



Impact chemistry of methanol: Implications for volatile evolution on icy satellites and dwarf planets, and cometary delivery to the Moon



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ABSTRACT

Methanol (CH₃OH) is one of the primordial volatiles contained within icy solids in the outer solar nebula. This paper investigates the impact chemistry of CH₃OH ice through a series of impact experiments. We discuss its fate during the accretion and evolution stages of large icy bodies, and assess the possibility of intact delivery of cometary volatiles to the lunar surface. Our experimental results show that the peak shock pressures for initial and complete dissociation of CH₃OH ice are approximately 9 and 28 GPa, respectively. We also found that CO is more abundant than CH₄ in the gas-phase products of impact-induced CH₃OH dissociation. Our results further show that primordial CH₃OH within icy planetesimals could have survived low-velocity impacts during accretion of icy satellites and dwarf planets. These results suggest that CH₃OH may have been a source of soluble reducing carbon and that it may have acted as antifreeze in liquid interior oceans of large icy bodies. In contrast, CH₃OH acquired by accretion on icy satellites and Ceres would have been dissociated efficiently by subsequent impacts, perhaps during the heavy bombardment period, owing to the expected high impact velocities. For example, if Callisto originally contained CH₃OH, cometary impacts during the late heavy bombardment period would have resulted in the formation of a substantial atmosphere (ca. $\geq 10^{-4}$ bar) composed of CO, H₂, and CH₄. To account for the current CO levels in Titan's atmosphere, the CH₃OH content in its crust may have been much lower than that typical of comets. Our numerical simulations also indicate that intact delivery of cometary CH₃OH to the lunar surface would not have occurred, which suggests that CH₃OH found in a persistently-shadowed lunar region probably formed through low-temperature surface chemistry on regolith.

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1. Introduction

Methanol (CH₃OH) is one of the major primordial volatiles in the outer Solar System and is commonly found in comets and around young stellar objects (Dartois et al., 1999; Ehrenfreund and Charnley, 2000; Irvine et al., 2000; Bockelée-Morvan et al., 2004). Methanol may have been formed in interstellar molecular clouds through surface chemistry on dust grains (e.g., Hiraoka et al., 1994; Watanabe and Kouchi, 2002; Hama and Watanabe, 2013), which in turn would have become the precursors of solids in the solar nebula. Compared with other volatiles, the condensa-

tion temperature of CH₃OH is high under nebula conditions (CO: ca. 30 K; CH₄: ca. 50 K; CO₂: ca. 70 K; NH₃: ca. 80 K; CH₃OH: ca. 100 K: Mousis et al., 2009, 2011; Johnson et al., 2012). This suggests that CH₃OH would have been widely incorporated into icy planetesimals in the solar nebula beyond the H₂O snowline.

The gas-starved model of a (gas giant) circumplanetary subnebula implies that regular icy satellites were formed from planetesimals, which originated from largely unprocessed solar-nebula materials at the location where the gas giant formed (e.g., Canup and Ward, 2006; Alibert and Mousis, 2007). This suggests that primordial CH₃OH would have been contained in icy planetesimals in the circumplanetary subnebula. In fact, CH₃OH has been found in Enceladus' plumes, although its abundance (0.01% relative to H₂O) is significantly less than that typically contained in comets (ca. 1% relative to H₂O: Waite et al., 2009). Given the proposed temperatures in the Ganymede–Callisto formation region in the

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jovian subnebula (ca. 50–100 K; Canup and Ward, 2009), CH₃OH may also have been trapped in the icy satellitesimals in outer regions of the jovian subnebula. In fact, the presence of CO₂ ice in a fresh crater on Callisto suggests that its building materials would have contained primordial CO₂ (Hibbitts et al., 2002), supporting the notion that the nebular temperature in Callisto's formation region would have been low enough to trap CH₃OH in ice.

If primordial CH₃OH in icy planetesimals and satellitesimals survived the impact processes during accretion, it would have played an important part in the geological and chemical evolution of large icy bodies. First, CH₃OH would act as antifreeze, facilitating the existence of liquid water below the freezing point of H₂O (e.g., Shin et al., 2013). In particular, the presence of antifreeze may be important for maintaining interior liquid oceans and cryovolcanism for long periods on icy bodies that are not effectively heated by tidal processes, such as Callisto, Titan, Pluto, and Ceres (e.g., Fortes and Choukroun, 2010; Shin et al., 2013). Second, CH₃OH is an important source of soluble reducing carbon in interior oceans. Through subsequent geological processes, CH₃OH could have been converted into complex organic matter (e.g., Nna-Mvondo et al., 2008; Furukawa et al., 2008; Martins et al., 2013). Methanol would also provide chemical energy to biological processes as a reductant on potentially habitable icy satellites, such as Europa and Enceladus (Hand et al., 2007).

In addition to its importance on icy bodies, the presence of CH₃OH has been suggested as a possible indicator of volatile delivery from the outer to the inner Solar System. The persistently shadowed regions near the lunar poles could possess an abundance of volatile deposits, which have presumably accumulated from cometary and asteroid impacts (e.g., Watson et al., 1961; Heldmann et al., 2012). Remote sensing observations with the Lunar Crater Observation and Sensing Satellite (LCROSS) spacecraft have detected the presence of H₂O vapor, together with gaseous CH₃OH and NH₃, in the ejecta plume formed by an artificial impact onto a persistently shadowed region of the Moon (e.g., Colaprete et al., 2010; Paige et al., 2010). Given the presence of CH₃OH and NH₃ in comets (e.g., Bockelée-Morvan et al., 2004), the LCROSS observations might suggest the intact delivery of volatiles through cometary impacts onto the lunar surface (Colaprete et al., 2010). Alternatively, CH₃OH could also be produced from CO and H through surface chemistry at extremely low temperatures on regolith in a persistently shadowed region of the Moon (Crider and Vondrak, 2002). Based on chemical equilibrium calculations, Berezhnoy et al. (2012) suggest that CH₃OH would be thermochemically unstable in a high-temperature vapor cloud formed by a cometary impact on the Moon. However, the efficiency of impact-induced CH₃OH dissociation would be determined by the kinetics of the reactions, which, thus far, has been poorly investigated based on laboratory experiments.

Despite the importance of CH₃OH to many areas of research in planetary science, a lack of systematic investigations into the impact chemistry of CH₃OH leaves large uncertainties in the quantitative assessment of both its role in the evolution of large icy bodies and the origin of volatiles on the Moon. In the present study, we investigate impact-induced dissociation of CH₃OH through a series of impact experiments based on a CH₃OH–H₂O-ice target and using a chemically clean laser-gun system, which enabled us to investigate the chemical composition of gas species formed by impacts (e.g., Sekine et al., 2011). First, we determine the efficiency of impact-induced CH₃OH dissociation and the chemical composition of the degassing species (Section 3). Next, we discuss the survivability and fate of primordial CH₃OH on large icy bodies (Section 4). The origin of volatiles on the Moon observed by the LCROSS spacecraft is also considered using a combination of experimental results and hydrodynamic simulations of cometary impacts (Section 4). Finally, we present our summary (Section 5).

2. Methods

Fig. 1 shows the configuration of our experimental system, which consisted of a high-energy laser (a Nd:YAG oscillator and glass amplifiers), a stainless-steel vacuum chamber, and a quadrupole mass spectrometer (QMS: BGM-202 QuLee, ULVAC). In Section 2.1, we first explain the acceleration mechanism and our use of the laser gun. The impact conditions achieved in our experiments are also described in Section 2.1. Then, in Section 2.2, we explain the preparation of a CH₃OH–H₂O-ice target and the assumptions adopted to calculate the efficiency of impact-induced CH₃OH dissociation. More detailed descriptions of the laser-gun system and ice-target preparation in our experiments may be found in Ohno et al. (2008), Kawaragi et al. (2009), Fukuzaki et al. (2010) and Sekine et al. (2011).

2.1. Laser gun

Fig. 2 shows a schematic of the acceleration mechanism of our laser gun. We used a gold (Au) metallic foil, with thicknesses of 2.5, 5 and 10 μm (Nilaco Corporation, 99.95% purity), as impactor. The Au foil in the vacuum chamber was irradiated with a pulse laser, and the front surface of the Au foil (ca. 1 μm) was vaporized to form a plasma vapor (Fig. 2). Subsequently, the reaction of the expanding plasma vapor accelerated the other side of the foil (Fig. 2). The impactor was a thin disk with an approximate thickness of 1.5, 4, and 9 μm because the thickness of the original Au foil was reduced by approximately 1 μm by the laser irradiation. The diameter of the impactor was approximately 800 μm, corresponding to the spot diameter of the pulse laser (Fig. 2). Finally, the impactor collided with a CH₃OH–H₂O-ice target, and shock-induced degassing occurred. Using this method, we were able to significantly reduce chemical contamination by gun debris and combustion gases compared with the previous methods that used propellant guns or two-stage light-gas guns (e.g., Lange and Ahrens, 1987; Kato et al., 1995; Burchell et al., 1996; Stewart and Ahrens, 2005). Previous studies used direct laser irradiation to simulate hypervelocity impacts (e.g., Nna-Mvondo et al., 2008). However, the pressure and temperature conditions of vapor clouds formed by direct laser irradiation may be different to those achieved through actual impacts (Kadono et al., 2002). The laser-gun method enabled us to investigate gas species formed by actual hypervelocity impacts.

Table 1 summarizes the experimental conditions pertaining to the laser energy, the original thicknesses of the Au foil, the estimated impact velocities, and the peak shock pressures achieved

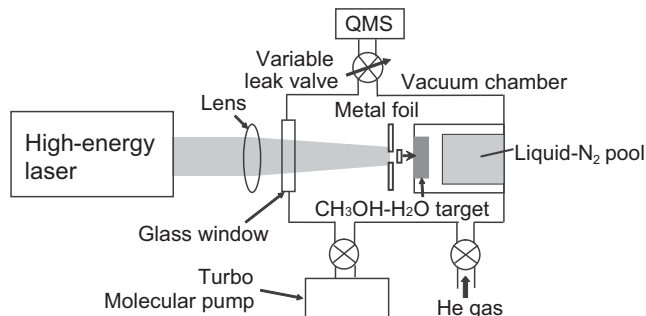


Fig. 1. Schematic diagram of the experimental setup of the laser-gun system. The laser system consists of a high-energy laser (an Nd:YAG oscillator and glass amplifiers), a stainless steel vacuum chamber, a quadrupole mass spectrometer (QMS), and a turbo molecular pump. A stainless-steel target holder containing CH₃OH–H₂O ice was placed in the vacuum chamber. The CH₃OH–H₂O ice was cooled with liquid N₂. Laser irradiation accelerates a gold (Au) metallic foil towards the CH₃OH–H₂O-ice target.

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