The surface diffusion of nitrogen atoms on amorphous solid water at low temperatures

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Molecular clouds are the birthplace of stars and characterized by low temperature and low density. Despite such an unfavorable environment for chemical evolution, over 150 chemical species have been identified. This chemical diversity cannot be explained by gas-phase reactions alone, and it has been known that surface reactions on amorphous solid water (ASW) existing on interstellar dust grains play important roles. Due to high vacuum, in gas phases, reaction heat cannot be released as thermal energy. In surface reactions, ASW contributes as a heat sink. Furthermore, the surface also enhances the association efficiency of the reactants. In such reactions, radical reactions are particularly important because they proceed even under low-temperature conditions due to barrierless reactions.

The nitrogen (N) is the sixth most abundant element in space, but it is not obvious how N atoms are incorporated into molecules [1]. A large fraction of N atoms is thought to exist as N_2, but it is difficult to detect N_2 by observation. In addition, N_2 is thought to be produced on the surface of icy grains by the recombination reaction of N atoms. Therefore, to understand the formation processes of molecules containing N-atoms, including N_2, it is important to clarify the behavior of N atoms on ASW through quantitative experiments; in this work, we focused on the thermal diffusion of N atoms on ASW.

In the present experiments, to detect trace amounts of N atoms on ASW, we used the combination of photo-stimulated desorption and resonance-enhanced multiphoton ionization techniques: PSD-REMPI method. Using this method, radical on the surface of ASW were photo-desorbed by a weak nano-second laser pulse at 532 nm, they were selectively ionized by the REMPI laser, and the ions were detected by a time-of-flight mass spectrometer. When N atoms are deposited onto the ASW surface and detected using this method, the signal intensity is proportional to the surface number density of N atoms. Therefore, this method enables tracking the time evolution of the surface number density of N atoms.

When N atoms are continuously deposited onto ASW, we found that a steady-state, where the PSD-REMPI intensity became constant, was achieved. Under this condition, the supply rate of N atoms is balanced by the consumption rate, which is predominantly determined by the diffusive recombination reaction ($2N \rightarrow N_2$). The consumption rate increases at higher temperature due to enhanced thermal diffusion, resulting in the lower steady-state intensity. From the temperature dependence of steady-state intensities, we determined the activation energy for surface diffusion to be 930 ± 60 K. This value is significantly different from ~ 300 K predicated by quantum calculations [2] The discrepancy can be explained by considering that the experimental value represents the diffusion from the deepest potential sites, which is the rate-determining step for N-atom recombination, whereas the predicated value is an average from those from various potential sites. In fact, the calculated activation energy from the deepest potential site is approximately 900 K [2].

References

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