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Anomaly in the Sound Velocity of the Aqueous Solution of Casein

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The concentration dependency of the supersonic velocity in the aqueous solution of casein is measured at 1 mc/sec. And it is found that the velocity suddenly changes near 8~9% concentration. This seems to be due to the hydrogen bond formed in the solution.

§ 1. Introduction

Recently the dynamical properties of synthetic chain molecules have been chiefly studied over the wide frequency region.^{1) 2)} From these results it is expected that the dynamical properties of supersonic region seem to be closely related to the intra and inter molecular interaction of those molecules.

As the chain protein molecules especially have many ions in themselves, the intra and inter molecular interaction is expected to play an important role to their dynamical properties. Therefore the dynamical properties of supersonic region seem to be important to understand the structure of protein molecules.

In this work the sound velocity of the aqueous solution of casein is measured at 1 mc/sec and it is found that the sound velocity suddenly changes to large value near 8~9% weight concentration. Compared with the results of the viscosity measurement, this phenomenon is considered to be due to the fact that the hydrogen bond (for example, $\text{NH} \cdots \text{H}$ -bonding) in casein molecules becomes stronger at this concentration and the network begins to be formed.

§ 2. Experiment

The casein solution was made as following; at first, the casein was swelled in water. After that about 0.5 N NaOH aqueous

solution was added drop by drop into that till the casein was dissolved. In this process the casein was always stirred. The pH of the solution was kept about 6 at the last moment.

The sound velocity was measured by the pulsed method. The schematic diagram of apparatus is shown in Fig. 1. The frequency

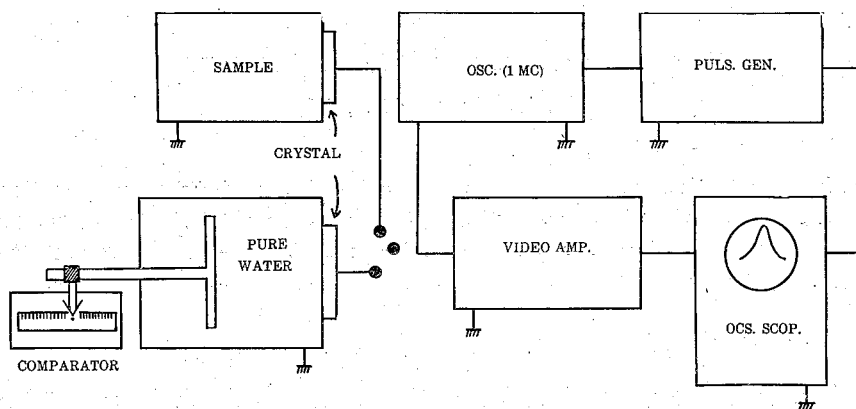


Fig. 1. Schematic diagram of apparatus.

was 1 mc/sec. The quartz crystal was set on the one wall of the vessel. It was used as the sonic source driven by the pulse generator, while it was also used as the receiver which catches the reflected wave from the opposite side of the vessel. The time by which the sound wave required to go and return in the vessel was measured by the oscilloscope. On the other hand, the pure water of which the sound velocity was known was measured by the same method using the vessel with the variable wave path. Compared the position of the signals of both cases on the oscilloscope, the sound velocity of casein solution can be obtained. If the distance between the crystal surface and its opposite wall of the vessel is l , the wave path in the case of pure water is l_w , and the sound velocity in pure water is v_w , the sound velocity in the solution v is shown by the following relation:

$$v = v_w l / l_w$$

§ 3. Result and Discussion

The concentration dependency of the sound velocity at 20°C is

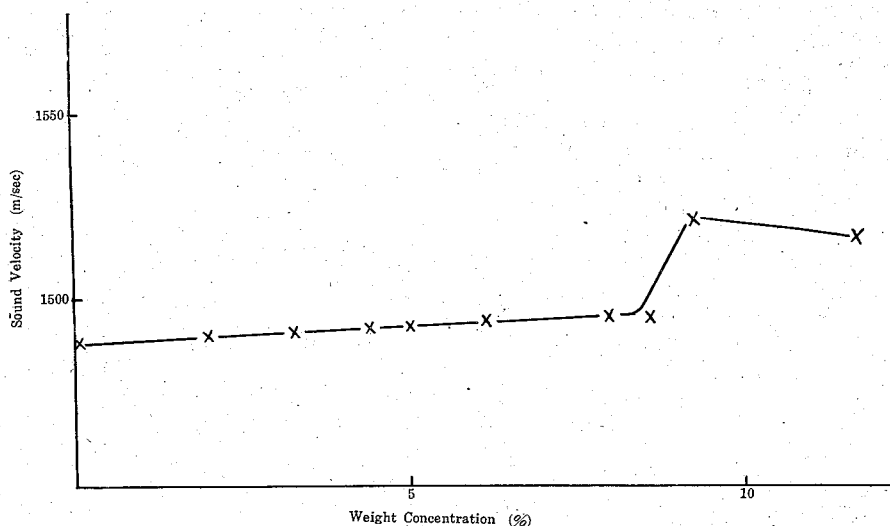


Fig. 2. The concentration dependency of the sound velocity. (pH 6.4, 20°C)

plotted in Fig. 2. It shows that the sound velocity in the casein solution suddenly changes at 8~9% weight concentration. The sound velocity of the solution v is related to the density ρ and the volume elasticity B of the solution by the following equation:

$$v = \sqrt{\frac{B}{\rho}}$$

Accordingly, the anomaly of the velocity of the casein solution is expected to be due to either the density or the volume elasticity. But the abnormal change of the density is not found. Therefore it is reasonable that this anomaly of the velocity is mainly due to the change of the volume elasticity. In other word, it is considered that the cohesion between molecules becomes stronger at 8~9% in the aqueous solution of casein and its volume elasticity increases.

This fact is supported by the viscosity measurement. Fig. 3 shows that the viscosity also becomes abruptly large at 8~9%. Fig. 4 and 5 show the concentration dependency of the activation energy of viscosity. Till about 6%, the activation energy is 3 Kcal/mol, but beyond 8~9%, it increases till 5~6 Kcal/mol. This energy corre-

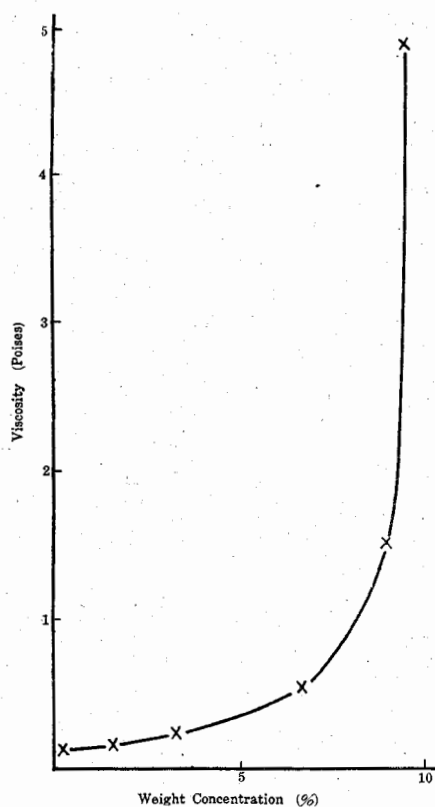


Fig. 3. The concentration dependency of the viscosity. (pH 6.6, 20°C)

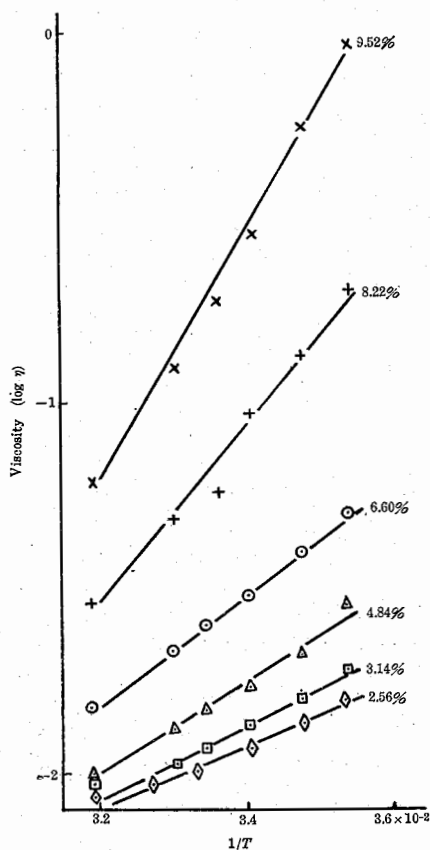


Fig. 4. The temperature dependency of the viscosity. (pH 6.6)

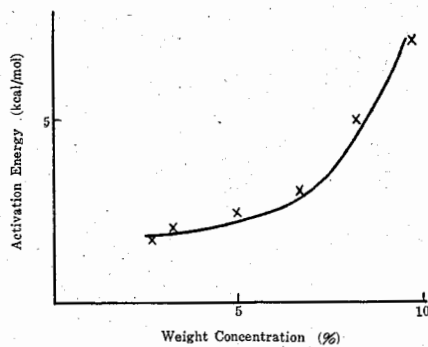


Fig. 5. The concentration dependency of the activation energy. (pH 6.6)

sponds to the binding energy of hydrogen bond. From these results, it is considered that the inter and intra molecular interaction of the hydrogen bond type becomes effective near 9% concentration and the three dimensional network structure is formed in solution.

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