14.729	19.318	4.528	1.155	1.712	1858
	5:1	1541	1111	2.347	3.790	0.388	0.239	0.894	15.178	19.684	4.715	1.185	1.712	1722
313	1:1	1532	1087	1.845	3.919	0.395	0.211	0.933	12.411	14.068	2.971	0.964	1.665	1075
	2:1	1537	1091	1.961	3.879	0.393	0.219	0.925	13.015	15.359	3.370	1.014	1.676	1205
	3:1	1541	1096	2.147	3.842	0.391	0.231	0.917	14.074	17.514	4.054	1.099	1.688	1263
	4:1	1552	1102	2.215	3.767	0.387	0.236	0.909	14.136	18.394	4.354	1.112	1.710	1857
	5:1	1546	1107	2.296	3.779	0.387	0.243	0.898	14.757	19.138	4.665	1.156	1.711	1750
323	1:1	1538	1081	1.642	3.910	0.394	0.204	0.939	10.977	11.880	2.429	0.855	1.662	1152
	2:1	1542	1085	1.783	3.876	0.392	0.214	0.931	11.784	13.381	2.873	0.921	1.673	1211
	3:1	1546	1089	1.942	3.841	0.391	0.225	0.924	12.688	15.140	3.420	0.994	1.683	1239
	4:1	1557	1094	2.107	3.770	0.387	0.236	0.916	13.415	17.148	4.059	1.059	1.703	1806
	5:1	1553	1099	2.186	3.772	0.387	0.243	0.906	13.962	17.900	4.363	1.099	1.706	1779

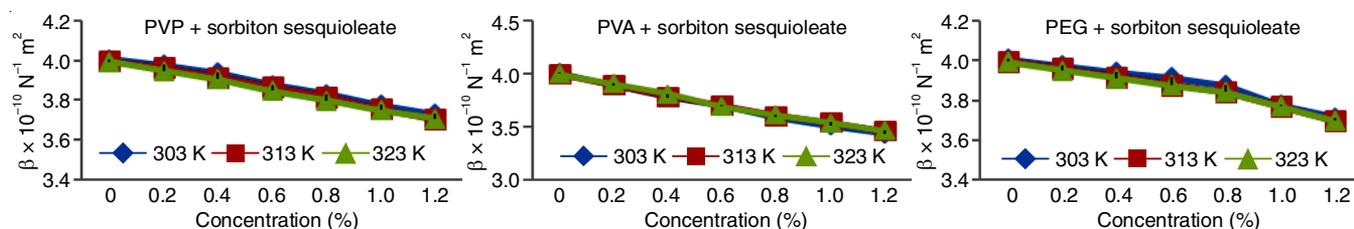


Fig. 2. Adiabatic compressibility vs. concentration of aqueous solution of some polymers + sorbitan sesquioleate at different temperatures

static effects of polymers on the surrounding molecules [14]. The adiabatic compressibility falls with increase in temperature. The soap molecules in dilute solutions ionize in cations and anions, due to the decrease in adiabatic compressibility. These ions in solution are bordered by a coating of solvent molecules, firmly sure and oriented towards the ions. The orientation of solvent molecules round the ions is attributed to the effect of electrostatic field of ions, which disturbs the internal pressure and drops the compressibility of the solution. The inter molecular free length depends upon adiabatic compressibility and shows a behaviour similar to that of compressibility and inversely to that of ultrasonic velocity. On increasing the concentration the inter molecular free length decreases. This shows that there is a structural rearrangement in the binary mixture and suggests the presence of strong solute-solvent interactions [15]. If the temperature increases, density and compressibility decreases and ultrasonic velocity increases. It shows that the interactions between solute and solvent molecules are significant. Due to thermal expansion of liquids an increase in temperature reasons the free length to increase which is similar to adiabatic compressibility.

The increase of acoustic impedance values with solute concentration can be recognized to the effective solute-solvent interactions. It is also observed that acoustic impedance value for sorbitan sesquioleate + PVA is greater than that for other two systems. The non-linear variation of Rao's constant ( $R_a$ ) with concentration forecast the strong intermolecular link between the molecules of sorbitan sesquioleate with polymers. The positive values of solvation number indicate the structure forming tendency of the polymers [16]. A change in post micellization area exhibited by the values of solvation number, which may be credited to greater consumption of solvent molecules. This reduces the repulsive forces acting between polar heads of ionic micelles [9]. The increase of internal pressure with concentration and the positive values of solvation number confirm an increase in cohesive force, which again support the structure forming tendency of solutes.

If the concentration of sorbitan sesquioleate is increased the absorption coefficient value is also increased. This is due to the fact that the maximum reaction takes place at higher concentration [17]. The result shows that the transfer of energies more probable between two different molecules than with the molecules of the same species [18]. Free volume is the null space between the molecules because of rough packing of molecules. Fig. 3 shows the variation of free volume with concentration and temperature. The increase of free volume shows that the strength of interaction increases with solute concentration. It represents that there is strong interaction between the solute and solvent molecules [19]. If the concen-

tration of sorbitan sesquioleate + polymer mixtures is increased, the cohesive energy also increased and this increment implies that the solute-solvent had a very strong bond in between them [20].

**UV analysis:** From the absorption study, the mixture of sorbitan sesquioleate + PVA has extremely good correlation  $R^2 = 0.986$ , but poor correlation in mixture of sorbitan sesquioleate + PVP,  $R^2 = 0.742$  and mixture of sorbitan sesquioleate + PEG,  $R^2 = 0.957$ . Aqueous mixture of sorbitan sesquioleate + PVA has high polarization power than the other two systems of sorbitan sesquioleate + PVP and sorbitan sesquioleate + PEG. In all the cases, polarization increases with increase in absorption as shown in Table-2.

Sample name	Concentration ( $\times 10^{-3}$ ) mol $m^{-3}$	Absorbance (Nm)
Sorbitan sesquioleate + PVP	0.0004	0.763
	0.0006	0.851
	0.0008	0.932
	0.0010	1.745
Sorbitan sesquioleate + PVA	0.0004	0.515
	0.0006	0.742
	0.0008	0.873
	0.0010	1.032
Sorbitan sesquioleate + PEG	0.0004	0.943
	0.0006	0.987
	0.0008	1.154
	0.0010	1.285

**FTIR analysis:** The Fourier transform infrared analysis was conducted to verify the possibility of interaction of chemical bonds between non-ionic surfactant with polymers. The FT-IR spectra of pure non-ionic surfactant (sorbitan sesquioleate) and sorbitan sesquioleate blended in the ratio 1:1 with PVP, PVA or PEG were recorded in the transmittance mode in the range of 4000-400  $cm^{-1}$  at a resolution of 1.0  $cm^{-1}$ .

The spectrum of sorbitan sesquioleate shows the characteristic absorption of band positions and intensities observed in FTIR spectrum. In sorbitan sesquioleate the peaks at 3536 and 3185  $cm^{-1}$  were due to the presence of stretching N-H group. The band at 2031  $cm^{-1}$  indicates the presence of  $C\equiv C$  group. The C-H stretching band was found in the range of 2915  $cm^{-1}$ . A band at 2915  $cm^{-1}$  in sorbitan sesquioleate shifted to 2968  $cm^{-1}$  in sorbitan sesquioleate + PVP, 2961 and 2943  $cm^{-1}$  in sorbitan sesquioleate + PVA was found to lie in the region of stretching of methyl and propyl groups and in the case of sorbitan sesquioleate + PEG the C-H stretching band at 2863  $cm^{-1}$  was found.

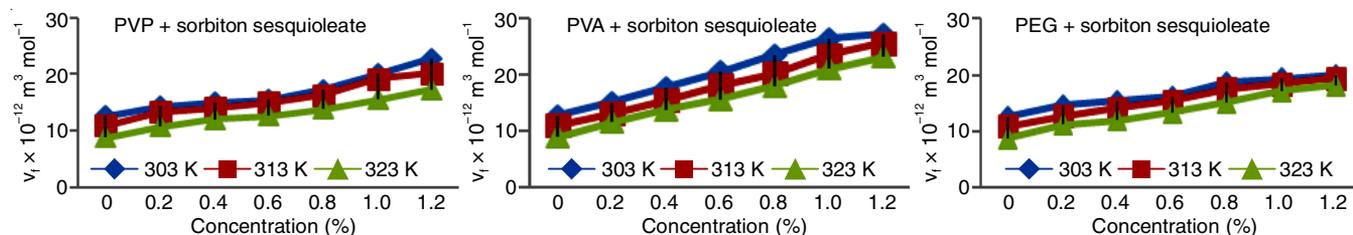


Fig. 3. Free volume vs. concentration of aqueous solution of some polymers + sorbitan sesquioleate at different temperatures

The intense peak at 1298  $\text{cm}^{-1}$  in pure PVP shifted to 1278  $\text{cm}^{-1}$  indicates the presence of C-O group in sorbitan sesquiolate + PVP. The peak at 2396  $\text{cm}^{-1}$  in pure PVA moved to 2301  $\text{cm}^{-1}$  indicates the presence of C=C band and 1372 to 1368  $\text{cm}^{-1}$ , 1096 to 1105  $\text{cm}^{-1}$  shows the presence of C-O for sorbitan sesquiolate + PVA. In sorbitan sesquiolate + PEG, 1540  $\text{cm}^{-1}$  lifted from 1698  $\text{cm}^{-1}$  in pure PEG mention C=C group. For sorbitan sesquiolate + PVP the band at 2843, 873  $\text{cm}^{-1}$ , the peak at 958  $\text{cm}^{-1}$  in sorbitan sesquiolate + PVA and 908, 841  $\text{cm}^{-1}$  at sorbitan sesquiolate + PEG show newly created C-H stretching vibration bands. In sorbitan sesquiolate + PEG the new peak at 1105  $\text{cm}^{-1}$  was assigned to C-O band. However, sorbitan sesquiolate with PVA revealed a significant chemical interaction which is realized by observing all spectral bands of sorbitan sesquiolate + PVA loaded. Besides, FTIR studies showed that the sorbitan sesquiolate + PVA distribution was homogeneous.

### Conclusion

The present study reveals that the intermolecular interaction of sorbitan sesquiolate with polymers (PVP, PVA and PEG) and the molecular association of sorbitan sesquiolate + PVA system is quite high compare to sorbitan sesquiolate + PVP and sorbitan sesquiolate + PEG systems. From UV analysis, the mixture of sorbitan sesquiolate + PVA has extremely good correlation  $R^2 = 0.986$  compared to other two mixtures. FTIR spectra, the result suggests that sorbitan sesquiolate + PVA blend has stronger intermolecular interaction than other blends. The present work clearly stated about that sorbitan sesquiolate + PVA has been used as a best additive and this composite had a lower foaming tendency due to lower surface tension and to make newer product.

### CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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