Research paper

Preparation and thermal decomposition behavior of ammonium dinitramide-based energetic ionic liquid propellant

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Abstract

The liquefaction of highly energetic materials without the use of solvents is one means of increasing the performance of liquid propellants. In the present study, energetic ionic liquid propellants (EILPs) were prepared by forming eutectic mixtures of the oxidizer ammonium dinitramide (ADN) with monomethylamine nitrate (MMAN) and urea. The physical properties, decomposition mechanism and combustion mechanism of these mixtures were assessed with the aim of eventually determining ignition methods and optimizing performance. Chemical equilibrium computations predict that the performance of ADN-based EILPs will be superior to that of hydrazine monopropellants. Thermal analysis of these EILPs during constant rate heating demonstrates that the majority decompose to generate various gases, including nitrous oxide, nitrogen dioxide, isocyanic acid, ammonia, carbon dioxide and water. Kinetics analysis also indicates that these EILPs can be safely stored for prolonged time periods at room temperature.

Keywords: ionic liquid propellant, ammonium dinitramide, thermal decomposition

1. Introduction

Energetic materials are now evaluated not only with regard to their performance but also their current or potential environmental and health impacts. Hydrazine is widely used as a liquid monopropellant for the attitude control of rockets and space satellites, since it readily decomposes to form hot gases in the presence of catalysts and may be stored at room temperature. However, this compound is also highly toxic and generates combustible vapors, and so is difficult to handle and reduces the operability of space crafts. To eliminate these problems, it would be beneficial to replace hydrazine with a less toxic monopropellant. As a result, many studies have pursued the development of new, environmentally safe (or "green")

propellants containing less toxic ingredients that provide suitable performance in terms of burning rate, specific impulse, stability and other important performance parameters. Among the potential candidates, hydroxyl amine nitrate (HAN, $NH_3OH^+NO_3^-$),[1)−5)] hydrazinium nitroformate [HNF, $N_2H_5^+C(NO_2)_3^-$][6)−10)] and ammonium dinitramide [ADN, $NH_4^+N(NO_2)_2^-$][11)−15)] are the most promising new energetic materials because they have the capacity to generate more energy than hydrazine while being less toxic. These three compounds have melting points of 48, 124 and 92°C, respectively and so are solids at room temperature and must be transitioned from solid to liquid before use. In many previous studies, HAN and ADN have been dissolved in water and/or methanol to
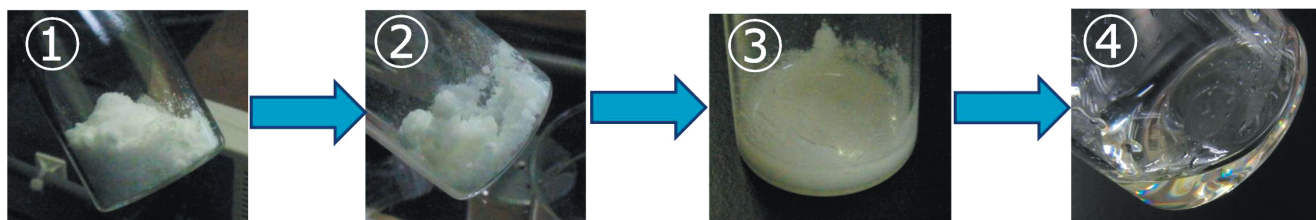
Figure 1　Preparation of an energetic ionic liquid propellant using freezing point depression.

allow their use as liquid propellants[16)-21)]. These propellants are now situated near realization. These propellants are close to being suitable for use in real world applications, but continue to face challenges related to ignition and combustion, due to the presence of the solvent.

In the present study, we attempted to prepare high performance liquid propellants by synthesizing ionic liquids to make energetic ionic liquid propellants, or EILPs. The most readily accepted definition of an ionic liquid is a salt or mixture of salts having a melting point below 100°C[22)]. The first ionic liquid to be reported was ethylamine nitrate [$(C_2H_5)NH_3^+ NO_3^-$], which is a colorless to slightly yellowish liquid with a melting point of 12°C and was described by Walden in 1914[23)]. Since ionic liquids have several unique properties, including low volatilities, low liquidus temperatures and high ionic conductivities, they have been researched with regard to applications in medicine[24)], electric batteries[25)], recyclable energy devices[26)] and functional materials[27)]. The various properties of ionic liquids may also allow these compounds to serve as liquid propellants, since their low volatility decreases the risk of exposure during handling as well as the threat of explosion. In addition, their low liquidus temperatures could enable the preparation of liquid propellants without the use of solvents. As such, these compounds may perform well as liquid propellants, making it possible to extend space mission time spans and reduce propellant tank volumes. Therefore, EILPs could drastically reduce the costs associated with the manufacture and use of rockets and satellites.

Based on the above, energetic ionic liquids (EILs) are expected to have applications in various devices. Indeed, over the past several years, a large number of publications have described energetic ionic salts and EILs[28)-30)]. Many of these EILs are pure compounds consisting of the cation of a nitrogen-containing hetero ring compound together with a nitrate, perchlorate, dinitramide or azide anion. These compounds tend to have low melting points because the interionic interaction between the anion and cation is minimal. However, there are concerns that problems may arise with EILs, including high sensitivity and difficulty in achieving sufficient purity. In the present work, we obtained ionic liquids by preparing eutectic multicomponent mixtures. This type of ionic liquid can be prepared only by mixing materials (Figure 1) and allows for a diverse range of performance and properties. A typical example is a 1 : 2 molar mixture of choline chloride and urea, which has a melting point of 12°C, a value that is far less than the melting point of either pure choline

chloride (302°C) or urea (133°C)[31)]. To date, this concept has not been applied to the synthesis of liquids for space propulsion and our goal was therefore to assess the applications of EILPs as monopropellants. In this paper, EILPs were prepared by forming eutectic mixtures and their thermal behaviors were investigated. Based on the results, the future potential of EILPs is discussed.

2. Calculation and experimental
2.1 Preparation of EILPs

To ensure sufficiently high performance, the EILPs fabricated in this study were each composed of a mixture of an oxidizer and a fuel. We initially examined the optimum compositions of such EILPs. As noted, it is necessary to employ highly energetic compounds so as to ensure sufficient performance and it is also a requirement to obtain a eutectic system so as to obtain a suitably low melting point. One promising new oxidizer that may be employed for this purpose is ADN (Figure 2), the primary physical properties of which are summarized in Table 1. ADN is a readily soluble, colorless, crystalline solid at room temperature with a melting point of 92°C. It offers high energy content, good oxygen balance, low toxicity and a relatively low melting point and, for these reasons, the authors have previously researched the synthesis, decomposition and combustion of this compound[32)-40)]. ADN has been found to form eutectics with various nitrates, including ammonium nitrate (AN), potassium nitrate and sodium nitrate, and the melting points of these mixtures have been shown to decrease to approximately 60°C[32), 33)]. In this study, ADN-based EILPs were prepared by mixing ADN with monomethylamine nitrate (MMAN) and urea, both acting as fuels, since these compounds have
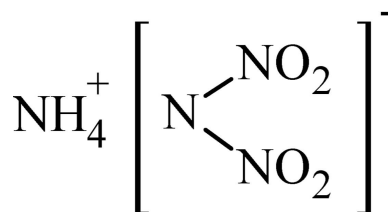

Figure 2　The structure of ADN.

Table 1　The physical properties of ADN.

Molecular weight	124.07
Density (solid) (25°C) [g cm^{-3}] (X-ray diffraction)[42)]	1.820
Density (liquid) (100°C) [g cm^{-3}] (pycnometer)[43)]	1.560
Melting point [°C][44)]	91.5−92.5
Heat of formation [kJ mol^{-1}][13)]	−148
Oxygen balance [%]	+25.8
Critical relative humidity (25°C) [%][42)]	55.2

been shown to form eutectics when combined with AN[41].

The ADN used in this work was obtained from the Hosoya Pyro-Engineering Co., Ltd. and urea was purchased from Wako Pure Chemical Industry, Ltd. Both were used as received without further purification. MMAN was synthesized from the room temperature reaction of a 40% aqueous solution of monomethylamine with nitric acid (1.42 g cm^{-3}), both obtained from Wako Pure Chemical Industry, Ltd., followed by drying of the product under vacuum.

2.2 Performance calculations

The heat of combustion (ΔH) and vacuum specific impulse (I_{vac}) of the ADN-based EILPs were calculated using the NASA-CEA chemical equilibrium program[45] and compared to the values for a standard hydrazine monopropellant. During ΔH calculations, the temperature and pressure were set at 298 K and 0.1 MPa. The I_{vac} calculations were based on the frozen flow assumption, with a combustion chamber pressure of 1 MPa and a nozzle open area ratio of 50.

2.3 Thermal decomposition behavior of ADN-based EILPs

The thermal behaviors of ADN-based EILPs were characterized using sealed cell differential scanning calorimetry (SC-DSC, TA Instrument, Q200). Each sample, weighing approximately 1 mg, was loaded into a stainless steel cell and sealed under air, then heated from −35 to 400°C at 10 K min^{-1}.

During the heating of samples, mass loss, thermal behavior and decomposition gases were analyzed simultaneously using thermogravimetry-differential thermal analysis-infrared spectrometry (TG-DTA-IR) instrumentation. This apparatus consisted of two parts : a TG-DTA instrument (Shimadzu Co., Ltd., DTG-60) and an IR spectrometer (Shimadzu Co., Ltd., IRPrestige-21). Samples of approximately 3 mg were loaded into open aluminum pans and then heated to 350°C at 5 K min^{-1} under an argon flow of 100 mL min^{-1}. The gases generated via thermal decomposition during TG-DTA analysis were transferred to a gas cell for IR analysis through a stainless steel tube. The transfer tube and IR gas cell temperatures were set at 200°C.

To assess the stability of the ADN-based EILPs, decomposition kinetics were determined using Calvet calorimetry (SETARAM Instrumentation, C80). Samples of approximately 50 mg were loaded into sealed stainless steel pans with glass inserts and heated to 300°C at 0.063, 0.125, 0.25 or 0.5 K min^{-1}. The kinetic parameters and long term stability were calculated using the AKTS-Thermokinetics software package[46].

3. Results and discussion
3.1 Preparation and performance prediction of ADN-based EILPs

Figure 3 presents a photographic image showing the typical results obtained when mixing equivalent masses of ADN, MMAN and urea in a vial followed by manual

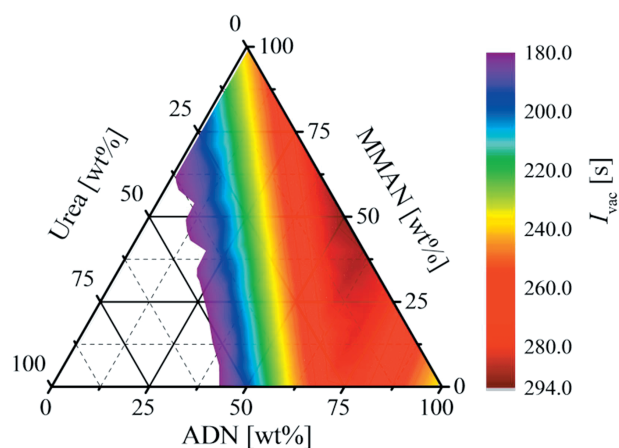**Figure 3** An ADN/MMAN/urea (1/1/1) mixture at room temperature.

Figure 4 Vacuum specific impulse values calculated for ADN /MMAN/urea mixtures using NASA-CEA.

agitation. In such cases, the mixture liquefied after only a few minutes. ADN-based EILPs could therefore be prepared by mixing ADN with MMAN and urea at room temperature. Figure 4 summarizes the I_{vac} values calculated for ADN-based EILPs using NASA-CEA. The I_{vac} calculated for hydrazine monopropellant under the same conditions was 231 s. In contrast, the I_{vac} of the EILP was found to range as high as 293 s, a value that is significantly higher than that of hydrazine. An ADN/ MMAN/urea mixing ratio (by mass) of 40/40/20 (EILP 442) generated a particularly low melting point, such that the mixture remained liquid at 0°C, and had an I_{vac} of 251 s (8.7% higher than that of hydrazine). An ADN/MMAN/ urea=60/30/10 mixture (EILP631) whose melting point is near the room temperature (25°C) had the highest I_{vac} (287 s, 24% higher than that of hydrazine) among the ADN-based EILPs and remained liquid at near room temperature. The ΔH values determined for EILP442 and hydrazine at 298 K and 1 bar from NASA-CEA calculations were −8.0 and −1.9 kJ g^{-1}, respectively.

The SC-DSC results obtained from EILP442 at 10 K min^{-1} are shown in Figure 5, in which three heat generation events are evident from approximately 130 to 290°C and the peak temperature were 193, 310 and 345°C, respectively. The total heat value of these peaks was 3.5 kJ g^{-1}. Although EILP442 is not the highest performance composition, its decomposition heat was found to exceed the ΔH value of hydrazine monopropellant. Thus it is evident that ADN-based EILPs exhibit higher energy and
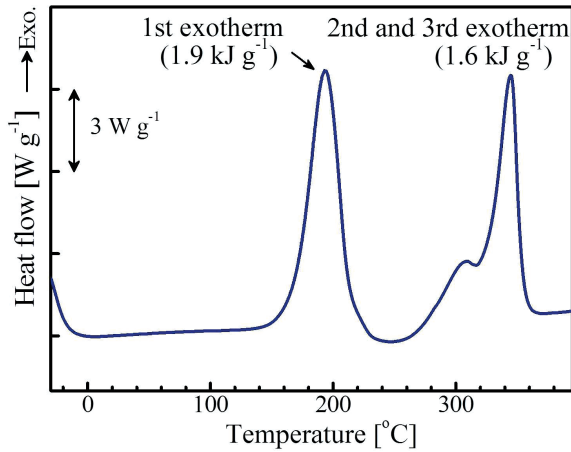
Figure 5　SC-DSC plot for EILP442.

Figure 7　IR spectrum of evolved gas from the decomposition of EILP442.

I_{vac} on the basis of both theoretical and experimental evaluations.

3.2 Thermal decomposition behavior

The TG-DTA results obtained from ADN-based EILP 442 are shown in Figure 6. Here a slight endotherm with concurrent mass loss is observed beginning at 110℃, with transition to the main exotherm at 140℃. The mass loss eventually reaches nearly 100%. Figure 7 presents the IR spectrum of the evolved gas from the decomposition of this same sample, in which peaks attributed to nitrous oxide (3500−3400, 2250-2100 and 1350−1200 cm^{-1}), nitrogen dioxide (1650−1550 cm^{-1}), isocyanic acid (2300−2200 cm^{-1}), ammonia (3350, 970 and 930 cm^{-1}), carbon dioxide (2400−2300 cm^{-1}) and water (broad peaks at 4000−3500 and 1800 −1400 cm^{-1}) are observed. These thermal analysis results demonstrated that almost all of the EILP samples generated these gases upon heating, indicating that ADN-based EILPs are potentially highly efficient propellants which does not produce condensed residue from the reactions. Isocyanic acid is known to be one of the main products of urea decomposition, and is produced at approximately 110℃ through an endothermic reaction[47),48)], which matches the temperature of the endothermic reaction seen in the TG-DTA data for EILP 442. Therefore, the thermal decomposition of these ADN-based EILPs is thought to begin with the decomposition of urea. The main reaction is exothermic even in an open
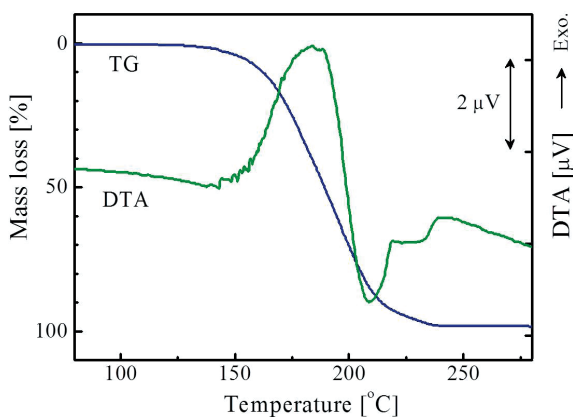
pan, indicating that the decomposition of these EILPs proceeds exothermically in the condensed phase.

3.3 Storage stability of ADN-based EILPs

The stability of the highest performing composition, EILP631, was subsequently investigated and the thermal behaviors obtained from the C80 analysis are presented in Figure 8. From about 110℃, an exotherm with an associated heat release of 2.8 kJ g^{-1} is observed. Figure 9 plots the dependence of the activation energy of the EILP 631 thermal decomposition as a function of reaction progress, as calculated from the results of the C80 analysis at different heating rates using the Friedman method[49)]. These calculations were based on the Equation (1):

$$\ln\left(\frac{d\alpha}{dt}\right) = \ln\left[A(\alpha)f(\alpha)\right] - \frac{E_a(\alpha)}{RT} \tag{1}$$

where α is the reaction progress, t is time, k is the reaction rate constant, $f(\alpha)$ is the reaction model (a function of the reaction progress), A is the pre-exponential factor, R is the gas constant and E_a is the activation energy. In this study, α was determined from the ratio of the heat value at a given time, Q, to the total heat release of the first exothermic peak, Q_{tot}, as in Equation (2).
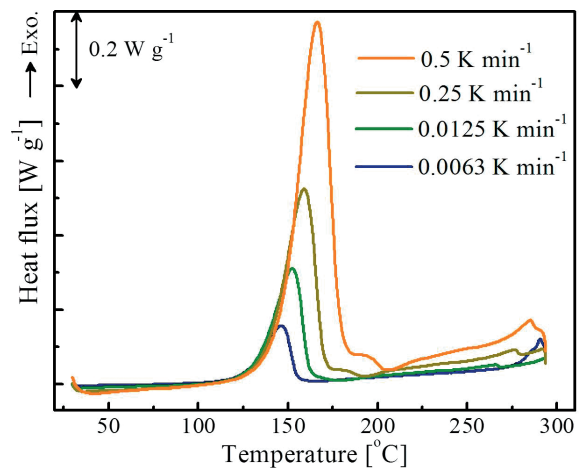
$$\alpha = \frac{Q}{Q_{tot}} \tag{2}$$

Figure 6　TG-DTA plots obtained from EILP442.

Figure 8　Exothermic behavior of EILP631 from C80 analysis.

Figure 9 Activation energy plots for EILP631 and pure ADN.

Figure 10 Predicted stability of ADN-based EILP631 during isothermal storage.

For a given value of α, the relationship between ln (da/dt) and T^{-1} at several heating rates gives a straight line, and the activation energy at each α can then be calculated from the slope of this line. For comparison, the E_a values for pure liquid ADN were also calculated using the same method and are provided in Figure 8. It is evident that the relationship between activation energy and α for EILP631 is different from that of pure ADN. The E_a of EILP631 is constant between 130 and 145 kJ mol^{-1}, up to $\alpha = 0.7$. In contrast, the E_a of pure ADN decreases moving from 130 kJ mol^{-1} to the end of the reaction. In addition, the value of the EILP631 activation energy is generally higher compared with the value for pure ADN at every α. These activation energies indicate that the decomposition mechanism of ADN-based EILPs is different from that of pure ADN, and that the EILPs are more stable. From previous studies, the reactivity of ADN is decreased by the presence of AN or urea[11),32)−34)] because these additives reduce the concentrations of unstable molecules such as dinitramic acid and nitrogen dioxide. In this composition, therefore, both the nitrate anions in MMAN and the urea are thought to work to stabilize the EILPs.

The stability of EILP631 at temperatures around ambient was predicted from kinetic parameters using the AKTS-Thermokinetics software package. Figure 10 shows the relationship between the extent of decomposition and storage time. As an example, EILP631 is predicted to decompose by approximately 2.5% over three years storage at ambient temperature. It is well-known that ADN decomposes to AN during long term storage[32),33)], and that the formation of AN significantly decreases the propulsion performance[34),50),51)]. On the assumption that all of the exothermic reaction seen in the C80 analysis was due to the decomposition of ADN to AN, the associated decrease in I_{vac} was predicted to be only about 0.1% based on calculations using NASA-CEA. These results indicate that ADN-based EILPs can be stored at room temperature for prolonged time periods with no deleterious effects.

4. Summary

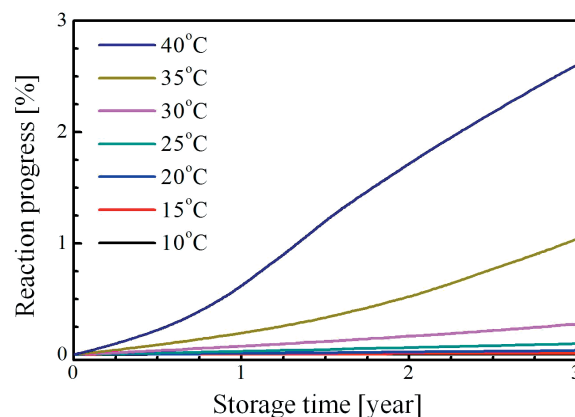As a means of developing high performance, low toxicity liquid propellants, this study focused on eutectic EILs. The EILPs synthesized in this study have a number of desirable properties, including low volatilities, low liquidus temperatures and simple preparation. Thus we successfully prepared EILPs based on the highly energetic material ADN without the use of solvents.

The results of SC-DSC and chemical equilibrium calculations both demonstrate that ADN-based EILPs will exhibit higher performance than hydrazine monopropellants. The majority of these mixtures react to generate gases such as nitrous oxide, nitrogen dioxide, isocyanic acid, ammonia, carbon dioxide and water upon heating and thus represent highly efficient propellants. These EILPs also exhibit suitable storage stability in the vicinity of ambient temperature.

There are still many challenges associated with the utilization of EILPs due to their unique properties. One of these is determining a suitable means of ignition. Generally, ionic liquids are low volatility liquids, which makes ignition difficult. The use of catalysts, such as are applied with hydrazine monopropellants, is not viable because EILPs do not contain a solvent and their flame temperature is too high. As an example, calculations using NASA-CEA predict an adiabatic flame temperature of 2657 K for EILP631, compared to 873 K for hydrazine. In addition, ionic liquids generally have high viscosities and therefore it will be necessary to develop a new method of igniting these novel propellants. A second issue is the design of EILPs. It will be vital to fully understand the effects of ingredient properties on the characteristics of EILPs, such as melting point, density, viscosity and ignition performance, so as to develop propellants with the desired performance parameters. Our group is currently carrying out further investigations of ADN-based EILPs based on ADN/MMAN/urea[52)−54)] in an attempt to solve these distinctive technical issues[55)].

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