

Ultrasonic Sound Velocity Measurements for Liquid Water-Glycerol Mixtures

K. Nukumizu, Y. Kajihara and M. Inui

Hiroshima University, Higashi-Hiroshima, Hiroshima 739-8521, Japan

kajihara@hiroshima-u.ac.jp

In liquid water-(monohydric)alcohol mixtures, there are known many thermodynamic anomalies¹. The origin of the anomalies is widely believed its special mixing state and the thermodynamic excess functions have been discussed¹. On the other hand, there was also an opinion that the anomalies of the mixture are attributed to the anomalies of water itself². In either case, there is no direct experimental evidence and no conclusion has been reached.

In order to conclude this unsolved problem of water-alcohol mixtures, we study the system by the new experimental concept which we proposed ourselves³. The essence of the method is to deduce the relaxation intensity from the ratio of two sound velocities; one by a low-frequency method like ultrasonic wave and the other by a high-frequency one like inelastic x-ray scattering. In water-ethanol, water-methanol⁴ and water-glycerol⁵ mixtures, the composition dependence of this relaxation intensity at room temperature shows peak at pure water and it gradually decrease with increasing alcohol concentration. Thus we conclude that the origin of thermodynamic anomalies of water-alcohol mixtures are attributed to the large relaxation intensity of water itself, which is universal not only for water-monohydric alcohol mixtures but also for water-trihydric alcohol mixture.

In the present study, we have carried out ultrasonic sound velocity measurements for liquid water-glycerol mixtures in the wide temperature (150 to 320 K) and whole concentration region. The interesting point of the system is that glycerol-rich sample is hardly frozen below the melting point and we can obtain both the low-frequency limit and the high-frequency limit of the sound velocities by one ultrasonic measurement with changing the temperature, which was already reported for pure glycerol⁶. Detail of the results and the universal view for water-alcohol mixtures will be presented.

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