STUDY OF GAS PHASE IGNITION MECHANISM WITH SKIMMING TRANSIENT MASS SPECTROSCOPY

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FINAL REPORT

BY

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13. ABSTRACT

A series of pyrolysis experiments have been performed on Ammonium Perchlorate (AP) ranging from low temperature solid phase decomposition to high heating rate (10 C/sec) flash evaporation. The aim of these experiments was to study the effects of gas phase reactions and to determine the nature of AP pyrolysis at heating rates that are representative of actual propellant systems. All experiments were performed with the use of transient mass spectroscopy. The results may be summarized in three categories

1. High temperature flash oven pyrolysis results showed product gas spectra similar in nature to the solid phase decomposition spectra but did not show the existence of any Cl_2.

2. The flash evaporation experiments resulted in product gas spectra with a pronounced sublimation like character even at flash temperatures as high as 600°C. This suggests that AP at high heating rates essentially sublimes into the gas phase and that the solid phase reactions are too slow to take part in the pyrolysis process.

3. Some preliminary experiments were also performed with doped AP, but conclusive data was not obtained.

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Problem Statement.

As part of the Army's broad attack on the fundamental questions relating to propellant ignition, the present research was initiated in order to study the gas phase ignition mechanism of Ammonium Perchlorate (AP). The primary goal of the work was to study AP decomposition chemistry under conditions that are characteristic of actual propellant ignition and combustion. Namely, at surface temperatures in the range of 400 - 600°C with heating rates in the range of 10^5 - 10^7°C/sec. The technique used for the study was the rapid vaporization of a small sample of AP in order to form an unsteadily expanding and simultaneously reacting gas cloud in vacuum with initial pressure and temperature corresponding to that likely to be encountered in actual propellant combustion. This expanding cloud was sampled or "skimmed" for mass spectroscopic analysis. Several methods were used to accomplish vaporization. AP was flashed in a hot oven in front of the time-of-flight mass spectroscope; AP was sublimed onto a thin ribbon from there it was flashed into the mass spectroscope. Lastly, AP powder was physically placed on the thin ribbon from where it was flashed as before.

Results.

The research program was exploratory in nature, and it was aimed at answering such questions as:

Is transient mass spectroscopy a meaningful technique to use for the study of ignition mechanisms of solid propellant systems?

What is the effect of a relatively high pressure product gas cloud over the decomposing AP surface?

What is the nature of AP decomposition under realistic heating rates in the range of 10^5 - 10^7° C/sec?

What is the effect of controlled impurities on the decomposition of AP?

Some of these questions were answered during the course of the research program, but some of them were examined only in part. The following is a brief exposition of the results that were obtained and described in the publications listed in this report.

It was found that transient mass spectroscopy is one of the techniques that can be used to make a high pressure reaction compatible with the vacuum requirements of mass spectroscopes. Perhaps it may be considered as a parallel technique with high pressure sampling each having its own advantages and disadvantages. The results of the present work show that transient mass spectroscopy can be used to study propellant ignition and deflagration chemistry. Furthermore, with the flash evaporation technique, realistic propellant heating rates can be achieved. The reaction "freezing" feature of the gas-dynamic expansion of the product gas cloud was clearly demonstrated by the fact that sublimination products were preserved throughout the expansion, even at flash temperatures as high as 650°C.

High temperature flash oven pyrolysis results showed product gas spectra similar in nature to the solid phase decomposition spectra, but did not show the existence of any Cl_2.

Flash evaporation results indicated that at high heating rates AP predominantly sublimes into Ammonia and Perchloric acid and that the final decomposition takes place in the gas phase. Increasing the gas phase residence times or the quasi-steady product gas pressure in the high temperature oven resulted in an increase in the O_2 production. Nevertheless, the spectra still remained at high heating rates essentially sublimation like in character.

Doped AP samples were also flashed into the time-of-flight mass spectroscope. The technique of supporting the samples on the thin flash ribbon was developed relatively late in the program. Hence, the study of doped AP remains incomplete. Nevertheless, it was found that doped AP powder can be pyrolysed at extremely high heating rates and their decomposition chemistry can be compared to that of pure AP. It is hoped that future experimental work employing transient mass spectroscopy at high heating rates will explore the important questions relating to doped AP.

Scientific Personnel Supported

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* Year of graduation

Graduate Degrees Awarded.

Chul Cho, Master of Science, Mechanical and Aerospace Engineering, July, 1974

Reports and Presentations.

"Mass Spectroscopic Study of Flash-Evaporated Ammonium Perchlorate," Second Review of the Army's Research on Ignition Phenomenon Program, IBL, Aberdeen, Maryland, Novemoer, 1972

"Flash Pyrolysis of Ammonium Perchlorate," Paper No. E-5 in USA Ballistics Research Laboratories Report No. 1707, April, 1974

"Study of Gas Phase Ignition Mechanisms with Skimming Transient Mass Spectrometry," Chief Investigators Conference and Review of the Military Theme "Physics and Chemistry of High Energy Materials," Picatinny Arsenal, April, 1973

"Unsteady Expansion of a Gas From a Vaporizing Source Surrounded by Vacuum," by Chul Cho, Master's Thesis, University of Massachusetts, Amherst, July, 1974