

Anion-molecule reactions on methanol droplet and ice surfaces induced by fast ion collisions

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When condensed matter is exposed to radiation, various positive and negative fragment ions can be produced. Since some of these ions are highly reactive, ion-molecule reactions could produce complex molecules with high efficiency. It is possible, for example, that such processes occur on ice surfaces in space. However, information on complex ion-molecule reactions in condensed matter is limited. In particular, studies of negative ions are inadequate, and many of these reactions remain unclear.

In this study, we examined ion-molecule reactions among fragments produced by fast ion irradiation on the surfaces of methanol droplets and ice. Recently, we have developed an experimental system for secondary ion mass spectrometry of positive and negative ions emitted from droplet surfaces induced by MeV-ion irradiation. This enables comprehensive and quantitative analysis of the complex product ions emitted from the surface. Notably, coincidence measurements with transmitted ions allow quantitative comparisons with high precision.¹ The droplet target also has the advantage of always irradiating a fresh surface.

Figure 1 shows a negative ion mass spectrum.² The result is compared with that of the CH₃OD droplet. The peak at $m/z = 59$ did not shift even after deuterium substitution, indicating that this ion does not contain a D atom. As a reaction that does not involve D atoms, we propose an association reaction between CH₃O⁻ and CO, which is also expected to be formed.³ Although still preliminary, measurements of secondary ions from the ice surface are in progress. We have confirmed the emission of similar product ion species. This is because the time scale of secondary ion emission is short, on the order of picoseconds, and thus, their production and emission processes are common regardless of phase. On ice surfaces in space, where methanol and CO are mixed, such anion-molecule reactions can occur not only in fast heavy ion collisions. The present results indicate that anion-molecule reactions can be a potentially important process as one of the pathways for complex molecule formation.

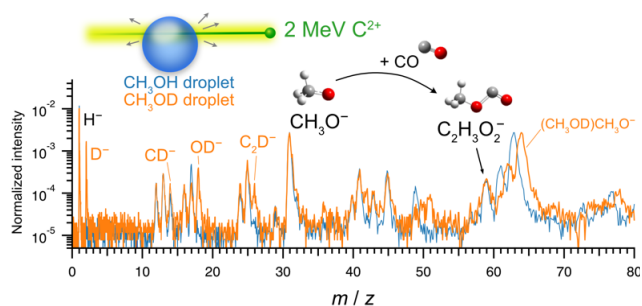


Fig.1 Mass spectra of the negative secondary ions emitted from CH₃OH (blue) and CH₃OD (orange) droplets.

¹ Majima, T. et al. Fast-ion-induced secondary ion emission from submicron droplet surfaces studied using a new coincidence technique with forward-scattered projectiles. *J. Chem. Phys.* **2020**, *153*, 224201.

² Majima, T. et al. Fast heavy-ion-induced anion-molecule reactions on the methanol droplet surface. *J. Phys. Chem. A* **2022**, *126*, 8988.

³ de Barros, A. L. F. et al. Radiolysis of frozen methanol by heavy cosmic ray and energetic solar particle analogues. *Mon. Not. R. Astron. Soc.* **2011**, *418*, 1363.