The reorientation of the tetrahedral complex anion $\text{ZnCl}_4^{2-}$ and the self-diffusion of the cation in $(\text{CH}_3\text{NH}_3)_2\text{ZnCl}_4$ were studied by $^1\text{H}$ NMR spin-lattice relaxation time ($^1\text{H} T_1$) experiments. In the second highest-temperature phase, the temperature dependence of $^1\text{H} T_1$ observed at 8.5 MHz could be explained by a magnetic dipolar-electric quadrupolar cross relaxation between $^1\text{H}$ and chlorine nuclei, and the activation energy of the anion motion was determined to be 105 kJ mol$^{-1}$. In the highest-temperature phase, the activation energy of the self-diffusion of the cation was determined to be 58 kJ mol$^{-1}$ from the temperature and frequency dependence of $^1\text{H} T_1$.

Key words: Nuclear Magnetic Resonance; Molecular Motion; Cross Relaxation; $(\text{CH}_3\text{NH}_3)_2\text{ZnCl}_4$. 