



# Experimental investigation of reduced volatile formation by high-temperature interactions among meteorite constituent materials, water, and nitrogen



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## ARTICLE INFO

### Article history:

Received 14 August 2013

Revised 4 November 2013

Accepted 28 November 2013

Available online 7 December 2013

### Keywords:

Astrobiology

Prebiotic chemistry

Impact processes

Atmospheres, chemistry

## ABSTRACT

Late heavy bombardment (LHB) of extraterrestrial objects supplied carbon with metals to the prebiotic Earth. The early oceans were the major target of these impacts, followed by interactions among the atmosphere, oceanic water, and meteorite constituent materials under high-temperature and high-pressure conditions. Post-impact reactions of these hypervelocity impacts have the potential to produce reduced volatiles and organic compounds, including amino acids. Therefore, understanding the reactions in post-impact plumes is of great importance for the investigation of prebiotic organic compounds. The composition of post-impact plumes has been investigated with thermochemical calculations. However, experimental evidence is still needed to understand the reactions in dynamic systems of post-impact plumes. The present study investigates the effects of reaction temperature and availability of water on products from iron, nickel, graphite, nitrogen, and water in a dynamic gas flow system to investigate reactions in post-impact plumes. Results of this study indicate the formation of CO, H<sub>2</sub>, NH<sub>3</sub>, and HCN by hypervelocity oceanic impacts of iron-rich extraterrestrial objects. The formation of methane was limited in the present experiments, suggesting that the quenching rate is an influential factor for methane formation in post-impact plumes. Availability of water vapor in the plume was also an influential factor for the formation of reduced volatiles that controlled the CO formation rate from graphite. These results provide experimental evidence for the formation of reduced volatiles in post-impact plumes, which influenced the formation of prebiotic organic compounds.

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## 1. Introduction

Geological evidence suggests that ancient oceans were formed more than 4.2 billion years ago (Gya) (Wilde et al., 2001; Cavosie et al., 2005). This suggests that the surface of the Earth at that time was cooled and ready for the accumulation of prebiotic organic compounds—potential procurers of life.

Lunar crater records suggest that the Earth experienced late heavy bombardment (LHB) of extraterrestrial objects at around 4.0–3.8 Gya (Culler et al., 2000; Valley et al., 2002). Sedimentary rocks formed at later periods suggest the existence of life (Rosing, 1999; House et al., 2013), implying that the LHB period was the potential time period for the origin of life.

In the Earth's history, a wide size range of extraterrestrial objects has been accreted on the Earth's surface (Neukum et al., 2001). According to estimations by previous researchers, crater-forming bodies were the major mass fluxes during the LHB

compared with meteors and meteorites (Anders, 1989; Chyba and Sagan, 1992).

Osmium isotope and highly siderophile elements in lunar impact melt breccias suggest that extraterrestrial objects similar to iron-bearing chondrites, such as ordinary and enstatite chondrite, were among the major projectiles during the LHB (Puchtel et al., 2008). This argument is supported by a theoretical study indicating that the majority of the LHB projectiles originated from the E-belt, which is composed of asteroids having enstatite–chondrite-like surfaces (Bottke et al., 2012). Other researchers have suggested that extraterrestrial objects similar to iron meteorite were the greatest flux of hypervelocity impacts during the LHB, estimated based on the frequency of fall meteorites and survivability of impactors (Bland and Artemieva, 2006; Pasek and Lauretta, 2008).

Hypervelocity impacts of extraterrestrial objects generate impact heating owing to shock waves and the release of kinetic energy. This heating generates post-impact reactions between reduced extraterrestrial objects and terrestrial objects. The formation of hydrocarbons, hydrogen cyanide, acetaldehyde, and ammonia by these post-impact reactions has been suggested by many previous studies that investigated the impact and post-impact reactions

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of extraterrestrial objects (Mukhin et al., 1989; Gerasimov et al., 2002; Sugita and Schultz, 2003; Nakazawa et al., 2005; Sekine et al., 2006; Kurosawa et al., 2013). Furthermore, Pasek et al. (2013) have shown the flux and existence of reactive phosphorus from meteorites to the oceans of the early Earth. The formation of an amino acid, carboxylic acids, and amines has been demonstrated in shock-recovery experiments that simulated the impacts of extraterrestrial objects containing iron and carbon (Furukawa et al., 2009). Therefore, hypervelocity impacts during the LHB were potentially the largest source process of the compounds that may have contributed to the origin of life (Chyba and Sagan, 1992; Nakazawa, 2006, 2008).

The bulk compositions of these impact-generated gases have been investigated using thermochemical calculations (Hashimoto et al., 2007; Schaefer and Fegley, 2010). Schaefer and Fegley (2010) suggest that the impact of extraterrestrial objects such as H chondrite produces many gases composed of H<sub>2</sub>, CO, CO<sub>2</sub>, CH<sub>4</sub>, H<sub>2</sub>S, SO<sub>2</sub>, and NH<sub>3</sub>. Hashimoto et al. (2007) indicate that even extraterrestrial objects similar to the oxidized meteorite group (CI chondrite) produce many reduced gases.

In hypervelocity oceanic impact processes, projectile and target materials are fragmented/vaporized in dynamical flow in plumes with atmosphere and vaporized water (Pierazzo and Melosh, 1999, 2000; Furukawa et al., 2007). In such dynamical systems, mixing with the terrestrial atmosphere or precipitation would have quickly quenched parts of the plumes. The volatiles produced in these quickly quenched environments are difficult to estimate using only thermochemical calculations, and instead require experimental investigations.

Therefore, in the present study, we conducted heating experiments of carbon, iron, nickel, water, and nitrogen in a dynamic quenching system to investigate the products in post-impact plumes. The effects of reaction temperature and water content of nitrogen on the composition of produced volatiles and solid residues in impact plumes were determined.

## 2. Experimental methods

### 2.1. Materials and procedures of heating experiments

The heating experiments were conducted in a gas-flow system made specifically for the present study (Fig. 1). The gas flow line was composed of a bubbling flask connected to a nitrogen tank, a reaction tube covered by a furnace, and a sample recovery unit. The bubbling flask was filled with ultrapure water (18.2 MΩ; Simple Lab; Millipore). Nitrogen (99.99995 vol%) was introduced into the bubbling flask to be saturated with water vapor at controlled temperatures. Subsequently, nitrogen was introduced to the reaction tube.

A gridded powder composed of 200 mg of iron (99.9 wt%, powder, <45 μm in diameter; Wako Pure Chemical Industries), 20 mg of nickel (99.95 wt%, sponge; Wako Pure Chemical Industries), and 30 mg of graphite (99.7 wt%, powder, ~5 μm in diameter; Kojundo Chemical Laboratory) was enfolded with a piece of silica wool (60 mg). The silica wool was wrapped in a thin gold sheet and inserted into the reaction tube made of silica glass. A gas mixture consisting primarily of nitrogen gas with varying amounts of water vapor was pumped through the reaction tube, reacting with the solid starting material and then transferred into the sample recovery unit.

The sample recovery unit consisted of two parts: a trap flask and a gas sampling bag. The gases introduced into the trap flask were bubbled through 50 ml of sodium hydroxide solution (100 mmol/l; Wako Pure Chemical Industries) or hydrogen chloride solution (10 mmol/l; Wako Pure Chemical Industries) for the

recovery of acidic volatiles or alkaline volatiles, respectively. The outlet gas from the trap flask was then collected in the gas sampling bag for the analyses of low- and nonpolar volatiles.

All experiments were conducted for 20 min. To set the water content of nitrogen (0.04, 0.11, and 0.27 mol<sub>H<sub>2</sub>O</sub>/mol<sub>N<sub>2</sub></sub>), the temperature of the bubbling flask was set at 30, 50, and 70 °C, respectively, with a fixed nitrogen flow rate (0.37 l/min). This flow rate corresponds to a linear velocity of ~5.8 m/min and a heating duration of ~1.6 s.

### 2.2. Analysis of low- and nonpolar volatiles

Low-polar and nonpolar gases (e.g., H<sub>2</sub>, CO, CO<sub>2</sub>, and CH<sub>4</sub>) were analyzed using gas chromatography (6850; Agilent Technologies) with a thermal conductivity detector (TCD). A constant flow of helium at 5 ml/min was used as a carrier gas. A capillary column (HP-PLOT Molesieve, 30 m length, 0.53 mm diameter, and 50 μm film thickness; Agilent Technologies) was used for the separation of these gases at 40 °C. The temperature of the TCD was kept constant at 180 °C.

### 2.3. Analysis of alkaline volatiles

A hydrogen chloride solution in the trap flask for alkaline volatiles was dried up to remove excess hydrochloric acid. The dried residue was dissolved in 5 ml of water. The concentration of alkaline volatiles in this solution was analyzed using ion chromatography (861 Advanced Compact IC; Metrohm) equipped with a cation exchange column (YK-421, 4.6 × 125 mm; Shodex). The temperature of the column was kept constant at 40 °C. Solutions of three different acids were used as eluents for the chromatographic separation: 5 mM of tartaric acid, 1 mM of dipicolinic acid, and 1.5 g/l of boric acid (all from Wako Pure Chemical Industries).

### 2.4. Analysis of hydrogen cyanide

The concentration of HCN collected in the sodium hydrogen solution was analyzed using an ultraviolet–visible (UV–Vis) spectroscopy system (8453; Agilent Technologies) at a wavelength of 620 nm. For the UV–Vis spectroscopy analysis, HCN was subjected to a color reaction based on the pyridine–pyrazolone method. The detailed procedure for the pyridine–pyrazolone method can be found elsewhere (Epstein, 1947).

### 2.5. Analysis of solid residue

The solid residues remaining after the experiments were analyzed by X-ray powder diffraction to identify their mineral composition (XRD; X-Pert Powder; Philips Co.). The XRD system was equipped with a copper target. All diffraction profiles were obtained at a step size of 0.01°, with a divergence slit of 1° and a receiving slit of 0.3 mm. The sample powder of 200 mg was mixed with 20 mg of fluorite powder for use as a reference material. Several solid residues were analyzed with scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM–EDS).

## 3. Results

### 3.1. Components of solid residues and volatile products

The XRD profile of the solid starting material showed 002 reflection of graphite, 110 of iron, 200 of nickel, and 211 of iron. Following the experiments, differences in mineral composition were found between the upstream part and the downstream part of the solid residues, with the upstream part being more oxidized

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