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## Original Research Article

# Ultrasonic and spectroscopic investigation of aqueous polyvinyl alcohol (PVA) solutions

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### ABSTRACT

Using distilled water as solvent, different weight percentages of polyvinyl alcohol solutions were prepared and were then examined using the FTIR, UV-visible, and ultrasonic pulse echo techniques. In physics, ultrasound is a vital and efficient research tool. Understanding the physicochemical behavior of liquids and determining the nature of molecular interactions have become increasingly important goals of the ultrasonic study of liquid mixtures. Additionally, it offers a powerful and trustworthy instrument for examining ultrasonic properties of polymer solution in the context of phase separation research. Ultrasonic velocity ( $u$ ), Density ( $\rho$ ) and viscosity ( $\eta$ ) for aqueous polyvinyl alcohol have been measured at 2 MHz and at temperature range 288K-308K and at concentration range 0.05wt% to 0.3 wt% by pulse echo technique. From this data, acoustic parameters such as adiabatic compressibility ( $\beta_a$ ), acoustic impedance ( $Z$ ), relaxation time ( $\tau$ ) and free length ( $L_f$ ) were calculated for aqueous PVA solutions. The results are interpreted as per the molecular interactions in the aqueous PVA solutions and compared with the results obtained from FTIR and UV- visible spectroscopy. PVA can be employed as a tablet binder in the pharmaceutical industry due to the interactions observed in the solutions. The strength of molecular interactions in aqueous PVA solutions is indicated by thermo acoustic parameters including ultrasonic velocity, density, viscosity, adiabatic compressibility, acoustic impedance, relaxation time, free length, etc.

## 1. Introduction

Many researchers have become interested in polyvinyl alcohol (PVA) because of its exceptional qualities, including hydrophilicity, biodegradability, biocompatibility, and nontoxicity. It is a non-toxic, water-soluble polymer that has been extensively used in numerous research fields [1-2], including biomedical and drug delivery systems [3]. Additionally, it possesses exceptional resistance to oil, grease, and solvents as well as film-forming and emulsifying properties. Due to its high hydrophilicity and processability, it might be combined with various polymers. High tensile strength, abrasion resistance, and oxygen barrier characteristics can be found in PVA films [3]. It is a potentially useful material with good charge storage, high dielectric strength, and dopant-dependent electrical and optical properties. It too has a backbone made of carbon chains with hydroxyl groups joined by methane carbon. These OH groups can serve as a hydrogen bonding source, which helps to create polymer blends and composites. One of the key elements affecting PVA's properties and uses is the water absorption it experiences. Additionally, PVA has a high density of hydroxyl groups, which act as cross-linking sites after being exposed to radiation, chemicals or heat [4]. One of the main areas of study in both fundamental and practical aspects has been the

spectroscopic and ultrasonic analysis of polymer solutions. A spectroscopic investigation that demonstrates substantial results on the polymer solution's spectroscopic and ultrasonic properties is required to modify and improve its properties.

## 2. Materials and methods

### 2.1 Materials

The polyvinyl alcohol solutions were made by adding a predetermined amount of water with a given weight of PVA. Then stirred it till to obtain clear solution. Then, the concentration was expressed in weight percent. Its range was set at 0.05 weight percent to 0.3 weight percent ( i.e. 0.05wt%, 0.1 wt%, 0.15wt%, 0.2wt%, 0.25wt%, and 0.3 wt%).

### 2.2 Methods

Using the MHF-400 pulser receiver at 2 MHz and a temperature range of 288 K to 308 K, the ultrasonic velocity was measured. Viscosity were measured by Oswald's viscometer's and density by Picknometer. With a precision of 0.1K, the temperature was maintained using a thermostatically regulated water circulation system with a Plasto Craft LTB -10 thermostat. The standard formulae shown below were used to determine various thermos-acoustical properties using experimental data on ultrasonic velocity, density, and viscosity [5-6].



(Eq. 1) to determine ultrasonic velocity.

$$u = (2d / t) \text{ m/sec,} \tag{1}$$

where,  $d$  is the separation between transducer and reflector and  $t$  is the travelling time period of ultrasonic wave.

(Eq. 2) to determine density

$$\rho = \frac{M_l}{M_w} \rho_w \text{ kg m}^{-3} \tag{2}$$

where,  $M_l$  and  $M_w$  are the mass of liquid and water respectively.  $\rho_w$  is the density of water.

(Eq. 3) to determine viscosity

$$\eta = \frac{\rho_l t_l}{\rho_w t_w} \eta_w \tag{3}$$

where  $\rho_l$  is the density of experimental liquid,  $\rho_w$  is the density of water,  $\eta_w$  is the viscosity of water,  $t_l$  is the time required to experimental liquid to flow from mark A to B in viscometer, and  $t_w$  is the time of flow of water.

(Eq. 4) to determine adiabatic compressibility

$$\beta_a = \frac{1}{u^2 \rho} \tag{4}$$

(Eq. 5) to determine acoustic impedance

$$Z = \rho \cdot u \tag{5}$$

(Eq. 6) to determine relaxation time

$$\tau = \frac{\eta}{\rho} \cdot \beta_a \tag{6}$$

(Eq. 7) to determine free length

$$L_f = K_j \beta_a^{1/2} \tag{7}$$

where  $K_j$  stand for Jacobson's constant.

### 2.3 Characterizations

In present work author used UV spectrophotometer (UV-1800 pc) of Shimadzu Company for UV-vis characterization and Bruker Alpha FTIR instrument No. 10066587 for FTIR characterization and MHF-400 pulser receiver for ultrasonic characterization by pulse echo technique.

## 3. Results and discussion

### 3.1 Ultrasonic analysis

The following Figures 1(a), 1(b), 1(c), 1(d), 2(a), 2(b) and 2(c) discuss how thermo-acoustical properties change for various concentrations (wt%) at temperatures between 288 K and 308 K at 2MHz frequency:

The variation in ultrasonic velocity and density with concentration (wt%) is depicted in Figure 1(a) and 1(b). Viscosity ( $\eta$ ) versus concentration (wt%) is plotted in Figure 1 (c).

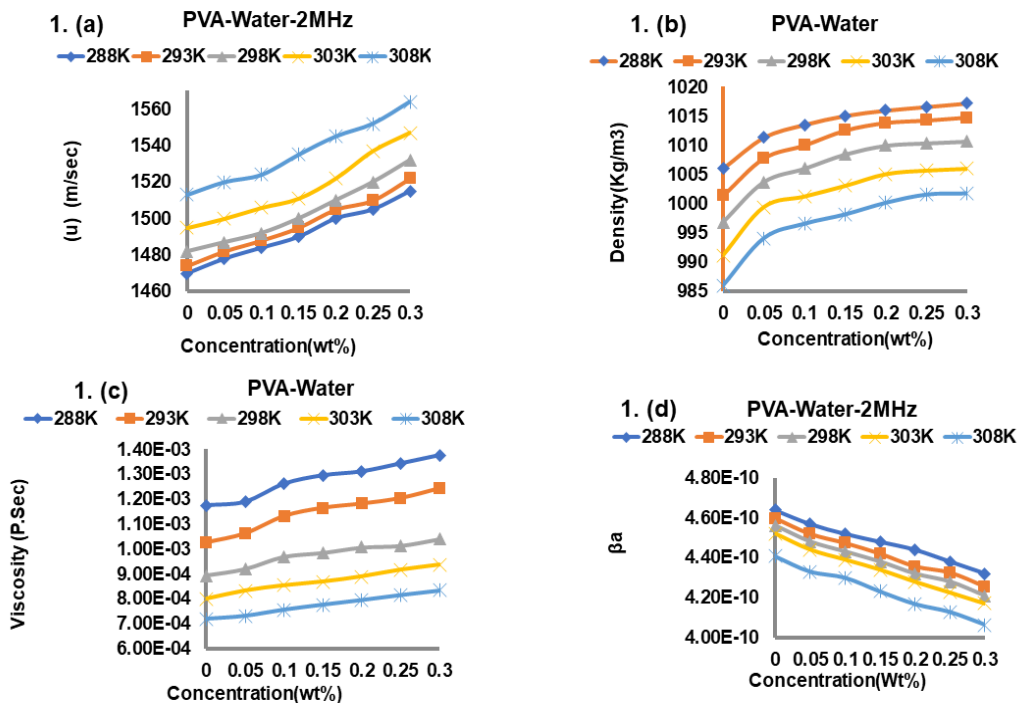


Figure 1: (a) ultrasonic velocity, (b) density, (c) viscosity, (d) adiabatic compressibility

It has been found that viscosity marginally increases as polyvinyl alcohol's weight percentage in water rises. According to Kauzman and Eyring, a mixture's viscosity is greatly influenced by its entropy, which is connected to both the liquid's structure and the interactions between its

constituent molecules. As a result, molecular interaction as well as molecule size and shape have an impact on viscosity. In the study of molecular interaction, the measurement of viscosity in binary mixtures provides some trustworthy

information. The strength of the molecular interactions between interacting molecules is determined by viscosity [7].

The viscosity of the polyvinyl alcohol + water system is increased by the dipole-dipole interactions of the permanent dipoles in component molecules. The metric for intermolecular association, dissociation, or repulsion is called adiabatic compressibility. It also controls how the molecules that make up the parts are oriented.

The fluctuation of adiabatic compressibility ( $\beta_a$ ) vs concentration (wt%) is shown in Figure 1 (d). It has been found that as the concentration (wt.%) of polyvinyl alcohol in water increases, adiabatic compressibility decrease, indicating a significant intermolecular contact between the component molecules in this system and an associating property. It is obvious that several closely bound systems have formed in the PVA + Water system based on the rapid decrease in adiabatic compressibility with concentration increase. This might be brought on by a more rigid liquid structure linked to PVA's hydrogen bonds in water. Solvent molecules have been discovered to cause such a drop in compressibility in the

solution [7]. According to Kiyohara and Benson, adiabatic compressibility is the product of a number of conflicting effects. The creation of hydrogen bonds between constituent molecules, interstitial accommodation, or strong dipole-dipole interactions all result in more compact structures and lower adiabatic compressibility. The observed increase in degree of connection in the component molecules is indicated by the decrease in adiabatic compressibility with concentration. Therefore, when concentration increases, the intermolecular distance reduces. The main factor that alters with structure and affects ultrasonic velocity is compressibility [7-8]. There is infinite contraction in the component molecules, as evidenced by a decrease in adiabatic compressibility. The molecules pack more tightly as adiabatic compressibility declines, which reduces the intermolecular free length. Through hydrogen bonds, the strong molecular connection between dissimilar molecules is strengthened as adiabatic compressibility increases. It is discovered that adiabatic compressibility ( $\beta_a$ ) is practically reciprocal to acoustic impedance ( $Z$ ).

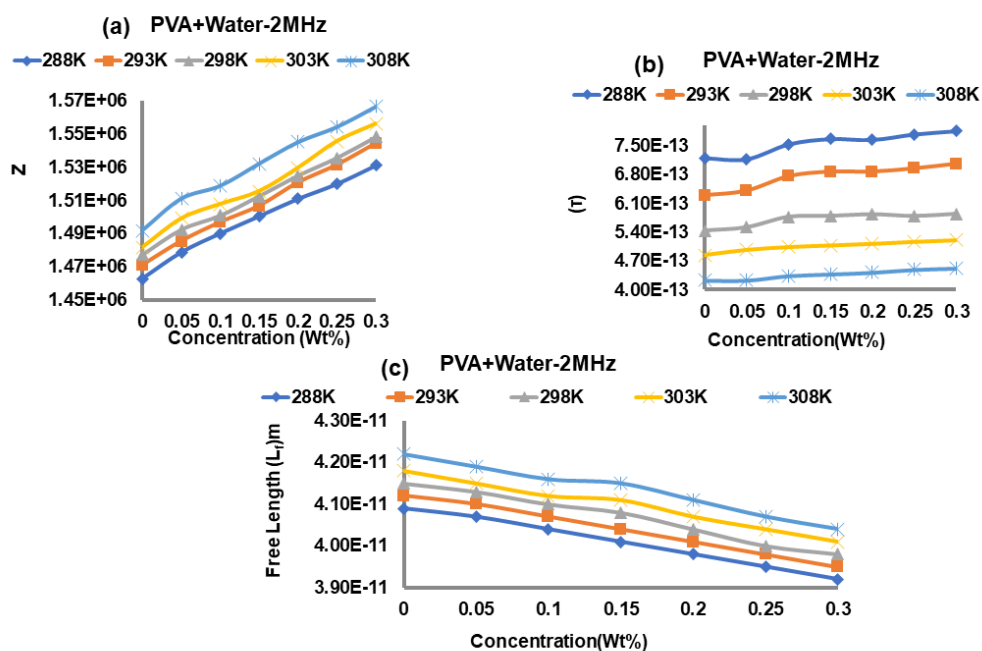


Figure 2: (a) Acoustic impedance, (b) Relaxation time, (c) Free length.

Acoustic impedance variation versus concentration can be seen in Figure 2(a). It has been found that when concentration (wt%) increases, so do the values of acoustic impedance. Because ultrasonic velocity increases with concentration, it satisfies the theoretical requirements. The decrease in intermolecular distance and relative decrease in gaps between the component molecules caused by intermolecular contact can be utilized to explaining the acoustic impedance ( $Z$ ) increases with concentration. This implies that significant interactions are also present in this liquid system.

The Figure 2(b) shows the relationship between relaxation time and concentration (wt%). The fact that relaxation time slightly increases as polyvinyl alcohol content in water rises indicates that polyvinyl alcohol molecules are highly stable. Since polyvinyl alcohol has a tight structure and unstable

molecules, the relaxation time is prolonged. Both the translational and the vibrational degrees of freedom can transmit energy, and both of these degrees are involved in the activity that is being seen [8]. Its behavior is determined by the liquid solution's viscosity and adiabatic compressibility. In this system, viscosity is crucial for lengthening relaxation times as concentration increases. With a rise in focus, the relaxation time rises. Due to its exceptional hydration, the solution is highly organized, and as a result, it is more likely to absorb ultrasonic energy[8-9]. Always expect that the mobility of the molecules in the solvent that is closest to the polymers will be lower than that of the molecules in pure solvent.

The curve of free length against concentration is shown in Figure 2(c). A measure of the intermolecular attraction between the constituents in binary mixes is the free length ( $L_f$ )

of a system. The intermolecular connection has weakened, as evidenced by the growth in free length. According to graphic behavior, free length decreases as polyvinyl alcohol concentration in water rises. Dipole-dipole interactions, hydrogen bonding-mediated association, or interstitial accommodation of dissimilar molecules if their sizes differ are all causes of a decrease in free length. Due to imperfect symmetry and a reduction in the amount of accessible space between the component molecules, free length decreases with concentration. Free length shortens as ultrasonic velocity quickens, exhibiting an inverse tendency that is well in line with the demands of theory. The decrease in free length as PVA concentration in water rises suggests that there are strong interactions between the solute and solvent, which may be supporting the behavior of the molecules in the binary solution that promotes structure. This results in an improvement of the closed structure, or an increase in the tightly packed structure of the component molecules. The increase in dipolar

association, which creates strong hydrogen bonds in the molecules of the liquid mixes, may be the cause of the decrease in free length. Free length shortens as ultrasonic velocity quickens, exhibiting an inverse tendency that is well in line with the demands of theory. The decrease in free length as PVA concentration in water rises suggests that there are strong interactions between the solute and solvent, which may be supporting the behavior of the molecules in the binary solution that promotes structure. This results in an improvement of the closed structure, or an increase in the tightly packed structure of the component molecules. The increase in dipolar association, which creates strong hydrogen bonds in the molecules of the liquid mixtures, may be the cause of the decrease in free length [7-9].

### 3.2 FTIR analysis

FTIR analysis is shown in Figure 3.

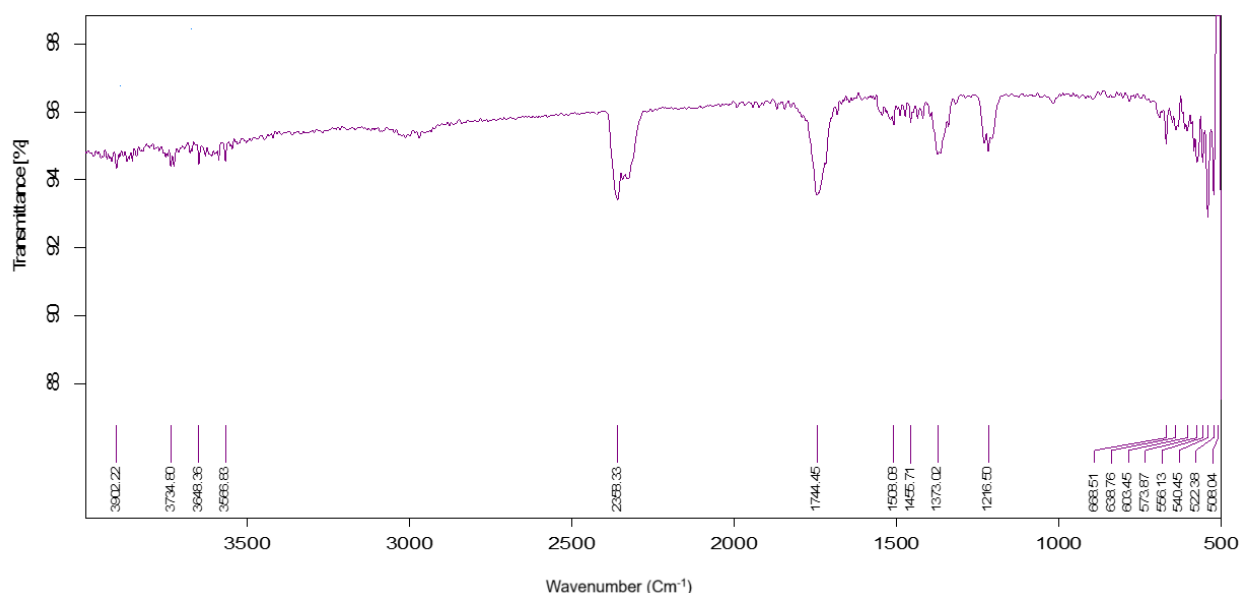


Figure 3: FTIR of aqueous PVA.

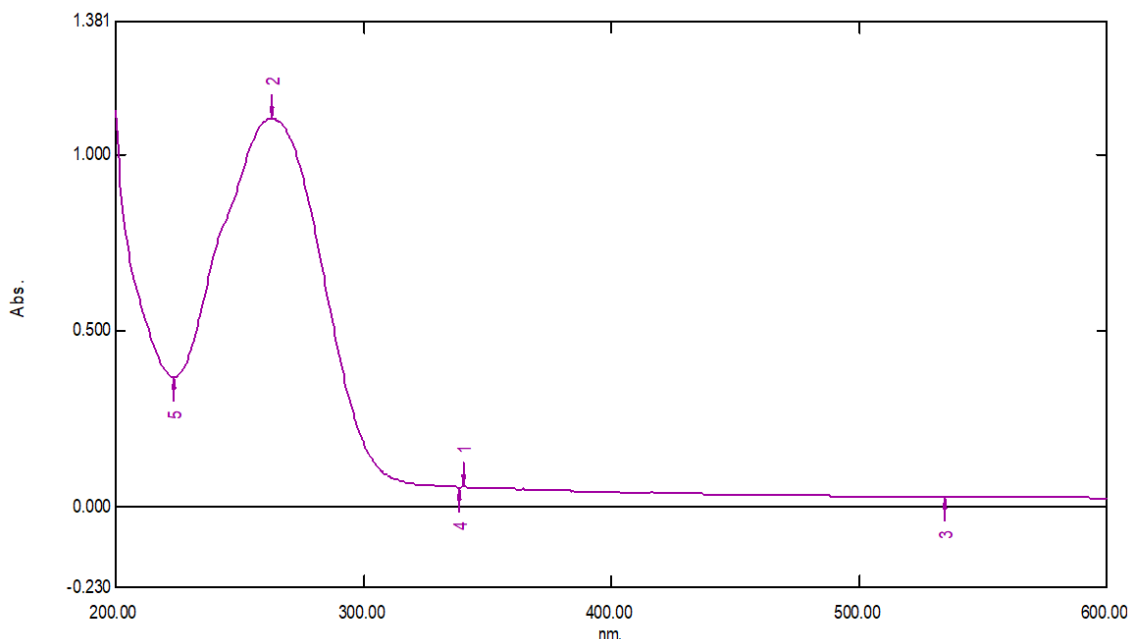
FTIR spectroscopy is a useful technique for identifying and investigating the presence of various functional groups in polymer solutions. The infrared spectra of various crystalline polymers have been extensively examined by polymer spectroscopists, but very little has been done to analyze the basic vibrations of organic polymers and copolymers. Because synthetic rubbers made of polyvinyl alcohol have such a wide range of industrial applications, it has been regarded as a significant polymer. The goal of the current inquiry is to examine the vibrational spectra of a polyvinyl alcohol polymer solution. Environmentally friendly and water soluble synthetic polymer with superior film-forming, emulsifying, and solvent- and oil-resistance capabilities is polyvinyl alcohol (PVA). Different weight percents of polymer solutions were created. Through FTIR spectroscopy, each material is examined for structural characteristics and miscibility behavior. We used a 1 (wt.%) aqueous PVA solution for the current FTIR investigation. The following graphic compares the FTIR spectra of aqueous Polyvinyl alcohol (PVA) in a 1 weight percent solution to that of pure PVA. The spectrum demonstrates the features of several functional groups,

absorption, stretching, and bending frequencies. The spectral peak at  $3648.36\text{ cm}^{-1}$  for the spectrum of an aqueous PVA solution is due to OH bonding. =C-H stretching (Olefinic) is thought to be responsible for the peak at  $3000\text{ cm}^{-1}$ . C=O bending may be seen in the typical band at  $1744.45\text{ cm}^{-1}$ . The C-C stretching (Aliphatic) and shifting are credited with causing the peak at  $1373.02\text{ cm}^{-1}$ . C-O stretching and shifting is attributed to the spectral peak at  $1216.50\text{ cm}^{-1}$ . While the spectral peak at  $3600\text{ cm}^{-1}$  in the spectrum of pure PVA is attributable to OH bonding. Between Pure PVA and an aqueous solution of 1 weight percent PVA, there is a slight shift in the OH bonding. Additionally, the peak for Pure PVA at  $2917\text{ cm}^{-1}$  is due to =C-H stretching. As shown by the typical band at  $1690\text{ cm}^{-1}$ , C=O bending. C-C stretching is also thought to be responsible for the peak at  $1373\text{ cm}^{-1}$ . The  $1081\text{ cm}^{-1}$  spectral peak is attributed to C-O stretching. Therefore, there is a slight wavelength shift in each peak. This indicates that there were no new bonds that were comparable to those in pure PVA were discovered in the aqueous PVA 1(Wt.%) solution. It is because the aqueous PVA solution has poor molecular interaction [10-13].

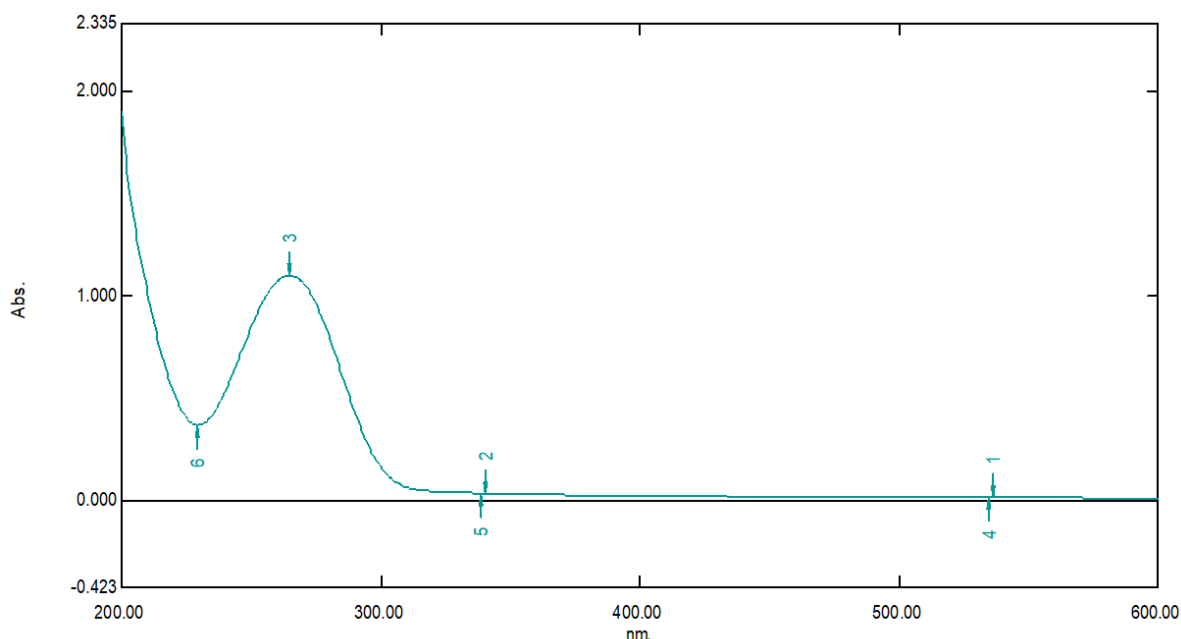
### 3.3 UV-visible analysis

Because it offers crucial details regarding the absorbance, transmittance, and reflectance of polymeric materials, UV-vis spectroscopy is a particularly intriguing approach [13]. PVA is

a crucial polymer because it has excellent optical characteristics including great transparency. Figures 4 and 5 shows the UV absorbance spectra of aqueous PVA at various weight percentage concentrations.



**Figure 4:** UV-vis aqueous PVA 0.05 wt.% solution ( $E_g = 4.71$  eV and  $\lambda = 263$  nm for peak-2).



**Figure 5:** UV-vis aqueous PVA 0.25 wt.% solution ( $E_g = 4.58$  eV) and  $\lambda = 268.5$  nm for peak-3).

PVA's absorbance spectra in water exhibits a distinctive peak at 194 nm that can be attributed to the compound's carbonyl groups C=O and C=C. There have been reports of a reasonably strong band at 270 nm, which is connected to the presence of residual acetate groups in PVA [13-15].

PVA exhibits distinctive absorbance bands associated to high-energy absorption at 263 nm for 0.05 wt%, 267.5 nm for 0.15 wt%, and 265 nm for 0.25 wt% solution. The electrical transitions  $n^*$  (R-band) and  $^*$  (K-band) can be attributed to these bands, respectively. With a slight variation in the position of the bands at various concentrations (wt%) in

the aqueous PVA solutions, all the bands seen in neat PVA. Thus, the UV-vis spectra revealed an absorption that was primarily in the UV region and barely detectable at visible wavelengths. For the peaks of various concentrations in weight percent listed below, energy band gaps are determined for the current inquiry. Sharp peak at wavelength 3 (or 268 nm) and energy band gap calculated as 4.62 eV are discovered for 0.25 weight percent. The peak is located at 2 (263 nm wavelength) and the energy band gap is 4.71 eV for 0.05 weight percent.

Additionally, it has been found that the energy band gap of the polymeric material reduces with an increase in the weight

percentage of the aqueous PVA solution. This leads to the conclusion that as concentration decreases, the energy band gap widens, increasing the dielectric characteristics of PVA.

#### 4. Conclusions

The temperature affects thermoacoustical characteristics, which also depend on the concentration (wt%) of the polymer solution. The presence of weak polymer-solvent interactions is confirmed by the variation in acoustical characteristics (wt%) for various solutions. The spectral analysis clarifies specific chemical interactions that are confirmed by the measurement of thermo-acoustical characteristics. Research on thermoacoustical characteristics suggests that polymer solutions can form single phases and exhibit miscibility. FTIR study indicates that there were no new bonds that were comparable to those in pure PVA were discovered in the aqueous PVA 1(Wt.%) solution. It is because the aqueous PVA solution has poor molecular interaction.

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