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論文の要旨
Summary (Abstract) of doctoral thesis contents

High performance and low-toxic monopropellant is required as an alternative to hydrazine for overall cost reduction to the construction and operation of the propulsion system. Ammonium Di-Nitramide-based Ionic Liquid (ADN-based IL) is one of the potential alternatives in this regard. It can be produced by mixing only three solid powders: ADN, monomethyl amine nitrate (MMAN), and urea. The liquefaction of ADN raises its density to around 1.5 times as high as that of hydrazine. The range of theoretical specific impulse is higher than that of hydrazine in almost every composition selected by the author. However, the IL has extremely low-volatility and considerably high viscosity when compared to conventional monopropellants. In addition, combustion mechanisms of ILs have not been clarified yet. Therefore, the technical feasibility of the thrusters with ADN-based IL is not clear.

The objective of this study is to clarify the combustion process of the ADN-based IL and to evaluate its application to a thruster. In this evaluation, the author focused on the processes of ignition and the flame holding, the stay time in combustion chamber, and the technical feasibility of propellant feed system.

ADN/MMAN/urea = 30/50/20, 40/40/20 wt.% are selected as candidate compositions among compositions at intervals of 10 wt.% of each substance, with consideration not only for providing higher specific impulse than that of hydrazine, but also for compatibility with a combustion chamber made of a SiC/SiC composite material in terms of thermal and chemical resistance. The respective density-specific impulse related to these compositions are 1.49 and 1.62 times as that of hydrazine, and both of them melt at around -30 degrees C.

The strand burning tests with samples of end-burning cylindrical liquid columns poured into a fused silica tube, were conducted under constant ambient-pressure conditions ranging from 0.15 MPa to 2 MPa, in order to investigate the combustion process of the ADN-based IL. The five compositions ADN/MMAN/urea=30/50/20, 35/45/20, 40/40/20, 37.5/37.5/25, 35/35/30 wt.% were tested as samples for investigation on the effects on combustion of mass fraction of urea in the IL and mass ratio of ADN to MMAN. At first, the combustion characteristics were obtained in the tests. The self-sustainable combustion (SSC) was confirmed at higher pressure range for all the compositions. It is confirmed that the linear burning rate tends to increase with increase in mass ratio of ADN to MMAN and with decrease in mass fraction of urea in the IL.

The combustion process was investigated on the basis of the temperature data and the video images obtained in the burning tests. The combustion wave structure was identified by both direct visualization of the combustion wave using a high-speed video camera and intrusive temperature measurement in the flame with a thermocouple. According to the behavior of bubbles on the surface of the IL in the strand burning tests, there are three

distinct phases in the combustion wave structure: liquid phase, gas-liquid phase, and gas phase. From the temperature distribution (in the direction from the liquid surface to the combustion gas in the flame), it is confirmed that there is a region of constant temperature preceding a region of rapid temperature rise. Then, the temperature reaches a level near the theoretical adiabatic flame temperature. The rapid rise in temperature shows the ignition in the gas phase. The location of ignition is assumed to be the burning surface. According to the comparison between the observed still image of combustion wave and the temperature distribution, there are two constant temperature regions in the gas-liquid phase and two step combustions occur in gas phase. The author, thus, discussed reactions in the gas-liquid phase and in the gas-phase separately with the aid of experimental demonstrations and numerical simulations with CHEMKIN-PRO for the strand burning.

It is inferred that the combustion process of the ADN-based IL is assumed as follows. The temperature of the IL adjacent to the burning surface increases due to heat conduction. Then, the ADN decomposes and the urea evaporates at around 408K before the MMAN dissociates into $CH_3NH_2$ and $HNO_3$ at around 523K. The mixed gas of the processes and the liquid ammonium nitrate (AN) are produced on the burning surface. As a result of the dissociation of liquid AN, the pre-mixed gas is produced and then it burns in two steps because, the reductions of NOx and nitric acid with nitrogen compounds ($CH_3NH_2$ and $NH_3$ and urea) occur in advance of the complete burning reaction.

The completeness of combustion was evaluated from a viewpoint of the length from the burning surface to the second flame in the strand burning. The ADN-based IL showed higher completeness than simple ADN because the length of the IL flame is much shorter than that of the ADN flame under the same ambient pressure conditions. This can be attributed to the reductions of $NO_X$ with relatively large amount of nitrogen compounds ($CH_3NH_2$ and $NH_3$ and urea).

For the evaluation of application to a thruster, the author focused on the processes of ignition and flame holding and the residence time in the combustion chamber. The technical feasibility of the propellant feed system was also analyzed.

In order to realize ignition in short time, atomization should be conducted. According to the estimation of heating time based on the heat diffusivity, atomization is required for restriction of ignition time to tens of milliseconds. In order to have a cascade of successful ignition from some heated droplets to all the droplets, it is required that the spray is condensed and adequate number of drops be heated due to the extremely low-volatility of IL. This can be achieved for example, through a heater made of porous ceramic with large area for heating all drops.

For proper flame holding in the combustion chamber, conventional flame holding of premixed gas is required. In addition, it is necessary to form the recirculation flow of the premixed gas after the flame holder which sets in the combustion chamber.

In the evaluation of the stay time in combustion chamber, thruster combustion model was constructed and the stay time in terms of atomization, heating and gas-phase combustion was evaluated separately. The stay times for thrusts of 1, 3, 10 N were calculated by summing the three partial stay times. As a result, the partial stay time for gas-phase combustion is 0.06-1.17ms in the promising compositions of the IL selected above. Thus, it is small enough comparing to the other partial stay times. On the other hands, the partial stay time for heating is dominant in the stay time. In the case of thrust of 1 N, the stay time is within conventional stay times of liquid propulsion systems, 2-40 ms, in the condition that the discharge coefficient of the injector is more than 0.81. However, in cases of 3 N and 10 N, the stay times are over 40 ms. This results in the characteristic length of the thruster being considered longer than conventional ones. Therefore, it is required to design the thruster system to reduce the length required to heat the IL in combustion chamber. One of the ways this is possible is by disturbing the flow of spray by heater with porous construction.

Next, feasibility of the propellant feed system with the IL is discussed in the composition of ADN/MMAN/urea=40/40/20 wt.%. The pressure and the weight of the propellant tank might increase because the IL has much higher viscosity (332mPas at 0 degree C) than that of hydrazine. Therefore, the author modeled the simple propellant feed system and evaluated the feed pressure and the pressure loss of the products in the feed system. According to the experimental results of condition of the flame propagation, quenching diameter was estimated and it was found that the inner diameter of capillary tube should be less than 3 mm in combustion pressure of 1 MPa. The diameter is further estimated to be around 1–2 mm because the pressure loss of summation of capillary tube and injector should be 10-30% of combustion chamber pressure. As a result, a fine capillary tube can be designed to have low pressure loss. Meanwhile, pressure loss of filter is the most dominant in the total pressure loss in the feed system in the present study. With the selected nominal-2-pore-size filter and a fine capillary tube, the feed pressure is less than 2MPa for the case of thrust less than 3N. To decrease the pressure loss of filter, viscous resistance coefficient of filter should be decreased by selecting a filter with a large cross-sectional area. Therefore, the design and construction of the propellant feed system is feasible if the capillary tube and the filter are designed properly.

From the above results, the evaluation of the application to the thruster is as follows. Spray ignition and a structure to hold the flame in combustion chamber are required to apply the IL to the thruster. In addition, because the time for heating the drops is the most dominant in the evaluation of the stay time, thruster system should be constructed to reduce the heating length of the drops. Meanwhile, there is the feasibility for the design and construction of the propellant feed system if capillary tube and filter are designed properly. Nevertheless, the IL can still be applied to the thruster of at least 1N.

In conclusion, the combustion process of the ADN-based IL was clarified with the aid of experimental demonstrations and numerical simulations for the strand burning. The application of the ADN-based IL to the thruster was evaluated from four viewpoints. It is found that the ADN-based IL is a promising monopropellant with high performance and combustion completeness. It can be applied to the thruster at least in the condition of low thrust level.

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# 博士論文審査結果の要旨

# Summary of the results of the doctoral thesis screening

出願者の論文「ADN 系イオン液体の燃焼に関する実験研究」は，高性能かつ低毒性の一液式推進剤の有望な候補として期待されているものの難揮発性かつ高粘性であるアンモニウムジニトラミド（ADN）系イオン液体について，一次元的な燃焼実験及び数値計算に基づく解析によってその燃焼過程を解明し，さらに宇宙機用スラスタへの適用性を評価したものである．

その成果として，ADN 系イオン液体の燃焼波構造は，液相，気液二相，気相で構成されており，その燃焼過程は，低温から順に液面の昇温，ADN の熱分解及び尿素の蒸発，MMAN の解離反応，液相の硝酸アンモニウムの解離反応，メチルアミンと硝酸の発熱反応による第一段階目の燃焼，窒素化合物による NOx の還元反応を伴った第二段階目の燃焼であるという解釈を示している．

またスラスタへの適用性については，ADN 系イオン液体の物性や前述の燃焼過程に関する新しい解釈も踏まえて，4 つの観点から評価している．点火方法については，ADN 系イオン液体の難揮発性や着火応答性の観点から噴霧点火が望ましく，保炎するためには予混合燃焼と同様に再循環流の形成が必要であるとしている．燃焼の完結性（燃焼効率）に影響する滞留時間に関しては，ADN 系イオン液体の昇温時間が支配的な要因であり，従来型スラスタの一般的な滞留時間を大きく超えることから，昇温に要する特性長さを低減する機構が求められるとしている．また，ヒートソークバックを抑制するキャピラリーチューブと異物汚染を防ぐフィルタを適切に設計することによって，高粘性の ADN 系イオン液体についても従来型スラスタと同レベルの供給圧力が設定可能であるとしている．以上により，昇温に課題はあるものの，少なくとも推力 1N 級の小推力スラスタへ適用は可能と結論付けている．

この論文に対して，審査委員で審査を実施し，以下のような結論を得た．

- 通常は固体であるADN, MMAN, Ureaからなるイオン液体に着目し，液体ロケット燃料として適切な配合比率が存在することを解析により見いだし，実験的に燃焼特性（燃焼速度およびその圧力依存性），高い燃焼完結性を示している．
- そのイオン液体の燃焼過程について，液相・気相での段階的な燃焼機構を実験および解析により解明している点は，新規性が極めて高い．
- 同液体のスラスタへの適用に関する考察については，燃焼機構の解明を踏まえて，微粒化・昇温過程について，解析的考察を行い，基本的成立性を示している．
- 特に，微粒化については，線香花火からのアナロジーにより，従来とは異なる分裂モデルの可能性を，予備実験により示唆している．
- また，微粒化・昇温・燃焼の各過程の中で昇温時間が支配的になることを示している．長い昇温時間はスラスタ長にとって不利になることから，ヒータ設置などによる解決策を提案している．
- これらにより，低毒性であるが，難燃性・高粘度のADN系イオン液体を液体推進系と

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して使用することの基本的成立性を示しており，この面においても新規性が認められる.

以上のことから，出願された論文は博士（工学）の学位論文として相応しいレベルに達していると判定した.

さらに審査委員全員参加の下で，関連する専門分野及びその基礎分野についての口述試験,英語論文要旨の提出による英語試験及び学位論文の公開発表会を実施した.出願者は,学位論文の要点を明確に説明し,質疑応答に対しても真摯に的確な対応を行ったことから,専門及び基礎分野に対する知識及び学力は十分であると判断された．さらに，内容についても, ADN系イオン液体の燃焼過程の解明とその宇宙機用スラスタへの適用性に関する考察の両面について十分な成果を挙げており，オリジナリティが十分に認められると判断した．また出願者は，学位論文の内容について，これまでに５回の国際会議発表，２編の英語論文が学術雑誌に掲載されていること，さらに英語の論文要旨も提出済みであり，語学力も十分な水準にあると判断された.

以上より，出願者は学位授与に相応しい知識と学力を有していると認められ，審査員全員一致で合格と判定した.

以上