In the present study, the IR-Soret forced Rayleigh scattering (IR-SFRS) method has been developed to measure the mass diffusion coefficient of methanol aqueous solutions in an electrolyte membrane. This method allows the independent measurement of mutual diffusion coefficient with the partition coefficient, and it also provides a noncontact and high-speed (~100 ms) measurement technique. In applying IR-SFRS to measure the mass diffusion coefficient in electrolyte membrane, it is critical to ensure that the influence of the coherent scattered light both from window materials and membrane. In the case when the diffracted light intensity is not larger enough than that of the scattering light, the heterodyne detection provides accurate measurement. Therefore, we have reconstructed the IR-SFRS apparatus using the heterodyne detection and measured the mass diffusion coefficient of methanol aqueous solutions in electrolyte membrane. The achievements are summarized as follows. (1) IR-SFRS using heterodyne detection was constructed. (2) For evaluation of the new apparatus, the mass diffusion coefficient of toluene \( n \)-hexane solutions at room temperature was preliminarily measured. The deviation of measured value from literature value was ±1% and standard deviation was ±10%. (3) The mass diffusion coefficient of 13 wt% methanol aqueous solutions in Nafion 117 at room temperature was measured. First, we explain that SFRS is possible to measure accurate diffusion coefficient of methanol aqueous solutions in electrolyte membrane compared with other different methods. Second, we discuss the diffusion phenomenon of methanol aqueous solutions in Nafion 117 from measured values.