



# Viscosity, density, electric conductance and activation parameters of viscous flow for sodium salicylate in aqueous poly (vinyl alcohol) solution

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## ABSTRACT

Viscosity, density and electric conductance of the solution of sodium salicylate + poly (vinyl alcohol) + water at the temperatures 293.15, 298.15, 303.15, 308.15 and 313.15 K are measured experimentally. The partial molar volume, the molar conductance at infinite dilution, and the activation parameters of viscous flow were obtained. An approximately linear relation between the activation free energy of viscous flow and the molar conductance at infinite dilution is observed. Our result shows that, although PVA increases the viscosity of the medium, the diffusivity of ions does not decrease, the solvation volume of salt at infinite dilution changes little. Therefore, the increase in diffusivity is connected with the decrease in the free space of ion moving when PVA is added into water.

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## 1. Introduction

The dissolution and diffusion rate control for hydrophilic drugs is an important task in pharmaceutical technology because it can lead to a better bioavailability. A broad variety of release control methods have been developed. Among them, the method of introducing a component into drug carrier to prolong drug release and decrease drug diffusivity has been widely used [1,2]. The effect of the carrier composition on drug dissolution and diffusion has been our interest in recent years [3–5].

Poly (vinyl alcohol) (PVA) is commonly used as a component to form hydrogels. PVA hydrogels have been used in a number of biomedical applications and drug delivery matrices [6,7]. This is because of their inherent non-toxicity, good biocompatibility, as well as their rubbery and swelling nature in aqueous solutions. Drug diffusion rate in aqueous solution is basically governed by the restrictive effect of the carrier on drug mobility, whether due to a reduction in free volume or an increase in medium viscosity. Systems of this type generally show an inverse relationship between release rate and viscosity, which is predicted by the Stokes–Einstein equation, so that medium viscosity has been used as a routine predictor of a resistance to diffusion [5,8]. The macro-viscosity of aqueous polymer solution is related to drug diffusion coefficients through  $D/D_0 = \eta_0/\eta$ . Where  $\eta$  and  $\eta_0$  are the viscosities of the polymer solution and pure solvent, respectively.  $D$  and  $D_0$  are diffusion coefficients for drug in the presence and absence of polymer, respectively. Drug diffusion is not only related to the properties of the medium, but also related to the

structure and properties of the drug molecule itself, and to the interaction between solute and solvents. From the relation between  $D$  and  $\eta$ , it is difficult to discover the connection between viscosity and the intrinsic properties of the drug itself. Therefore, it is necessary to select some proper parameters to express the relationship between them.

In this paper, viscosity, density and electrical conductance of ternary solution of sodium salicylate + PVA + water were determined experimentally. The molar conductance, partial molar volume and viscous  $B$ -coefficient of sodium salicylate in PVA aqueous solution were evaluated. Using Eyring's transition state treatment, the activation parameters of viscous flow, which reflects the effect of the concentration of PVA on the motion of sodium salicylate in viscous flow, have been obtained. Using these parameters, the effect of PVA on the diffusion of sodium salicylate is discussed.

## 2. Experiments

### 2.1. Materials

Sodium salicylate ( $\text{HO}_2\text{C}_6\text{H}_4\text{COONa}$ ), analytical agent with purity of 99.0%, was dried at 60 °C under vacuum for 24 h before use. PVA is of reagent grade. Its polymerization degree is about 1750. They were purchased from the National Chemical Company of China. Water was obtained by distilling deionized water.

### 2.2. Density and viscosity measurement

PVA aqueous solutions were prepared in dilute concentration region to prevent the formation of hydrogel. Sodium salicylate was

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**Table 1**  
Densities ( $\rho/\text{g}\cdot\text{cm}^{-3}$ ) of sodium salicylate (3)+PVA (2)+water(1) solution

$m_3/\text{mol}\cdot\text{kg}^{-1}$	293.15 K	298.15 K	303.15 K	308.15 K	313.15 K
		$m_2/\text{mmol}\cdot\text{kg}^{-1} = 0$			
0	0.99817	0.99702	0.99561	0.99401	0.99221
0.0031559	0.99838	0.99717	0.99578	0.99420	0.99230
0.0062649	0.99871	0.99752	0.99602	0.99432	0.99257
0.025091	0.99987	0.99874	0.99712	0.99566	0.99372
0.050378	1.00156	1.00032	0.99890	0.99727	0.99543
0.075921	1.00328	1.00204	1.00052	0.99895	0.99727
0.10154	1.00496	1.00364	1.00215	1.00050	0.99852
0.12749	1.00665	1.00538	1.00387	1.00221	1.00024
0.15363	1.00832	1.00704	1.00540	1.00372	1.00175
0.17996	1.01003	1.00871	1.00714	1.00539	1.00346
		$m_2/\text{mmol}\cdot\text{kg}^{-1} = 11.355$			
0	0.99816	0.99696	0.99560	0.99399	0.99225
0.012548	0.99914	0.99794	0.99655	0.99491	0.99311
0.025128	0.99993	0.99874	0.99730	0.99570	0.99390
0.037734	1.00079	0.99957	0.99814	0.99650	0.99467
0.050409	1.00167	1.00046	0.99901	0.99738	0.99553
0.063130	1.00256	1.00129	0.99979	0.99816	0.99638
0.075895	1.00335	1.00212	1.00061	0.99901	0.99714
0.10159	1.00501	1.00376	1.00225	1.00063	0.99871
0.12752	1.00671	1.00544	1.00393	1.00226	1.00039
0.15359	1.00838	1.00704	1.00552	1.00381	1.00201
0.17999	1.01012	1.00873	1.00721	1.00544	1.00349
		$m_2/\text{mmol}\cdot\text{kg}^{-1} = 22.745$			
0	0.99835	0.99717	0.99570	0.99422	0.99241
0.012567	0.99923	0.99802	0.99665	0.99492	0.99324
0.025135	1.00009	0.99885	0.99736	0.99573	0.99402
0.037715	1.00095	0.99974	0.99830	0.99665	0.99481
0.050428	1.00174	1.00052	0.99900	0.99744	0.99570
0.063123	1.00271	1.00143	0.99992	0.99830	0.99646
0.075895	1.00346	1.00217	1.00073	0.99911	0.99729
0.10159	1.00514	1.00387	1.00236	1.00065	0.99888
0.12753	1.00685	1.00550	1.00407	1.00230	1.00046
0.15363	1.00853	1.00716	1.00563	1.00391	1.00206
0.17998	1.01027	1.00890	1.00727	1.00560	1.00368
		$m_2/\text{mmol}\cdot\text{kg}^{-1} = 34.101$			
0	0.99844	0.99726	0.99586	0.99425	0.99249
0.012561	0.99936	0.99817	0.99675	0.99510	0.99328
0.025141	1.00017	0.99905	0.99766	0.99600	0.99419
0.037721	1.00102	0.99975	0.99836	0.99672	0.99491
0.050428	1.00190	1.00064	0.99921	0.99758	0.99573
0.063136	1.00274	1.00146	1.00002	0.99835	0.99652
0.075895	1.00364	1.00239	1.00091	0.99924	0.99737
0.10161	1.00524	1.00398	1.00251	1.00079	0.99895
0.12751	1.00699	1.00571	1.00418	1.00243	1.00050
0.15363	1.00871	1.00739	1.00588	1.00413	1.00221
0.17998	1.01038	1.00901	1.00749	1.00575	1.00382
		$m_2/\text{mmol}\cdot\text{kg}^{-1} = 45.514$			
0	0.99871	0.99748	0.99601	0.99446	0.99265
0.012529	0.99949	0.99823	0.99690	0.99527	0.99346
0.025109	1.00036	0.99918	0.99776	0.99615	0.99433
0.037709	1.00118	0.99994	0.99851	0.99688	0.99500
0.050435	1.00211	1.00085	0.99944	0.99783	0.99595
0.063098	1.00283	1.00159	1.00013	0.99846	0.99663
0.075908	1.00368	1.00242	1.00094	0.99932	0.99745
0.10159	1.00541	1.00408	1.00263	1.00095	0.99905
0.12752	1.00722	1.00585	1.00436	1.00263	1.00074
0.15364	1.00885	1.00747	1.00595	1.00427	1.00233
0.17993	1.01051	1.00920	1.00764	1.00589	1.00391
		$m_2/\text{mmol}\cdot\text{kg}^{-1} = 56.892$			
0	0.99879	0.99762	0.99619	0.99458	0.99284
0.012542	0.99965	0.99845	0.99697	0.99538	0.99357
0.025103	1.00051	0.99923	0.99788	0.99628	0.99441
0.037722	1.00123	1.00004	0.99859	0.99701	0.99515
0.050429	1.00218	1.00097	0.99949	0.99783	0.99600
0.063111	1.00297	1.00174	1.00026	0.99862	0.99675
0.075908	1.00381	1.00261	1.00111	0.99947	0.99761
0.10159	1.00551	1.00425	1.00275	1.00104	0.99917
0.12747	1.00726	1.00590	1.00436	1.00268	1.00076
0.15366	1.00891	1.00757	1.00605	1.00432	1.00237
0.17996	1.01058	1.00926	1.00767	1.00595	1.00403

dissolved in PVA aqueous solution to form ternary solution. Densities were measured with an Anton Paar DMA 602 vibrating tube densimeter with an uncertainty of  $\pm 0.00002 \text{ g}\cdot\text{cm}^{-3}$ . The temperature

of the cell was controlled by circulating water from a water bath with the temperature within  $\pm 0.01 \text{ K}$ . The densimeter was calibrated by using dry air and degassed water as calibration substances. Viscosities were measured by means of a suspended level Ubbelohde viscometer. The efflux time of the fluid was measured with a digital stopwatch to  $\pm 0.01 \text{ s}$ . The viscometer was kept in a water thermostat controlled to  $\pm 0.01 \text{ K}$ . Viscometers were calibrated by water as described in our previous work [5]. Two viscometers were used in the experiments. Detection was performed at least in six replicates for each composition at each temperature. The flow time of water for two viscometers is 124.31 and 130.02 s, respectively. The viscometer constants of equation  $\nu = \eta/\rho = Ct - Kt$  for two viscometers are  $C = 0.007257$  and  $0.006944$ ,  $K = 1.192$  and  $1.341$ , respectively. The estimated relative standard deviation for  $\eta$  was  $\pm 0.1\%$ . Literature data for density and viscosity calibration is from reference [9]. Other experimental details and procedures are the same as those described in reference [5].

### 2.3. Electrical conductivity measurement

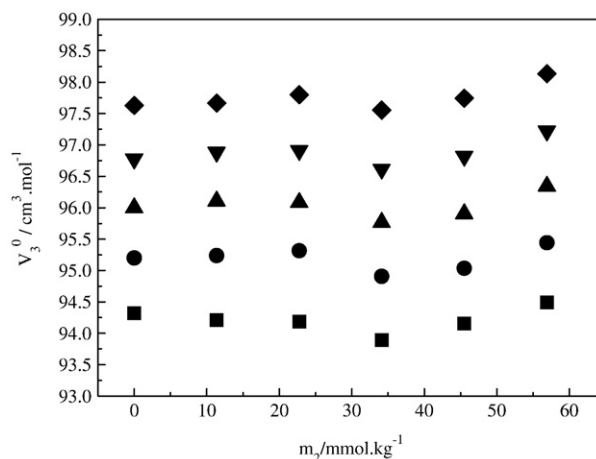
Electrical conductivity was measured using a digital conductivity meter, CON 1500 model, EUTECH Company, USA, with a cell having platinum electrodes. The electrode was calibrated by a standard sample of KCl aqueous solution at different temperatures [10]. The cell constant is  $1.218 \text{ cm}^{-1}$ . According to the instruction of the instrument manual, for electrode with cell constant of  $1.218 \text{ cm}^{-1}$ , the optimal conductivity range is from 10 to  $2000 \mu\text{S}\cdot\text{cm}^{-1}$ .

The conductivity was measured in a glass container, where sample solution was stirred by an electromagnetic stirrer. The temperature of the container was controlled by a water bath with a precision of  $\pm 0.01 \text{ K}$ . The container was sealed with a rubber plug, through which the conductivity cell has been inserted. Conductivity data was recorded at least three times at each experimental condition. The relative error of the measurement was  $< 0.1\%$ .

## 3. Results and discussion

### 3.1. Density and partial molar volume at infinite dilution

Densities of ternary solutions of sodium salicylate (3)+PVA (2)+water (1) are listed in Table 1. In our density experiment, the concentration of solute is less than  $0.2 \text{ mol}\cdot\text{kg}^{-1}$ . That is a dilute concentration range. Generally, for solid dissolves in solvent, the relation between density and the concentration of solute can be expressed in a polynomial expression. However, at the dilute concentration range, it



**Fig. 1.** The effect of the concentration of PVA on  $V_3^0/\text{cm}^3\cdot\text{mol}^{-1}$  at  $T =$ : ■, 293.15 K; ●, 298.15 K; ▲, 303.15 K; ▼, 308.15 K; ◆, 313.15 K.

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