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Acoustical and Spectroscopic studies in aqueous solutions of polymer and dextrin's binary complex formation

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Abstract : The ultrasonic velocity, density, viscosity in mixtures of poly ethylene glycol (PEG) and dextrin's (Maltodextrin (MD), β-Cyclodextrin (β-CD) and amylose) in different concentration ranges are measured at three different temperatures 303, 313 and 323K. PEG is used to make emulsifying agents, soaps, detergents, ointments, plasticizers etc. Dextrins are used as a tablet disintegrating agent and use of such excipient in wet, dry, and directly compressible tablet formulations for pharmaceutical use. From the experimental data, other related acoustical parameters such as adiabatic compressibility (β), intermolecular free length (L_f), internal pressure (π_i), Rao's constant (R_a), relaxation time (τ), acoustical impedance (Z_a), absorption coefficient (α/f^2), free volume (V_f) and cohesive energy (C_E) have been evaluated. All these parameters have been discussed separately to throw light on the polymer – dextrin's interactions. These properties are attributed to solute-solvent interactions through hydrogen bonding. The FT-IR analysis also carried out for vibration assignment and conformation of compatibility.

Introduction

Polyethylene glycol (PEG) is one of the most common and the simplest synthetic water soluble polymers used by human beings in everyday life. It is mostly used in adhesives, coatings, sealants, photoresists, solder masks and photopolymers[1]. Polyethylene glycol is an aprotic and powerful diluents that is also used as solubilizing agent for plastic, textile, paper and paint stripping interactions[2]. Interaction of cyclodextrin with Polyethylene glycol enhances the polymer stabilization as well as solubilization[3]. β-Cyclodextrin is a circular molecule linked by α-1,4 bonds. Hydrophilic outer tails and hydrophobic inner cavities allow β-CD to easily form an inclusion complex with a diversity of molecules to increase their solubility[4,5,6].

β-Cyclodextrin are also act as a potential candidate for the drug carrier because of their ability to alter physical, chemical and biological properties of the guest molecules through the formation of inclusion complex[7].

Maltodextrin is a mixture of glucose, disaccharides and polysaccharides, obtained by the partial hydrolysis of starch. Maltodextrin is a flavorless, easily digested carbohydrate made from cornstarch. A maltodextrin is a short chain of molecularly linked dextrose (glucose) molecules, and is manufactured by regulating the hydrolysis of starch and also it is freely soluble in water. Amylose is a spiral polymer made up of D-glucose units. In the present paper an attempt has been made to study the acoustical properties of polymer and dextrins. PEG has variety of applications in pharmaceutical applications [8]. Acoustical properties of polymer solutions have shown that ultrasonic velocity and its wide derived parameters provide much information on molecular interactions, which are of utmost importance for process involving polymer production and their uses[9].

Preparation of sample

Polyethylene glycol with molecular weight of 1500, Maltodextrin, β-Cyclodextrin and amylose was obtained from Himedia, India. Doubled distilled water was used to prepare the stock solution. A 0.4% of PEG standard stock solution was prepared initially and different concentration of dextrins was added to dissolve in PEG to form binary mixtures.

Experimental Techniques

For this different dissolved ultrasonic velocities of solutions were measured using a single frequency continuous wave ultrasonic interferometer (Mittal type, India) to an accuracy of ±0.05% at a frequency of 2MHz at 303, 313 and 323K. The temperature of the samples were maintained constant to an accuracy of ±0.1K using a thermostatically controlled digital water bath. The densities of the solutions were measured using a specific gravity bottle with an accuracy of ±0.01 kgm^{-3}.The viscosity was measured using Ostwald's viscometer to an accuracy of ±0.2%. The FTIR spectra were collected for these samples using Fourier Transform Infra Red Spectrometer (Spectrum RX, Perkin Elmer). In case of powder samples of PEG and dextrin's, the powder samples were mixed with KBr powder to form a pellet and placed in a sample cup and measured [10]. All spectra were collected in the range (4000-400cm^{-1}). The KBr technique was used to prepare the samples for infrared spectroscopy measurements.

Computational aspects of thermodynamic parameters

Thermodynamic parameters such as adiabatic compressibility (β), intermolecular free length (L_f), internal pressure (π_i), acoustic impedance (z_a), cohesive energy (CE) and Rao's constant (R) were calculated from empirical Jacobson's relations [11].

Adiabatic compressibility $\beta = 1/U^2 \rho$ (1)

have been calculated from the U-ultrasonic velocity and ρ- density of the medium using the Newton –Laplace equation

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

Where K_T is the temperature dependent constant known as Jacobson's constant (K_T=2.131x10^{-6}), β is the adiabatic compressibility

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

(Where, b stands for cubic packing, which is assumed to be for all liquids,

T-absolute temperature in Kelvin, Where M_{eff} is the effective molecular weight of the mixture ($M_{eff} = \sum m_i x_i$ 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 4.281x10^9 [12] for all liquids, R is the universal gas constant, η-Viscosity of the solution).

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

Relaxation time $\tau = 4/3\beta\eta$ (5)

Acoustic impedance $z_a = \rho U$ (6)

Absorption coefficient $\alpha/f^2 = (8\pi^2\eta/3\rho U^2)$ (7)

Free Volume $V_f = (M_{eff} U/K\eta)^{3/2}$ (8)

Cohesive energy $CE = V_f \pi_i$ (9)

Result and discussion

The aqueous property of ultrasonic velocity, density and viscosity of measured parameters and acoustical parameters of PEG+ β-Cyclodextrin, PEG+Maltodextrin, PEG+Amylose at 303, 313 and 323K are

presented in tables 1-3. The acoustical parameters such as adiabatic compressibility (β), intermolecular free length (L_f), internal pressure (π_i), Rao's constant (R_a), relaxation time (τ), acoustical impedance (Z_a), absorption coefficient (α/f^2), free volume (V_f) and cohesive energy(CE) have been computed and are shown in table 1-3.

The measured velocity of 0.4% PEG and different concentration of β-Cyclodextrin, Maltodextrin, Amylose increases with increase in both temperature and concentration. The plot between the ultrasonic velocity and concentration potential shows that the ultrasonic velocity is found to change linearly with concentration and temperatures (fig. 1, 4 & 7). This linear increase suggests that there is strong solute-solvent interaction in the liquid solution of PEG+Maltodextrin, where as compare to PEG+ β-Cyclodextrin and PEG+Amylose. The increase in velocity of Maltodextrin suggests that disturb ion of Maltodextrin structure is enhanced further with the addition of PEG [13,14].

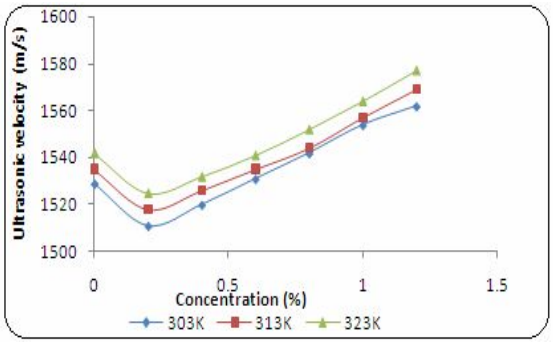


Fig:1.PEG+ β-Cyclodextrin (% Vs U)

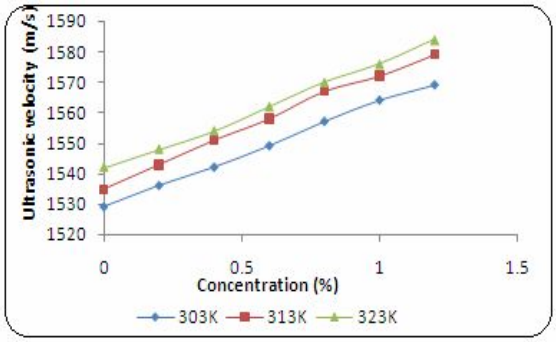


Fig:4.PEG+ Maltodextrin (% Vs U)

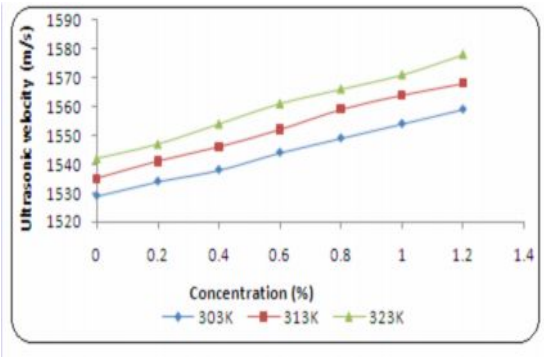


Fig:7.PEG + Amylose (% Vs U)

From the table (1, 2 & 3) the values of density increase with increases in concentration of PEG+ β-Cyclodextrin, PEG+Maltodextrin and PEG+Amylose, but it is found to decrease with increase in temperature. Viscosity values of PEG+Maltodextrin (from table 2) increases with increases in concentration than PEG+ β-Cyclodextrin and PEG+Amylose (from table 1 & 3).Viscosity values decreases with increase in temperature the study offer a convenient and simple way of investigating interaction phenomena in binary polymer systems [15] .Viscosity of a solvent or solution is a measure of cohesiveness or rigidity present between either ions (or) ion-solvent (or) solution. The viscosity increases linearly with concentration. This indicates that there exist a strong interaction between solute and solvent[16].

Adiabatic compressibility is the property of a substance capable of being reduced in volume by application of pressure, quantitatively the reciprocal of the bulk modulus. The adiabatic compressibility decreases with increase in concentration for all the three different temperature that shown in fig (2, 5 & 8). Decrease in adiabatic compressibility might be due to aggregation of solvent molecules around solute molecules [17]. The decrease of adiabatic compressibility with concentration of PEG+Maltodextrin solution studied here indicates the formation of a more number of tightly bound systems than the other two systems.

In the present investigation, the intermolecular free length of PEG+ β-Cyclodextrin, PEG+ Maltodextrin and PEG+Amylose is found to decrease with respect to the various concentration and increases with temperature. Decrease in free length is due to compression of liquid which indicates that the molecules are coming closer to each other; hence the intermolecular cohesion is stronger leading to strong molecular association.

Table 1: Ultrasonic velocity and related acoustical parameters for β cyclodextrin in aqueous PEG (0.4%) at different temperature

Temp K	Conc %	U ms^{-1}	ρ kgm^{-3}	η $x10^{-3}$ Nsm^{-2}	β $X10^{-10}$ $N^{-1}m^2$	Lf Å	π_i $X10^{10}$Pascal	R $X10^{-5}$	τ $X10^{-10}$	z_a $x10^6$ kgm^{-2} s^2	α/f^2 $X10^{-10}$	V_f $X10^{-10}$	Cohesive energy
303K	0	1529	1044	0.832	4.097	0.403	27.01	0.198	4.544	1.596	0.896	0.021	0.581
	0.2	1511	1052	0.857	4.163	0.407	2.455	1.565	4.756	1.589	0.938	0.456	1.120
	0.4	1520	1064	0.873	4.067	0.402	1.342	2.632	4.733	1.617	0.933	0.990	1.329
	0.6	1531	1081	0.886	3.946	0.396	0.972	3.468	4.661	1.655	0.919	1.510	1.468
	0.8	1542	1093	0.898	3.847	0.391	0.787	4.156	4.605	1.685	0.908	1.988	1.565
	1.0	1554	1104	0.914	3.750	0.386	0.679	4.727	4.679	1.715	0.901	2.403	1.633
	1.2	1562	1113	0.933	3.682	0.382	0.610	5.209	4.678	1.738	0.903	2.742	1.673
313K	0	1535	1038	0.779	4.088	0.403	26.849	0.199	4.245	1.593	0.837	0.023	0.641
	0.2	1518	1046	0.812	4.148	0.406	2.454	1.576	4.490	1.587	0.885	0.497	1.222
	0.4	1526	1054	0.845	4.074	0.402	1.353	2.660	4.589	1.608	0.905	1.045	1.415
	0.6	1535	1068	0.869	3.973	0.397	0.985	3.513	4.603	1.639	0.907	1.561	1.538
	0.8	1544	1079	0.886	3.887	0.393	0.800	4.212	4.591	1.665	0.905	2.033	1.627
	1.0	1557	1097	0.902	3.760	0.386	0.693	4.760	4.521	1.708	0.891	2.458	1.705
	1.2	1569	1104	0.917	3.679	0.382	0.620	5.259	4.497	1.732	0.887	2.833	1.757
323K	0	1542	1031	0.710	4.079	0.402	26.273	0.201	3.860	1.589	0.761	0.027	0.725
	0.2	1525	1039	0.764	4.138	0.405	2.440	1.589	4.214	1.584	0.831	0.549	1.340
	0.4	1532	1047	0.797	4.069	0.402	1.347	2.681	4.323	1.604	0.852	1.148	1.547
	0.6	1541	1056	0.832	3.987	0.398	0.985	3.557	4.422	1.627	0.872	1.676	1.651
	0.8	1552	1067	0.859	3.890	0.393	0.805	4.266	4.455	1.655	0.878	2.146	1.728
	1.0	1564	1079	0.881	3.788	0.388	0.698	4.847	4.449	1.687	0.877	2.564	1.790
	1.2	1577	1091	0.902	3.685	0.383	0.628	5.331	4.431	1.720	0.874	2.927	1.838

Table 2: Ultrasonic velocity and related acoustical parameters of Maltodextrin and aqueous solution of PEG (0.4%) at different temperature

Temp K	Conc %	U ms^{-1}	ρ kgm^{-3}	η $x10^{-3}$ Nsm^{-2}	β $X10^{-10}$ $N^{-1}m^2$	Lf Å	π_i $X10^{10}$ Pascal	R	τ $X10^{-10}$	z_a $x10^6$ kgm^{-2} s^2	α/f^2 $X10^{-10}$	V_f $X10^{-10}$	Cohesive energy
303K	0	1529	1004	0.832	4.097	0.403	27.017	0.198	4.544	1.596	0.896	0.021	0.058
	0.2	1536	1056	0.854	4.013	0.399	2.156	1.741	4.569	1.622	0.901	0.550	1.186
	0.4	1542	1069	0.875	3.934	0.395	1.063	3.208	4.588	1.648	0.905	1.356	1.144
	0.6	1549	1078	0.891	3.866	0.392	0.697	4.621	4.591	1.669	0.905	2.321	1.619
	0.8	1557	1085	0.913	3.801	0.389	0.579	5.988	4.626	1.689	0.912	3.350	1.741
	1.0	1564	1094	0.936	3.736	0.385	0.416	7.290	4.662	1.711	0.919	4.409	1.836
	1.2	1569	1107	0.953	3.669	0.382	0.348	8.505	4.661	1.736	0.919	5.523	1.926
313K	0	1535	1038	0.799	4.088	0.403	26.840	0.199	4.245	1.593	0.837	0.023	0.641
	0.2	1543	1047	0.808	4.011	0.399	2.149	1.759	4.320	1.615	0.852	0.601	1.293
	0.4	1551	1055	0.841	3.940	0.396	1.064	3.257	4.417	1.636	0.712	1.452	1.545
	0.6	1558	1066	0.872	3.864	0.392	0.705	4.682	4.492	1.660	0.886	2.418	1.706
	0.8	1567	1074	0.894	3.791	0.388	0.526	6.062	4.518	1.682	0.891	3.491	1.836
	1.0	1572	1083	0.915	3.736	0.385	0.421	7.376	4.557	1.702	0.898	4.597	1.937
	1.2	1579	1095	0.934	3.662	0.381	0.353	8.616	4.560	1.729	0.899	5.746	2.029
323K	0	1542	1031	0.710	4.079	0.402	26.273	0.201	3.860	1.589	0.837	0.027	0.725
	0.2	1548	1042	0.743	4.004	0.399	2.116	1.769	3.966	1.613	0.852	0.685	1.451
	0.4	1554	1052	0.784	3.936	0.395	1.057	3.268	4.113	1.634	0.871	1.617	1.710
	0.6	1562	1063	0.831	3.855	0.391	0.708	4.699	4.271	1.660	0.860	2.609	1.848
	0.8	1570	1070	0.866	3.791	0.388	0.532	6.088	4.376	1.679	0.891	3.672	1.955
	1.0	1576	1078	0.887	3.734	0.385	0.426	7.417	4.415	1.698	0.898	4.835	2.061
	1.2	1584	1084	0.908	3.676	0.382	0.356	8.713	4.450	1.717	0.899	6.024	2.146

Table 3: Ultrasonic velocity and related acoustical parameters in the amylose and aqueous solution of PEG (0.4%) at different temperature

Temp K	Conc %	U ms^{-1}	ρ kgm^{-3}	η $x10^{-3}$ Nsm^{-2}	β $X10^{-10}$ $N^{-1}m^2$	Lf Å	π_i $X10^{10}$ Pascal	R $X10^{-3}$	τ $X10^{-10}$	z_a $x10^6$ kgm^{-2} s^2	α/f^2 $X10^{-10}$	V_f $X10^{-10}$	Cohesive energy
303K	0	1529	1044	0.832	4.097	0.403	27.017	0.198	4.544	1.596	0.896	0.021	0.581
	0.2	1534	1061	0.863	4.005	0.399	2.146	1.753	4.607	1.627	0.908	0.550	1.180
	0.4	1538	1074	0.881	3.936	0.395	1.042	3.265	4.622	1.651	0.911	1.384	1.443
	0.6	1544	1082	0.897	3.876	0.392	0.675	4.760	4.635	1.670	0.914	2.408	1.625
	0.8	1549	1091	0.915	3.820	0.389	0.496	6.222	4.659	1.689	0.919	3.548	1.762
	1.0	1554	1099	0.926	3.767	0.387	0.390	7.663	4.650	1.707	0.917	4.831	1.885
	1.2	1559	1111	0.947	3.703	0.383	0.323	9.045	4.674	1.732	0.922	6.109	1.975
313K	0	1535	1038	0.779	4.088	0.403	26.849	0.199	4.245	1.593	0.837	0.023	0.641
	0.2	1541	1052	0.811	4.002	0.399	2.132	1.770	4.327	1.621	0.853	0.608	1.296
	0.4	1546	1065	0.837	3.928	0.395	1.041	3.298	4.383	1.646	0.864	1.506	1.569
	0.6	1552	1076	0.869	3.858	0.391	0.682	4.795	4.469	1.669	0.881	2.545	1.735
	0.8	1559	1082	0.892	3.802	0.389	0.502	6.287	4.521	1.686	0.891	3.722	1.868
	1.0	1564	1088	0.914	3.757	0.386	0.396	7.757	4.577	1.701	0.902	4.974	1.972
	1.2	1568	1097	0.932	3.707	0.384	0.327	9.178	4.606	1.720	0.908	6.311	2.067
323K	0	1542	1031	0.710	4.079	0.402	26.256	0.201	3.860	1.589	0.716	0.027	0.726
	0.2	1547	1046	0.752	3.994	0.398	2.105	1.784	4.004	1.618	0.789	0.685	1.443
	0.4	1554	1059	0.781	3.910	0.394	1.030	3.324	4.070	1.645	0.802	1.686	1.738
	0.6	1561	1065	0.827	3.853	0.391	0.679	4.856	4.247	1.662	0.837	2.767	1.880
	0.8	1566	1072	0.859	3.803	0.389	0.503	6.359	4.355	1.678	0.859	3.968	1.999
	1.0	1571	1080	0.885	3.751	0.386	0.399	7.830	4.425	1.696	0.872	5.260	2.101
	1.2	1578	1087	0.909	3.694	0.383	0.330	9.287	4.476	1.715	0.882	6.620	2.188

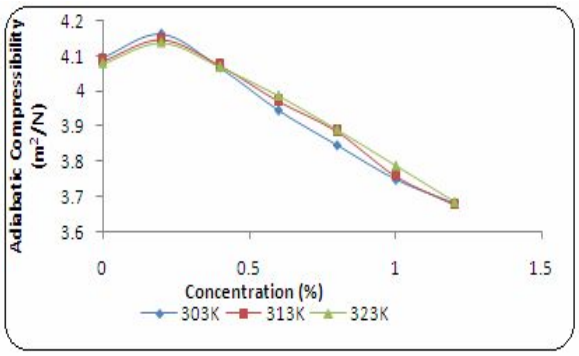


Fig:2.PEG+ β-Cyclodextrin (% Vs β)

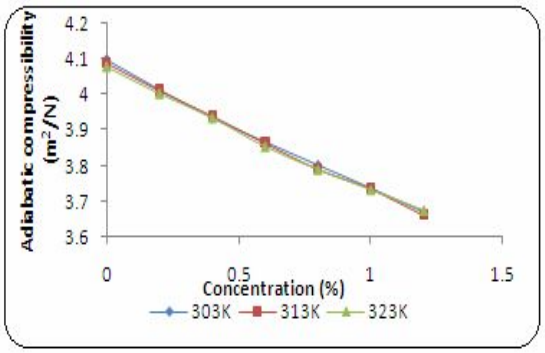


Fig:5.PEG+ Maltodextrin (% Vs β)

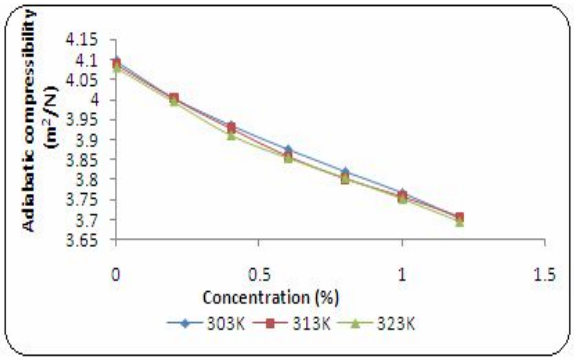


Fig:8.PEG+ Amylose (% Vs β)

Internal pressure of a solution is a single factor which appears to vary with intermolecular interactions namely solvation, quantum mechanical forces of dispersion, dielectric forces and solute-solvent interactions which play an important role in transport properties of solution. The internal pressure decreases with the rise in temperature because when the temperature is increased there is a tendency for the ions to move away from each other, reducing the possibilities for interaction, which may further reduce the cohesive forces and ultimately leads to decrease in the internal pressure.

Rao's constant is found to be increasing with increasing concentration and also with increasing temperature for PEG+ β-Cyclodextrin, PEG+Maltodextrin and PEG+Amylose. The increase of solute concentration is accompanied by an increase in relaxation time (τ) for all the three systems. The interaction causing associations between the solute molecules and the solvent molecules are responsible for the increase in the relaxation time. Such interaction is indeed also the reason for the increase in the ultrasonic velocity.

The acoustic impedance (Z_a) is governed by the internal and elastic properties of the medium. In the present investigation, acoustic impedance increases with increasing concentration and decreases with increasing temperature, which indicates the absence of complex formation and existence of solute – solute interaction is identified.

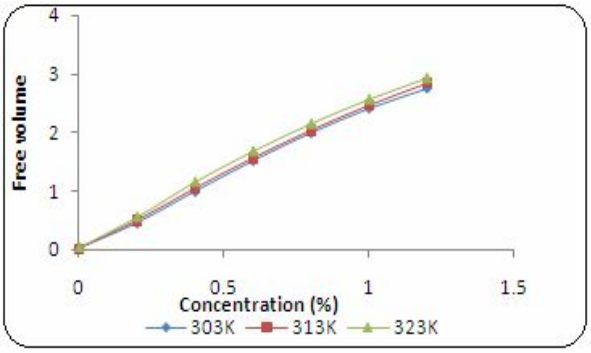


Fig:3.PEG+ β-Cyclodextrin (% Vs V_f)

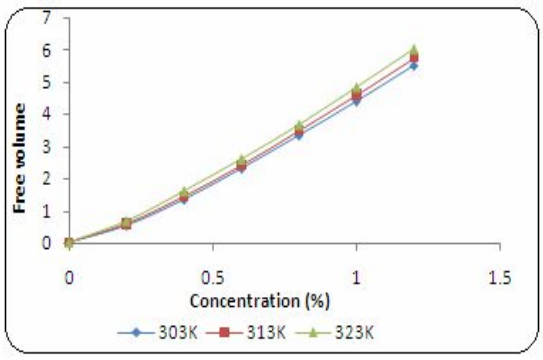


Fig:6.PEG+ Maltodextrin (% Vs V_f)

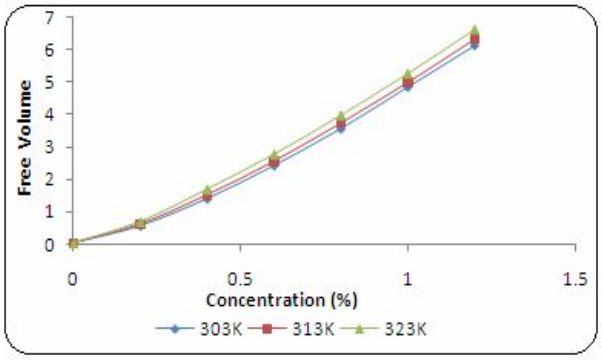


Fig:9. PEG+ Amylose (% Vs V_f)

Free volume is the average volume in which the centre of molecule can move due to the repulsion of the surrounding molecules. The free volume increases with increases in concentration (fig. 3, 6 & 9). The decrease in molecular association causes an increase in free volume. The increase in free volume is attributed to lose packing of the molecule inside the shield, which may be brought about my weakening of molecular interactions. Thus free volume is an inverse function of internal pressure[18].

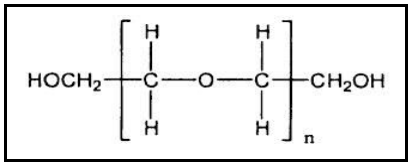
Cohesive forces are leading to the disturbtion of water structure in the systems containing the mixture of polymer molecules and the dextrin as a result of which water molecules are entrapped in voids formed during

FTIR spectroscopic study:

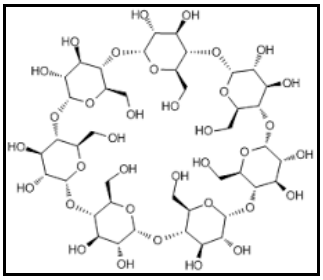
Fourier transform infrared is a powerful tool to study the formulation and possible interaction of polymer molecules in solid state. Polyethylene glycol is a polymeric linear chain consisting of ethylene oxide molecule in repetition. FTIR data of PEG, β-Cyclodextrin, Maltodextrin, Amylose, PEG+ β-Cyclodextrin, PEG+Maltodextrin and PEG+Amylose is shown in fig:1-3. From the FTIR spectra, the results suggest that PEG+Maltodextrin blend has stronger intermolecular interaction than PEG+ β-Cyclodextrin and PEG+Amylose blends. As shown in figure the PEG characteristics absorption peaks are 3431 cm^{-1} produced to polymeric chain and broad and steep peak produced due to prove the presence of OH group. A sharp intense band at 2887 cm^{-1} and 1104 cm^{-1} are due to stretching vibration of C-H and C-O bond. The symmetric vibrations are mostly displayed in the range of 1356 cm^{-1} and suggested –C-O-C.

Structure of samples:

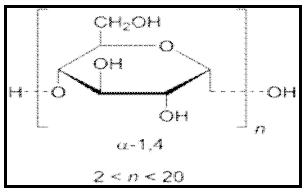
Poly ethylene glycol (PEG)



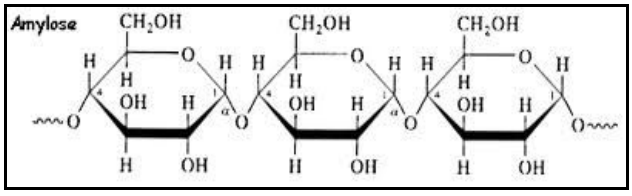
β- Cyclodextrin



Maltodextrin:



Amylose



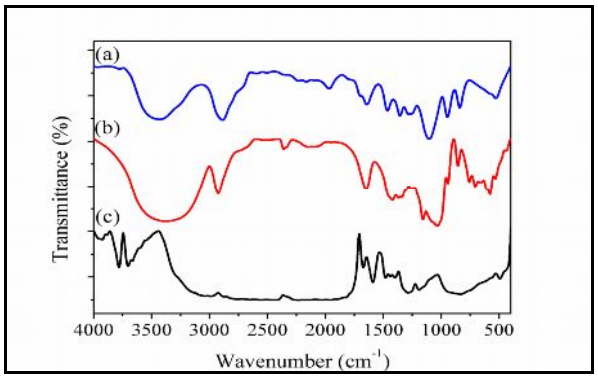


Fig.1 Overlapped FTIR spectra for a)PEG, b) β cyclodextrin and c) PEG+ β cyclodextrin blends

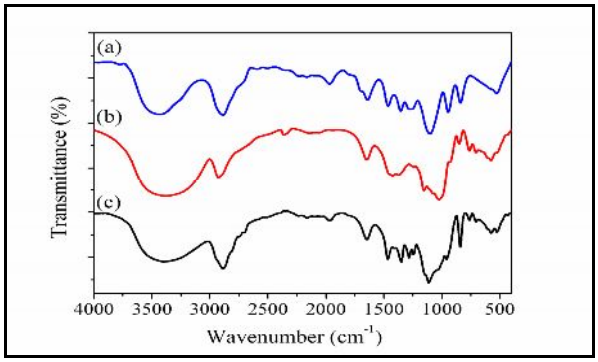


Fig.2 Overlapped FTIR spectra for a) PEG, b) Maltodextrin and c) PEG+Maltodextrin blends

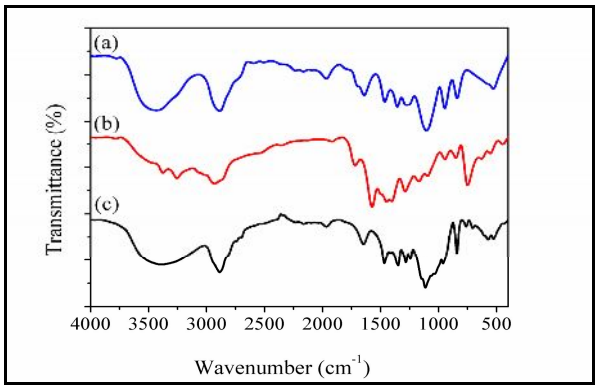


Fig.3 Overlapped FTIR spectra for a) PEG, b) Amylose and c) PEG+Amylose blends

A sharp band at 1642 cm^{-1} of PEG sifted to 1672 $cm^{-1}$and 1647 cm^{-1} in PEG+ β-Cyclodextrin, PEG+Amylose it indicate the presence of CO six member cyclic rings but in PEG+Maltodextrin indicate the interaction of presence of the stretching vibration of $\upsilon_{c\text{-}o}$ bond. The shift in the peaks associated with the dextrin group indicates the strength of bond.

Conclusion

In the present study,we confirmed that intermolecular attraction of PEG and different types of dextrin groups. The measured values, acoustical and FTIR data showed the solute-solute interaction was favored. Among the three different dextrin groups the potency of interaction was more in PEG+Maltodextrin compared to other two. Finally we conclude that PEG+ Maltodextrin has been used as a best additive in pharmaceutical, food industry and in biodegradability. This paper highlights the solute-solute interaction of polymer and dextrin groups.

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