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Thermite reactions with oxides of iron and silicon during combustion of magnesium with lunar and Martian regolith simulants



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ABSTRACT

It has been shown recently that mixtures of JSC-1A lunar regolith simulant with magnesium are combustible. Thermite-type reactions in these mixtures could be used for in situ production of construction materials on the Moon. Because of complex composition of lunar regolith, however, the mechanisms of these reactions are not well understood. Also, for Mars mission applications, it is important to explore the possibility of using Martian regolith in such mixtures. In the present paper, combustion of two Martian regolith simulants (ISC-Mars-1A and Mojave Mars) with magnesium is studied using thermodynamic calculations and combustion experiments. To understand the reaction mechanisms in these mixtures as well as in the mixtures of JSC-1A lunar regolith simulant with magnesium, thermoanalytical experiments are also conducted. It has been shown that the Martian regolith simulants form combustible mixtures with magnesium. The measured combustion temperatures and identified product compositions are in reasonable agreement with thermodynamic predictions. The mixtures of JSC-Mars-1A with magnesium at 20-30 wt% Mg exhibit higher temperatures and burn more vigorously than mixtures based on Mojave Mars, which is explained by the higher content of iron oxide in JSC-Mars-1A. For Mojave Mars, combustion is accompanied by oscillations in the front motion and by the formation of a layered structure of the product. This effect is more significant at lower concentrations of Mg. Thermoanalytical studies have shown that iron oxide plays a dominant role in the combustion of JSC-Mars-1A simulant with magnesium. However, Mg/SiO₂ mixture ignites at a temperature lower by approximately 100 °C than for Mg/Fe₂O₃. For Mojave Mars material and JSC-1A lunar regolith simulant, which include more silica and less iron oxide, silica exhibits a significant effect on the combustion, promoting reactions at lower temperatures. © 2015 The Combustion Institute. Published by Elsevier Inc. All rights reserved.

1. Introduction

Recently, it has been proposed to use combustion of lunar regolith with aluminum [1–3] and magnesium [4–7] for the production of construction materials on the Moon. Thermite-type reactions between regolith and the added metal lead to the formation of products that are sufficiently strong for construction applications. An important advantage of this approach is small energy consumption because the combustion process is self-sustained and only small amount of energy is required for ignition. The required metal could be brought from Earth or obtained *in situ* by recycling the lander parts and by processing regolith [8]. Trade-off studies could be conducted to compare combustion synthesis of construction materials from regolith and transported/recycled/recovered metal, incorporation of regolith into thermoplastics [9–11], and high-temperature sintering of regolith using microwave [12,13] or laser [14] radiation. Such studies, however, should involve a detailed mission analysis, which is beyond the scope of the present work.

Previous research has revealed significant advantages of Mg over Al in combustion with JSC-1A lunar regolith simulant. Combustion with Al requires significant preheating of the mixture [1], adding ilmenite as an oxidizer while keeping the regolith as an inert material [2,3], or adding polytetrafluoroethylene as an additional reactant [15]. In contrast, the mixtures of JSC-1A with Mg ignite with no need for preheating or adding reactants, leading to a steady or spinning propagation of the combustion wave over the pellet at relatively low concentrations of Mg [4–7]. Further, use of SHS compaction (quasi-isostatic pressing immediately after combustion) for JSC-1A/Mg mixtures has enabled fabrication of products with a compressive strength exceeding 10 MPa, i.e., stronger than common bricks (5 MPa) [7].

Because of a complex composition of lunar regolith, however, the combustion mechanisms of its mixtures with metals are not well understood. For example, the mineral composition of JSC-1A

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lunar regolith simulant includes (in order of decreasing mass fraction) anorthite (CaAl₂Si₂O₈), albite (NaAlSi₃O₈), forsterite (Mg₂SiO₄), wollastonite (CaSiO₃), enstatite (MgSiO₃), ferrosilite (FeSiO₃), fayalite (Fe₂SiO₄), and other crystalline and amorphous phases [4]. For simplicity, it is possible, at the cost of a decreased accuracy, to represent the regolith composition as a mixture of simple oxides such as SiO₂, Al₂O₃, Fe₂O₃, CaO, MgO and Na₂O. Among these, SiO₂ and Fe₂O₃ may exhibit thermite reactions with Mg:

$$SiO_2 + 2 Mg \rightarrow Si + 2 MgO, \ \Delta H^{\circ}_{rxn} = -146 \text{ kJ per mol } Mg$$
(1)
$$Fe_2O_3 + 3 Mg \rightarrow 2 Fe + 3 MgO, \ \Delta H^{\circ}_{rxn} = -328 \text{ kJ per mol } Mg$$
(2)

It is unclear, however, which of these two oxides plays a greater role in the combustion process. Silica has a higher concentration in JSC-1A regolith simulant (45.7 wt% vs 12.4 wt% for Fe₂O₃ [16]), but iron oxide thermodynamically exhibits a higher heat release.

Another issue that was not studied previously is the fact that the tested JSC-1A simulant is just one material that is similar chemically to regolith in some specific areas of the Moon surface [16,17]. Lunar dusts in other areas as well as Marian regolith are chemically different from JSC-1A [18,19]. Thus, the hypothesis that Martian regolith forms combustible mixtures with magnesium should be verified experimentally.

One objective of the present research was to investigate combustion of Martian regolith simulants (JSC-Mars-1A and Mojave Mars) with magnesium and compare their combustion characteristics with those of JSC-1A/Mg mixtures. This part of research included thermodynamic calculations and combustion experiments.

As noted above, to fabricate stronger products, SHS compaction could be used [7]. To study combustion behavior and mechanisms, however, experiments in a gas environment are more convenient. The choice of the gas environment is an important issue. As discussed previously [7], combustion of regolith mixed with Mg or Al is hardly possible in ultrahigh vacuum (pressure less than 10^{-7} Pa) of the Moon because of vaporization of the metals during ignition so that a pressurized reactor is required. On Mars, mixtures of regolith with Mg could be burned in CO₂, which constitutes 95% of the Martian atmosphere, but the low atmospheric pressure (0.6–1 kPa) would remain a problem. It has been reported that at pressure less than 25 Torr (3.3 kPa), ignition of JSC-1A mixed with FeTiO₃ and Mg led to the pellet disintegration [3]. On the other hand, no differences in the combustion behavior and front propagation velocity were observed for JSC-1A/Mg mixtures at pressures 10 kPa and 90 kPa [4]. Since the goal of the present work was to compare the Martian and lunar regolith simulants, all combustion experiments were conducted under identical conditions, i.e., in an argon environment at 1 atm. This was also convenient for comparison with thermoanalytical experiments, conducted at atmospheric pressure.

It should be noted that on Mars it could be advantageous to use compressed Martian "air" (which is mainly CO_2) as an environment in the reactor. From this point of view, it would be important to conduct experiments on combustion of Martian regolith simulants with Mg in a CO_2 environment. The use of CO_2 instead of argon may influence the combustion process because Mg particles burn with CO_2 [20–24]. In the present research, however, the goal was to compare the Martian regolith simulants with the lunar one so that only argon was used as the environment.

The second objective of the research was to clarify the mechanisms of reactions that occur during combustion of the regolith simulants with magnesium. This was achieved using thermoanalytical experiments and X-ray diffraction analysis of quenched samples.

2. Experimental

ISC-Mars-1A and ISC-1A regolith simulants were obtained from ORBITEC, while Mojave Mars simulant was provided by NASA. In contrast with the actual lunar dust, JSC-1A is a quite coarse powder, which needs to be ground to the particle size of less than $100 \,\mu\text{m}$ for combustion with magnesium powder [4]. The two Martian simulants are also coarse [18,19]. To compare the combustion characteristics of the three simulants, each of them was milled to a powder with a median diameter of $2-3 \mu m$. This was achieved with a planetary ball mill (Fritsch Pulverisette 7 Premium Line) at a rotation speed of 1100 rpm using zirconia-coated bowls and zirconia grinding balls (diameter: 1.5 mm; the balls-mixture ratio: 4:1). Each of the four cycles included a 10-min milling period and a 60-min pause for cooling. The particle size distributions of the milled powders were measured using a laser diffraction particle size analyzer (Microtrac Bluewave), with isopropyl alcohol being used as a carrier fluid. For example, Fig. 1 shows the particle size distribution for the milled JSC-Mars-1A simulant. The particle size distributions of two other milled powders are similar.

The milled regolith simulants were mixed with magnesium powder (-325 mesh, i.e., less than 44 µm, 99.8% pure, Sigma-Aldrich) in a three-dimensional inversion kinematics tumbler mixer (Bioengineering Inversina 2L) for one hour. The mixtures were then compacted into cylindrical pellets (diameter: 13 mm, height: 21–24 mm) using a uniaxial hydraulic press (Carver).

To study the combustion process, the pellet was placed inside a windowed steel chamber (diameter 30 cm, height 40 cm), connected to a compressed argon cylinder and a vacuum pump. All experiments were conducted in an argon environment at 1 atm. The pellet was installed on a brass pedestal inside the chamber and ignited at the top by a tungsten wire heated with a DC power supply. The temperature in the middle of the pellet during combustion was measured by a thermocouple (K-type, 254-µm diameter wire, Omega Engineering). The thermocouple, located in a two-channel ceramic insulator (outer diameter: 1.6 mm, Omegatite 200, Omega Engineering) was inserted into the pellet through a channel drilled perpendicularly to the pellet axis. Digital video recording (Sony XCD-SX90CR) was used for observation and for determination of the combustion front velocity. The combustion product compositions were characterized using X-ray diffraction analysis (Bruker D8 Discover XRD).

Thermal analysis of mixtures was conducted using a differential scanning calorimeter (Netzsch DSC 404 F1 Pegasus). The DSC was calibrated using standard materials (In, Sn, Bi, Zn, Al, and Au) obtained from the manufacturer. The temperature program was carried out for all these materials with three heating and two cooling segments so that the melting peak of each material was measured three times. All calibration runs were performed using alumina crucibles at a heating rate of 20 K/min and argon flow rate



Fig. 1. Particle size distribution for the milled JSC-Mars-1A simulant.

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