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Abstract

The study of aluminum combustion in pure oxigen and mixture (N_2-H_2-Cl_2-O_2) at high temperatures and pressures provides a simulation of combustion mechanism study of aluminum in solid rocket propellant. Computations were performed for predicting possible intermediate species and products. Emission spectra of important intermediate species were identified by using a spectrometer. Time histories of the AlO emission band show the continuum radiation of Al_2O_3 which indicates that AlO is a precursor to Al_2O_3 in the reaction scheme. The solid and liquid products were observed and analyzed by electron diffraction, X ray diffraction, transmission and scanning electron microscopes. The gaseous products were detected by using infrared spectrometry. These measurements are in good agreement with the computations and provide useful chemical kinetic data.

A SIMULATION STUDY OF THE COMBUSTION MECHANISM OF ALUMINUM IN SOLID ROCKET PROPELLANT AT HIGH TEMPERATURES AND PRESSURES IN A SHOCK TUBE

Liu Zichao, Beijing Institute of Aeronautics and Astronautics

I. Preface

The study of aluminum combustion in pure oxygen and a N_2-H_2-Cl_2-O_2 mixture using a high temperature and pressure environment (can reach 5000K and 40 atmospheric pressures) behind the reflective shock wave in a shock tube can provide a simulation study of the combustion mechanism of aluminum powder added in solid rocket propellant. Equilibrium components were computed and possible intermediate species and final products were identified for the combustion reaction under different conditions. Some primary intermediate species of aluminum combustion were identified by using a spectrometer and time histories of AlO were recorded. The discovery of continuum radiation of Al_2O indicates that AlO is a precursor to Al_2O_2. Solid and liquid products as well as their particulate conglomerates were observed and analyzed by electron and x-ray diffraction, transmission and scanning electron microscopes. The gaseous products were analyzed by using infrared spectrometry. All these measured results were in good agreement with the calculated results. This provided useful chemical kinetic data.

This paper was presented in the Conference of Combustion of the Association of Chinese Engineering Thermophysics which was held in Tianjing in October, 1984.

The performance of solid rocket propellant can be markedly improved by adding aluminum. For years, there have been many studies on this, but the question of chemical kinetics is still not very clear. Most of the studies on aluminum-oxygen reaction are conducted under 10 atmospheric pressures and there are basically no data on conditions above 10 atmospheric pressures. The data on reaction of aluminum in ammonium chlorite are even more rare. This has caused difficulties in estimating the performance of rocket motors. For instance, the conglomeration mechanism of Al_2O_3 particulates as a combustion product is not well understood, and this makes it difficult to estimate the effects of interflow caused by such mechanism.

The high temperature and pressure zone behind the reflective shock wave in a chemical shock tube can simulate the operation environment of a rocket motor. Figure 1 shows a schematic diagram of the aluminum reaction zone behind the reflective shock wave in the shock tube. The combustion process and emission spectra of intermediate species were recorded by using a high-speed camera and spectrometry. The experiments have shown that aluminum ignition starts at T_5, is between 2000-2400K and a blue-green light of AlO is emitted[1]. This is related to the 2300K melting point of the oxidation layer. Computations were performed for the chemical equilibrium components to predict the intermediate species and final products of $Al-O_2$ and $Al-N_2-H_2-Cl_2-O_2$ reactions, and they were used as guides for the experiments. Useful chemical kinetic data were obtained through the analysis of intermediate species and final products using spectra, electron and x-ray diffraction, electron microscope and infrared spectrometry.

II. Computation results of intermediate species and products for aluminum combustion

The conditions of working gases (pure oxygen or 0.1 N_2 + + 0.4 H_2 + 0.1 Cl_2 + 0.4 O_2 mixture) before aluminum combustion were computed first. Then the intermediate species and final products of aluminum participating in combustion under high temperature and pressure were computed. The minimized Gibbs free energy method under isobaric and isothermal conditions was applied in the computations to take true gaseous effects into consideration.

For the $Al-O_2$ reaction, when the incident Mach number Ms=9.1 and at P_5=28.9 atmospheric pressures and T_5=4418K, the composition of oxygen before the ignition of aluminum was computed to be 43% molar fraction of oxygen atoms and 56% molar fraction of oxygen molecules. The mass ratio of oxygen and aluminum was 1.32 and the aluminum was preheated to 900k. Twenty-three possible components appearing during the combustion were considered in the computations. The temperature after aluminum combustion was 4930K. The majority of the intermediate species were O (42%) and AlO (19.7%), and the remainder was Al, Al^+, AlO^+, AlO^-, Al_2O, AlO_2, Al_2O_2, Al_2O_3, O^-, etc.

For the $Al-(0.1N_2+0.4H_2+0.1Cl_2+0.4O_2)$ reaction, the mass ratio of mixture and aluminum was 2.6, and the components, specific heat and molecular weight of the mixture under various Ms, T_5, P_5 before the aluminum combustion were computed. The maximum conditions of the experiment were Ms=10.5, T_5=5100K and P_5=40 atmospheric pressures. For a typical condition of Ms=7.8, T_5=4149K, P_5=14.3 atmospheric pressures, the primary components before the ignition of aluminum are: O (17%), OH (14.3%), H (13.4%), O_2 (11.4%), H_2O (10.4%), Cl (9.8%), HCl (7%) and N_2, H_2, NO, etc. The appearance of 80 components was considered when aluminum participated in combustion. The computed results of 23 components are presented in Figure 2. The majority was Al, AlO, AlCl, HCl, H, O, OH, Cl, etc. The molecular weight of the combustion gas is 25.67g/mole and its specific heat is 1.118. Figure 3 shows the changes of 18 intermediate species during the cooling process. The final products were mainly water, water vapor, HCl, ammonium, N_2, H_2 and Al_2O_3 solid particles with water, HCl, Al_2O_3 as the majority. This is consistent with the results of rocket experiment measurement.[2]

III. Measured results of intermediate species and final products for aluminum combustion

The spectrometer recorded the $Al-O_2$ reaction spectra of Al(3944,3961.5Å), Al^+(5861A) and six bands of AlO(4373.7, 4470.5, 4648.2, 4842.3, 5059.3 and 5226.9Å). No bands of AlO_2, Al_2O and Al_2O_2 were found in the visible light range. The AlO band was very strong. The $Al-N_2-H_2-Cl_2-O_2$ reaction emitted a large amount of spectra and bands with some overlapping, but a spectra of Al, Al^+, AlO, AlH, H, O, etc. could still be identified. Some weak bands might have belonged to AlCl, $AlCl_2$ and AlOH. The time histories of AlO (Fig.4) shows that the maximum value of strength occurred 1.4 milliseconds after the ignition, then declined after 2 milliseconds. A strong continuum radiation appeared, subsequently lasting 14 milliseconds. Analyses show that this was emitted by Al_2O_2. It is very possible that AlO is a precursor to Al_2O_2.

X-ray diffraction identified the white powder produced from the $Al-O_2$ reaction to be Al_2O_3. These were spherical particulates with sizes ranging from 0.1 to 0.6 micron measured by an electron microscope, and they are in good agreement with the measurements of Al_2O_3 particulates exhausted from the rocket.[2] The solid products from the reaction of Al and the mixture were identified to be the Al_2O_3 particulate conglomerates suspended in the solution of HCl + H_2O. They were no longer spherical which indicates that the congomeration of particulates was due to the effects of HCl and H_2O.

The gaseous products from the reaction of aluminum and the mixture were analyzed and found to be water vapor and HCl vapor by using infrared spectrometry, and are consistent with the computation results. There were far more NO and NO_2 measured that those computed. This was caused by the nonequilibrium process during cooling.

IV. Conclusions

1. Region behind the reflective shock wave in a chemical reaction shock tube can provide a simulation study of combustion mechanism of aluminum in solid rocket propellant.

2. The computation results of intermediate and final products after combustion and the measured results using various methods are basically in good agreement. This provided some basic data for the study of aluminum combustion mechanism under high temperature and pressure.

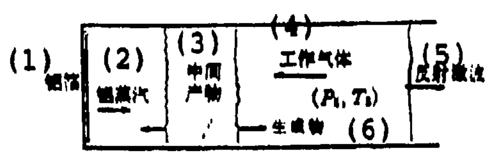

Fig.1 Schematic diagram of the reaction zone near aluminum surface behind the reflective shock wave
Key: (1) Aluminum foil; (2) Aluminum vapor; (3) Intermediate species; (4) Working gases; (5) Reflective shock wave; (6) products.

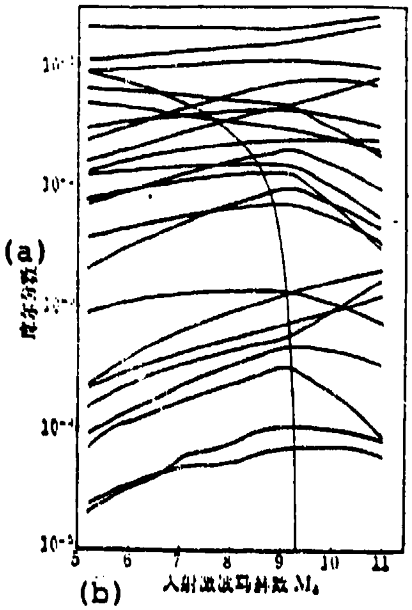

Fig. 2 Computation results of components of reaction of Al and mixture, from upper left-hand corner downward, respectively,
H、O、OH、Al_2O_3、H_2O、HCl、AlCl、AlO、Al、NO、AlOCl、AlO_2H、AlO_2、AlOH、Al_2O、$AlCl_2$、AlH、Al^+、N、AlO_2^-、Al_2O_2、HAlO、$AlOH^+$.

Key: (a) Molar fraction; (b) Incident shock wave Mach No. M_s.

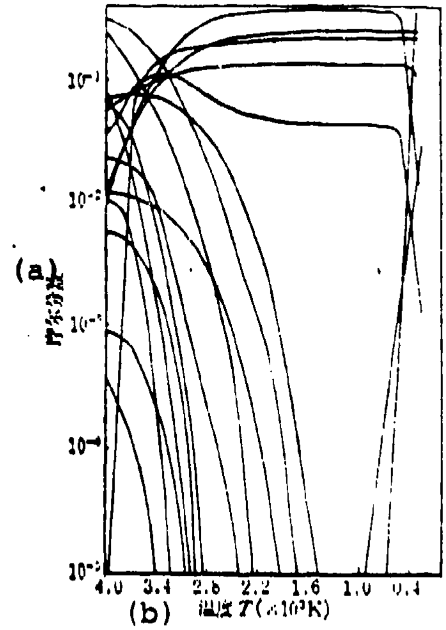

Fig. 3 Component change during cooling after the reaction of Al and mixture, from left dwonward, respectively, H, O, Al, Cl, AlOH, N_2, H_2, $AlCl_2$, HCl, NO, H_2O (vapor), Al_2O, AlO_2, Al_2O_2, AlH, Al_2O_3 (liquid, solid), H_2O (liquid), NH_3.
Key: (a) Molar fraction; (b) Temperature T(X10^3K).

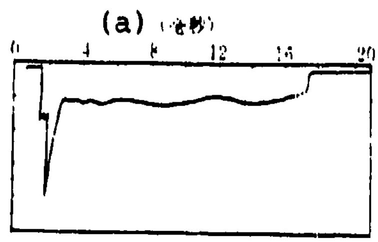

Fig.4 History of band strength of AlO at 4846.2Å.
Key: (a) millisecond.

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