In this thesis, I stated my overall opinion about the hydration behavior of water soluble compounds obtained by experimental results observing the molecular dynamics of both water and solute molecule in a much more broadband frequency range than that covered by the previous studies.

The hydration behavior of substances bearing nitro and cyano groups which are highly polar groups and have low donor number (DN) were discussed based on the combination of the results of near infrared (NIR) spectroscopic technique, dielectric relaxation (DR) spectroscopic technique, and terahertz (TH) spectroscopic technique. A water molecule affected by the presence of nitro and cyano compounds has one non-hydrogen bonded hydroxyl group (called dangling OH) at ca. 7050 cm$^{-1}$ from the results of NIR measurements. The propyl groups in nitro and cyano compounds have one additional dangling OH because the hydrophobic hydration occur the vicinity of hydrophobic propyl group. Moreover, the hydration number of nitro and cyano groups was determined to be zero by using DR measurement. The amount of dangling water of nitro and cyano compounds observed in terahertz region increased with increase in concentration. Consequently, nitro and cyano compounds dissolved into water without any hydrogen bond to water and with only dipolar interactions.
The hydration behavior of substances bearing a tertiary amino group, which has slightly lower dipole moment than other hydrophilic groups but greatly higher DN in dilute aqueous solution, was discussed based on the results of NIR and DR measurements. Since aqueous tertiary amine solutions indicate lower critical solution temperatures (LCSTs) at different temperatures dependent on alkyl chain lengths, the relationship between the hydration number and LCST showing behavior was discussed. Tertiary amines have hydration water possessing the longer hydration lifetime than that observed in other hydrophilic compounds examined before. The temperature dependence of hydration number was determined to be 16 at 10 °C to 8 at 25 °C with increasing temperature and the same results were obtained with different alkyl chains. On the other hand, the number of affected water molecules was determined to be 4 at 25 °C irrespective of alkyl chains. The effect of hydrophobic hydration is negligibly small for tertiary amine compounds because the hydration number and the affected number were determined to be constant values irrespective of alkyl chain species. On the other hand, the molecular dynamics and solution structure of aqueous triethylamine solution in middle to concentrated regime were investigated by means of DR and small angle X-ray scattering (SAXS) measurements. A new relaxation mode was discovered around \( \omega = 10^8 \text{s}^{-1} \), which was not observed in the dielectric relaxation spectra of dilute aqueous solutions of other hydrophilic and hydronutral compounds. The temperature dependence of the relaxation strength of the newly found relaxation mode increases abruptly approaching to LCST contrary to the relaxation times almost constant irrespective temperatures. On the other hand, the formation of cylindrical shape molecular associations is confirmed on the basis of SAXS experiments for 40 wt% aqueous TEA solutions at temperatures close to cloud points, and the associations were elongated with increase in temperature. The mechanism of the newly found relaxation was supposed to be the fluctuation of counterion distribution along the direction perpendicular to the elongated axis of the cylindrical molecular association of triethylamine.

I established new system to measure the viscoelastic behavior of liquids, of which viscoelasticity is too low to be detected by conventional mechanical rheometers, using modified dynamic light scattering (DLS) techniques. Moreover, the viscoelasticity of aqueous solution of chemically modified cellulose ethers determined using new DLS measurement techniques were discussed. The obtained average relaxation time in isolated situation was proportional to molar mass, \( M_w^{-2} \). On the other hand, the reciprocal of steady state compliance corresponding to the average modulus was proportional to the number density of molecules irrespective of the presence of entanglements. These observations strongly suggested that the chemically modified cellulose ethers behave as rigid rod-like particles in aqueous solution possessing hairpin like terminals in both sides.