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Table of contents

LIST OF FIGURES

LIST OF TABLES

NOMENCLATURE

Abbreviations

ADN	Ammonium Dinitramide
APU	Auxiliary Power Unit
BET	Brunauer-Emmett-Teller
CEA	Chemical Equilibrium with Application
DAQ	Data Acquisition Device
DMAZ	2-Dimethylaminoethylazide
EM	Engineering Model
ERPS	Electric Rocket Propulsion System
ESA	European Space Agency
GHS	Global Harmonized System
GNSS	Global Navigation Satellite System
GPIM	Green Propellant Infusion Mission
HAN	Hydroxylammonium Nitrate
HNF	Hydrazinium Nitroformate
HRPS	Hybrid Rocket Propulsion System
HTP	High-Test Peroxide
IR	Infrared
KITSAT-1	Korea Institute of Technology Satellite-1
KSLV-II	Korea Space Launch Vehicle
LD_{50}	Lethal Dose
LPF	Low Path Filter
LRPS	Liquid Rocket Propulsion System
MI	Mineral Insulated
NASA	The National Aeronautics and Space Administration
OMS	Orbital Maneuvering System
PRISMA	Prototype Research Instruments and Space Mission technology Advancement

RCS	Reaction Control System
SEM	Scanning Electron Microscopic
SIA	Satellite Industry Association
SRPS	Solid Rocket Propulsion System
TGA	Thermogravimetric Analysis
TMS	Thrust Measurement System
TRL	Technology Readiness Level

<u>Unit</u>

bar	Bar
℃	Degree Celsius
℃/min	Degree Celsius Per Minute
°F	Degree Fahrenheit
ft/sec	Foot Per Second
g	Gram
g/s	Gram Per Second
h	Enthalpy
K	Kelvin
kJ/mol	Kilojoule Per Mol
m/s	Meter Per Second
m^2/g	Square Meter Per Gram
mg/kg	Milligram Per Kilogram
mg/l	Milligram Per Liter
mm	Millimeter
mN	Millinewton
ms	Millisecond
N	Newton
Ns/kg	Newton-second per kilogram
ppmV	Parts per million by volume
s, sec	Second

t	Time
W	Watt

Symbols

A	Frequency Factor or Pre-Exponential Factor
AC	Alternating Current
A_t	Nozzle Throat Area
C^*	Specific Velocity
DC	Direct Current
E_a	Activation Energy
g_0	Standard Gravity
I_{SP}	Specific Impulse
I_{VAC}	Vacuum Specific Impulse
k	Rate Constant
$\dot{m}$	Mass Flow Fate
P	Pressure
$P_{chamber}$	Chamber Pressure
P_{max}	Maximum Pressure
P_{peak}	Peak Pressure
P_{prop}	Propellant Supply Pressure
R	Gas Constant
T	Temperature
$T_{chamber}$	Chamber Temperature
T_m	Peak Temperature
T_{max}	Maximum Temperature
β	Heating Rate
ε	Expansion Ratio
Φ	Diameter

Chemistry

Al_2O_3	Aluminium Oxide
C_2H_5OH	Ethanol
$C_3H_8O_3$	Glycerol
CH_3OH	Methanol
CO	Carbon Monoxide
CO_2	Carbon Dioxide
Cu	Copper
H_2	Hydrogen
H_2O	Hydrogen Dioxide
H_2SO_4	Sulfuric Acid
HCl	Hydrogen Chloride
HNO_3	Nitric Acid
Ir	Iridium
KOH	Potassium Hydroxide
KSO_3NH_2	potassium Sulfamate
N_2	Nitrogen
N_2H_4	Hydrazine
N_2O_5	Dinitrogen Pentoxide
NH_2CONH_2	Urea
NH_2COOCH_3	Methyl Carbamate
NH_2NO_2	Nitramide
NH_3	Ammonia
$(NH_4)_2SO_4$	Ammonium Sulfate
NO_2BF_4	Nitronium Tetrafluoroborate
NOx	Nitrogen Oxides

ABSTRACT

Design and evaluation of monopropellant thruster for ammonium dinitramide based low-toxicity propellant test

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본 논문에서는 저 독성 추진제 및 촉매의 합성, 추진제 성능평가를 위한 추력기의 설계/제작을 진행 하였고 해당 추력기를 바탕으로 추진제 및 촉매의 성능 검증을 위한 추력기 및 추력 측정 시스템(TMS)을 설계/제작하여 성능 검증을 수행하였다.

현재 미국, 유럽, 중국 등 우주개발 선진국에서는 기존 발사체 및 인공위성 자세제어용 추력기에 활용되어 온 하이드라진 기반 추진제의 대체를 추진제의 연구개발이 활발하게 진행 중이다. 이를 위해서 저 독성 추진제로 활용이 가능한 다양한 후보 물질들이 있으며 이들의 물리적 요구조건으로는 비교적 독성이 낮고 하이드라진 기반의 추진제와 성능이 유사하거나 뛰어난 특성을 가질 것을 요구하고 있다. 이러한 조건을 만족하는 대표적인 저 독성 추진제 후보군으로는 Ammonium dinitramide(ADN), Hydroxyl ammonium nitrate(HAN), 그리고 Hydrazinium nitroformate(HNF)등이 있다. 하지만 국내에서는 해당 후보물질을 기반으로 한 저 독성 추진제와 관련하여 연구의 기술 성숙도(TRL)가 아직 기초 단계에 머물러 있으며 해당 추진제와 관련한 연구기반이 부족한 실정이다. 이를 위해서 저 독성 추진제 및 촉매의 선정에서 부터 추진제의 시험 및 성능평가를 위한 저 독성 추진제 테스트 시스템을 설계 및 제작을 진행 하였다.

ADN 기반 단일 추진제와 촉매의 합성을 수행하는 한편 저 독성 추진제와 촉매의

분해특성을 파악하기 위해 저 독성 추진제와 귀금속 촉매 표본을 Thermogravimetric Analysis(TGA)를 통해 분석을 수행하였다. 그리고 TGA를 통해 수집한 데이터를 바탕으로 등전환법을 통해서 추진제의 촉매 간 분해 특성을 비교분석하였다.

성능평가를 통해 선정된 촉매와 추진제의 연소실험 및 추력기 성능 평가를 수행하였다. 이를 위해서 1 N급 추력기의 설계를 진행하였다. 추력기의 설계를 위해 추진제의 화학평형을 Chemical Equilibrium with Application(CEA)을 통해 계산하였고 이를 통해 얻어진 수치를 바탕으로 추력기를 설계 및 제작하였다.

최종적으로 촉매, 추진제 및 추력기 실험결과를 분석 및 평가하였다. 촉매의 성능평가를 위해 Scanning Electron Microscopic(SEM) 및 Brunauer-Emmett-Teller(BET) 분석을 진행하여 실험 전/후의 촉매의 성능 변화를 분석하였다. 그리고 선정된 촉매와 추진제간의 연소 성능평가를 시험용 추력기에 적용하여 진행하였다. 1 N급 추력기에 저 독성 추진제의 적용 시험을 진행하였다. 이때 TMS는 펜듈럼방식의 추력측정시스템으로 추력 마진 및 기타 충격으로부터 보호하기 위해 충분히 마진을 주고 설계하였다. 그리고 ADN 기반 저 독성추진제의 특성중 하나인 무연/무 화염 분해현상을 Mid-range Infrared(IR) 시각화를 진행하여 저 독성 추진제 분해현상 관측 및 성능 분석을 수행하였다.

I. Introduction

Konstantin Tsiolkovski (September 5, 1857-September 19, 1935) laid the theoretical foundation for space travel and rocket propulsion, human civilization has constantly attempted to challenge its advance into space[11]. This challenge triggered a full-scale space development competition between the United States and the Soviet Union during the Cold War, starting from the beep sound transmitted by Sputnik 1 on orbit. Since then, they has successfully carried out various missions beyond the Earth's gravity, along with large and small events and accidents in human civilization. In particular, through the Soviet Union's Vostok project and the Voskhod project, he reached the Soyuz Plan and floated the space station on orbit through the Salute Program. The United States succeeded in landing on the moon for the first time in the Apollo Project after the Mercury Plan and Gemini Plan, and boasted the technology of space development in both countries through these successes of big plans. Even after the end of the Cold War, as the demand for various operating ranges increased, the Space Development Organization was launched competitively around the world, and it is currently being researched to support various missions across civilian, civilian, and military forces based on the technologies accumulated at that time[15].

In particular, Fig. 2 was published by the Satellite Industry Association (SIA), a private organization, and according to the data, the size of the satellite industry, which was about $89 B in 2005, has grown significantly to about $227 B by 2018[1]. In particular, the size of the industry has doubled compared to 2008. This shows a significant increase in the size of the space launch vehicle and satellite manufacturing industry in the space projectile and satellite market. This increase in industrial size is attributed to increased demand for various technologies and environments linked to satellites, from military satellites [prospection, navigation (table. 1) and telecommunications] to civilian satellites [earth observation (sea surveillance, environmental pollution identification, forest identification, crop status check), weather satellites, communication satellites (general communications, broadcasting signal transmission and reception), and space telescopes]. And this increase in market size has also affected private companies' entry into the space development market. With Space X at the forefront, private companies such as ULA and Blue Origin have begun to enter the national space launch vehicle development project[2-5].

Meanwhile, in Korea, The space development plan like Fig. 3 is in progress. Beginning with KITSAT-1 in 1992, as a latecomer, the space development plan was under way[6]. In recent years, based on the mid- to long-term plan for space development, R&D has been conducted to achieve the goal by establishing six key tasks such as technology development of space launch vehicles, construction of a national satellite navigation system, and space exploration [7]. Based on its experience in launching and operating the Naro rocket, it successfully launched the Nuri test vehicle in 2018 as well as secured the possibility of launching satellites that had relied on other countries' launch vehicle. The examples described above confirm that active research is being carried out on spacecraft and satellite-related materials and components for space development at home and abroad. As spacecraft and satellite technologies are a high-tech industry, the need to secure basic and core technologies in the field is gradually increasing.

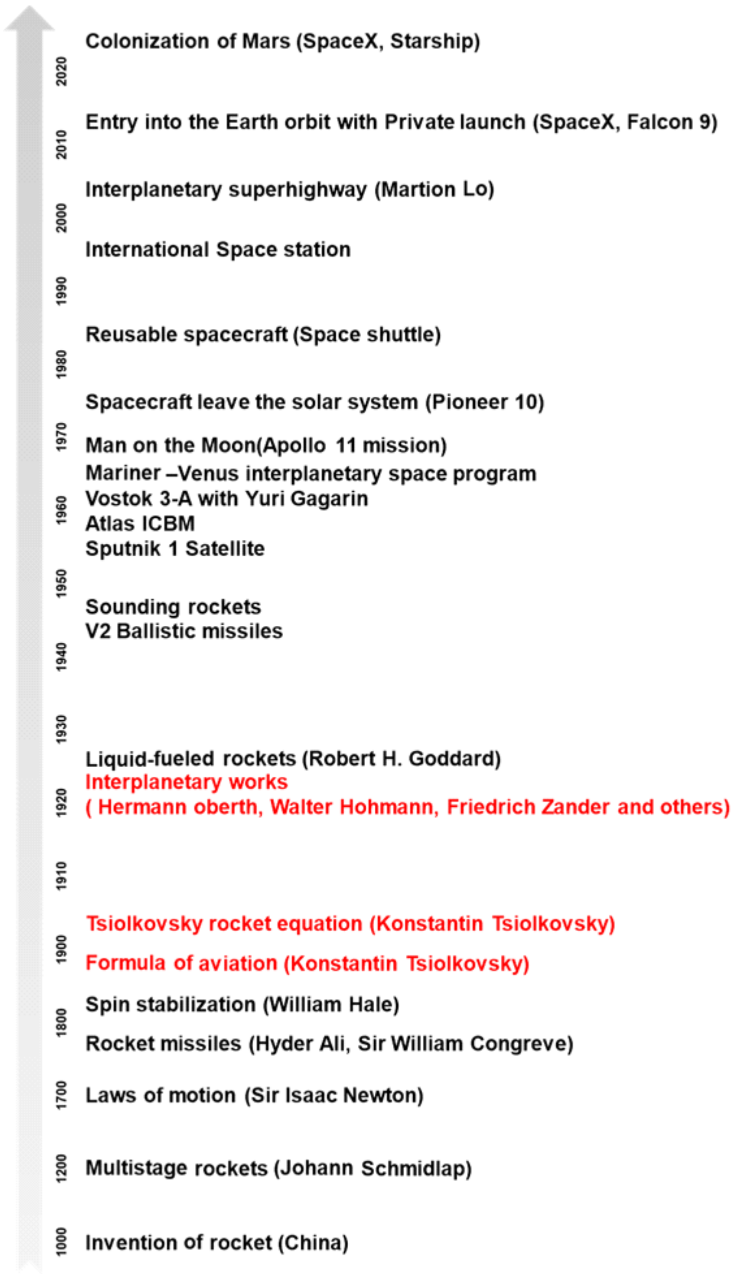


Fig. 1 Historical timeline of space exploration and development

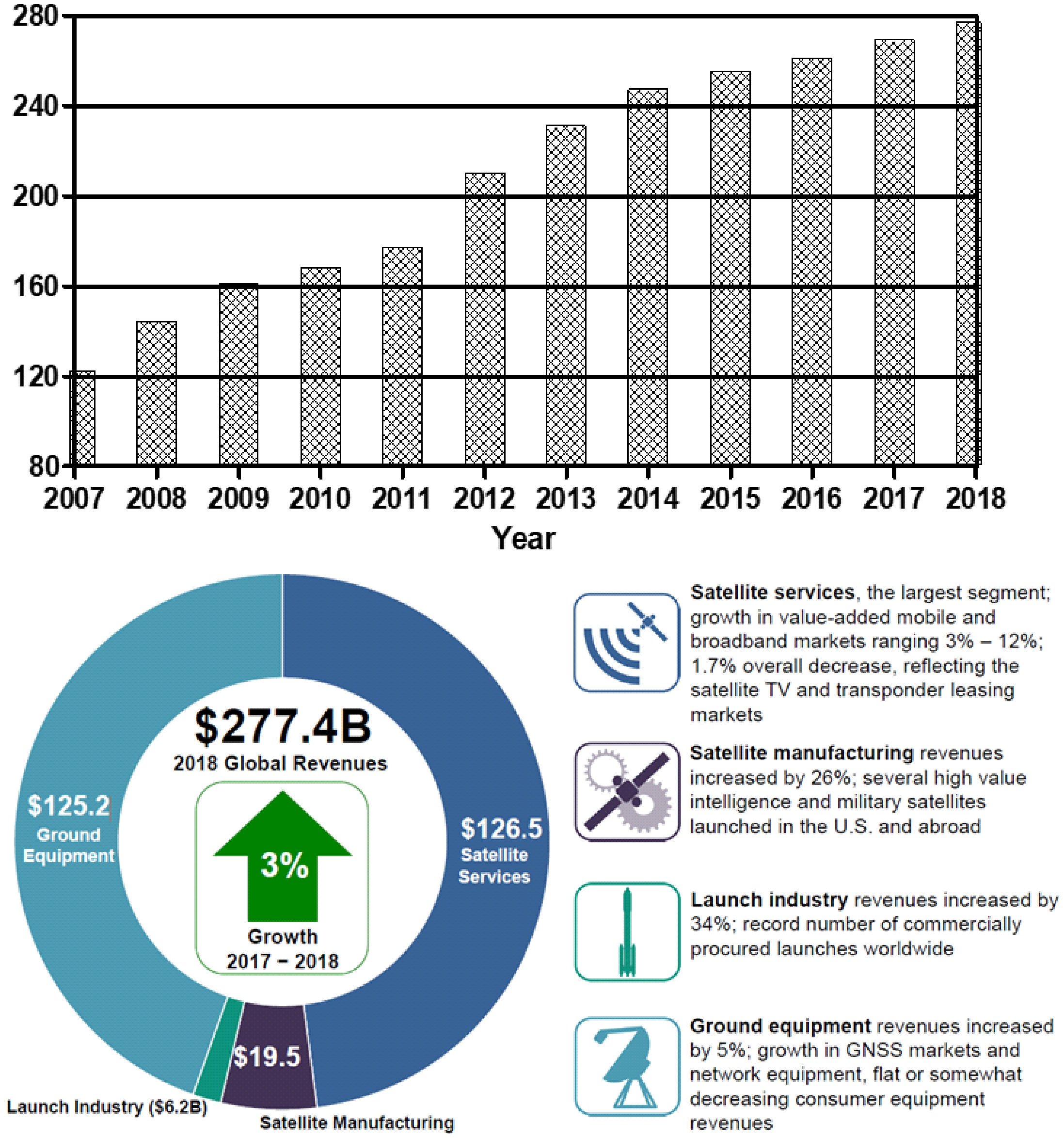


Fig. 2 Global satellite industry revenues[1]

Table. 1 Status of global navigation satellite system(GNSS) in each country

GPS (USA)	
No. of Satellites	31
First satellite launch	1978
Coverage	Global
Precision	5m

BEIDOU (CHINA)	
No. of Satellites	35
First satellite launch	2000
Coverage	Global
Precision	10m for public 10cm for military

GLONASS (RUSSIA)	
No. of Satellites	24
First satellite launch	1982
Coverage	Global
Precision	5m to 10m

GALILEO (EU)	
No. of Satellites	40
First satellite launch	2011
Coverage	Global
Precision	1m for public 1cm for military

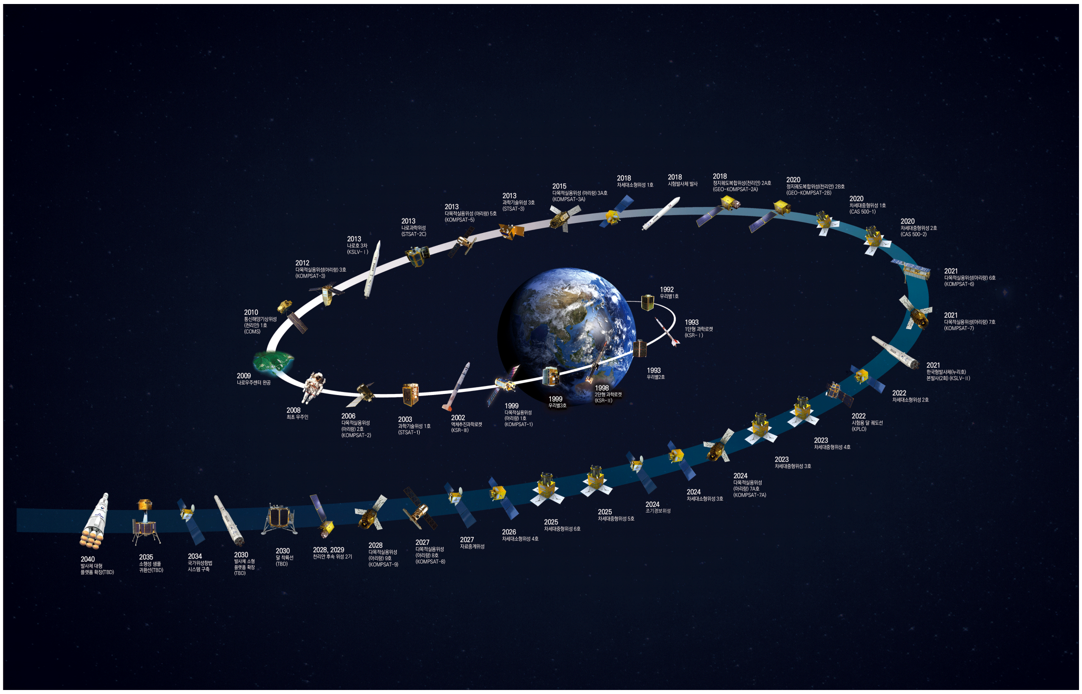


Fig. 3 Korea's long-term plan roadmap for space development[7]

A. Classification of Rocket Propulsion

The main systems that make up the rocket/satellite can be classified as shown in Fig. 4.

- Structure system
- Payload system
- Guidance & communication system
- Propulsion system

The basic and key technology for rockets and satellites to reach orbit is propulsion systems. The rocket/satellite achieves its goal through the power it gains through the propulsion system. In this case, propulsion refers to the reaction force generated by the injection of a substance generated inside an object. In particular, rocket propulsion means obtaining the force generated by the injection of material supplied from the rocket (generally, gas injection from the combustion process of fuel). This force is used as a means to overcome the drag generated during the course of an object by moving a stationary object or changing its speed[21, 22].

- Launch – Accelerate the spacecraft to reach its desired orbit from Earth (low, medium, high, and stationary orbit).
- Orbit insertion – Place the spaceship in mission orbit.
- Orbit maintenance and maneuvering – To maintain or move a spaceship into its target orbit.
- Attitude control – Torque generated to keep the spacecraft in the target direction

To perform these detailed tasks, the mission of the space launch vehicle/satellites is performed from the main engine through an externally mounted thruster as shown in Fig. 4-7.

Rocket propulsion systems can classified according to the various types like basic propulsion method (thermal, non-thermal), energy sources (chemical, nuclear or solar energy), roles (booster stage, multistage rocket, posture control, orbit maintenance, etc.), size, propellant type, configuration type, etc. Among them, propellant-based propulsion systems can be classified as shown in Table 2.

1. Cold Gas Propulsion

Cold gas propulsion system is the most classic type of propulsion system, which generates thrust by ejecting stored gas. However, compared to liquid/solid-based propellants, it has the disadvantage of low density and poor storage, and the performance index of propulsion systems such as specific impulse is generally low.

2. Chemical Rocket Propulsion

a. Solid Rocket Propulsion

Solid Rocket Propulsion System(SRPS) is the system in which solid propellants are stored in the combustion chamber. This solid propulsion system is ignited and burned until the propellant is depleted inside the combustion chamber. The combustion gas is accelerated by the converging-diverging(C-D) nozzle. Solid rocket system is relatively easy to operate but difficult to control thrust. Also, depending on the composition of the propellant, toxic substances may be ejected from the exhaust gas.

b. Liquid Rocket Propulsion

The Liquid Rocket Propulsion System (LRPS) is the system that stores propellants (fuel and oxidizer) in tanks and supplies propellants to the combustion chamber, which releases energy through chemical reactions. The liquid rocket propulsion system has two types of propulsion systems, which are divided into a monopropellant system and a bipropellant system according to the difference in the configuration of fuel and oxidizer. The advantage of a liquid rocket propulsion system is that it can have the best performance of all conventional chemical propulsion systems and can provide precise thrust control. The disadvantages are the complexity of the system structure and the high development cost. In general, a monopropellant system is less complex than a bipropellant system, but has poor performance.

c. Hybrid Rocket Propulsion

The Hybrid rocket propulsion system (HRPS) is usually a combination of liquid/gas oxidizer and solid fuel. The liquid/gas oxidizer is fed into the combustion chamber using a feeding system similar to a conventional liquid rocket. Solid fuels are vaporized by the heat generated during combustion and mixed with oxidizer vapors for combustion. The hybrid system is simpler than the bipropellant system and performs better than the solid propellant system. And because fuel and oxidizers are stored separately, they are generally safer than other systems. The disadvantages are lower density than solid propulsion systems and lower performance than liquid propulsion systems.

3. Electric Rocket Propulsion

The Electric Rocket Propulsion System (ERPS) is a system that uses electricity to add energy to the propellant. To add energy, ERPS uses electrothermal heat or ionizes the propellant and accelerates the propellant through ions or plasma in an electrostatic or electromagnetic field. In the first method, the system is similar to LRPS, except for the heat source. The advantage of ERPS is its high specific impulse performance compare to the chemical systems. The disadvantage is that it requires heavy and inefficient power source, and the thrust is usually low, 0.005 to 1N. In addition, long-term (weekly or monthly) acceleration with low thrust levels is necessary to increase the speed of the spacecraft.

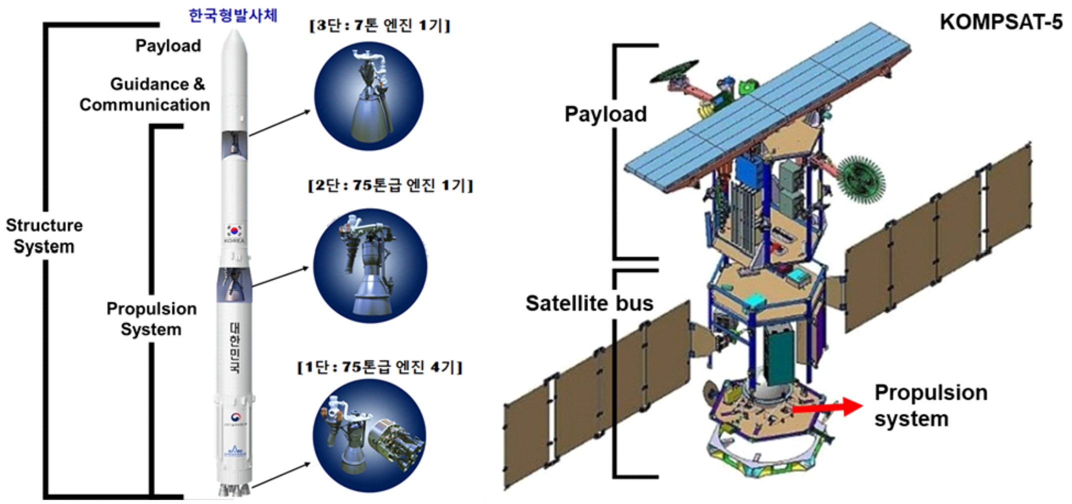


Fig. 4 Configurations of spacecraft/satellite systems

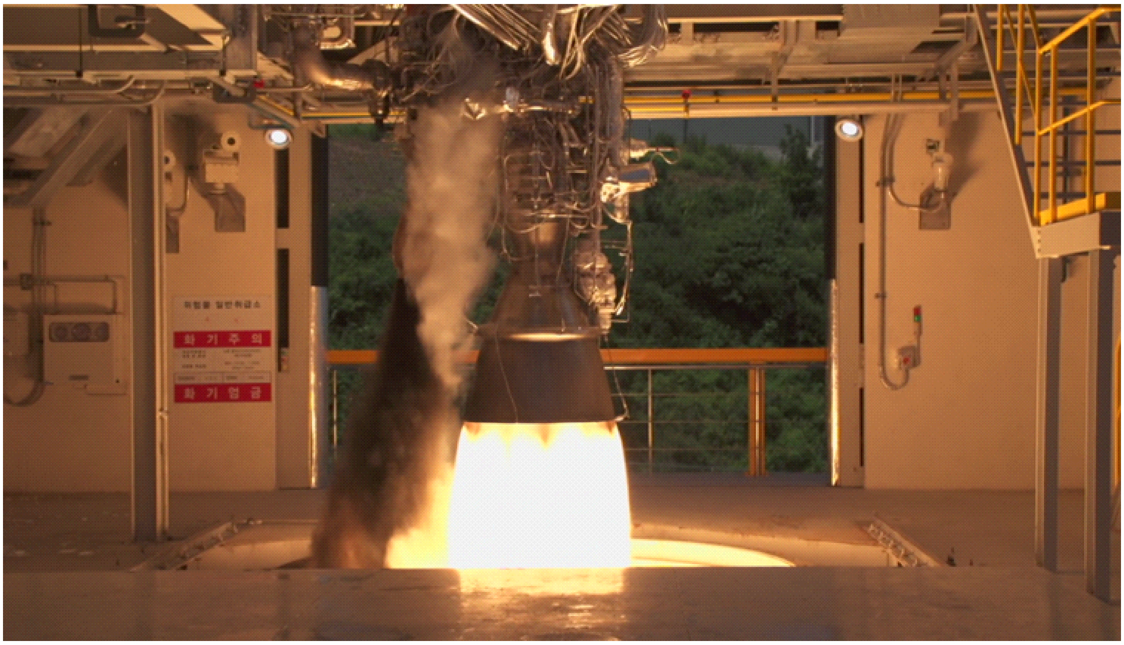


Fig. 5 KSLV-II Nuri 75 ton class liquid rocket engine[8]

Fig. 6 Apollo mission RCS Quad R-4D thruster[9]

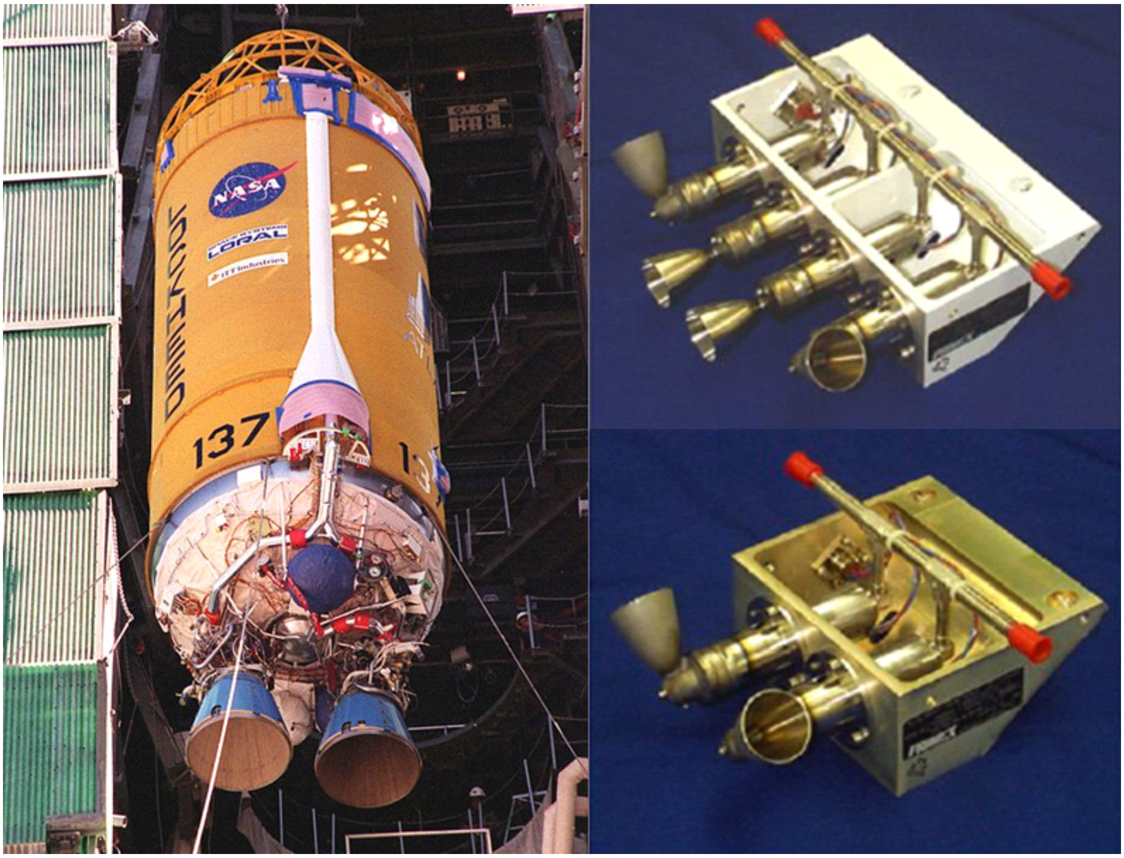
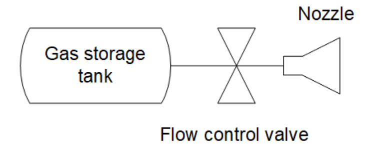
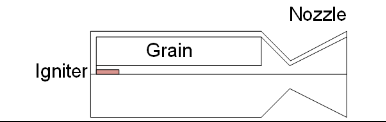
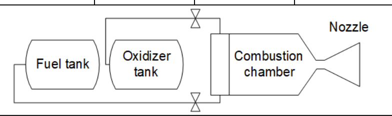
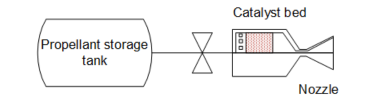
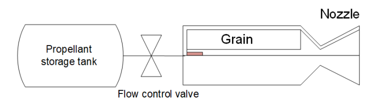
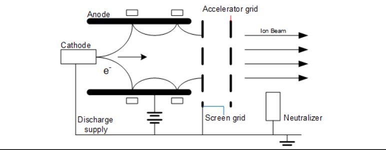


Fig. 7 Attitude control thruster in the Centaur stage rocket (Aerospace rocketdyne MRM-106D)[10]

Table. 2 Characteristics of space propulsion technique and its use

Sort	Schematic	Function: Launch	Function: Orbit insertion	Function: Orbit Maintenance and maneuver	Function: Attitude Control	Advantages	Disadvantages
Cold gas propulsion				○	○	• Simple structure • Reliability	• Low specific impulse • High-pressure propellant tanks • Propellant Storage tanks increase weight
Solid propellant propulsion		○	○			• Simple structure • High thrust level	• Not reusable • Thrust, not adjustable
Bipropellant propulsion		○	○	○	○	• High specific impulse	• Increased complexity and weight of the system structure
Monopropellant propulsion				○	○	• Simple structure than Bipropellant thruster • Relatively high specific impulse • Good reusability	• Impregnated catalyst technology needs • Lower than bipropellant thruster specific impulse characteristics
Hybrid propulsion		○	○	○		• Compared with liquid • Mechanically simple • Higher fuel density • Compared with solid • High theoretical ISP • Controllable thrust	• O/F ratio shift • Low regression rate
Electric propulsion			○	○		• Mass efficiency • Higher specific impulse • Longer operational lifetime	• Low thrust level • Poor power efficiency

B. Development of Low-toxicity Propellant

With the development of the rocket industry, the impact on the environment is also increasing proportionally and taking it seriously. Fig. 8 is showing a major exhaust produced by actual rocket launch situation. In particular, it has been confirmed through various research cases that exhaust gases from rockets during launch and re-entry can affect environmental pollution [20, 26, 27]. This influence comes entirely from the type of propellant used in the launch process. In addition, problems are continuously raised, which are caused by environmental pollution due to harmful substances generated during the production and handling of rocket propellants, and human hazards due to long-term exposure of toxic substances handlers.

Hydrazine or hydrazine-based propellants are widely used in thruster systems for attitude control of space launch vehicles and satellites. Fig. 9 represents the physical properties of hydrazine, and hydrazine is a material first synthesized by German chemist Julius Wihelm Theodor Curtius in 1887. It shows that it can be activated immediately by heating a propellant or by catalytic decomposition. These characteristics are used not only as a primary fuel for spacecraft, but also as an auxiliary power Unit (APU) for aircraft.

Recently, research to replace hydrazine is gradually increasing along with environmental regulations in the field of space propulsion. It is not easy to replace the performance and reliability of hydrazine propellants that have been in use for a long time, but some problems with hydrazine propellants (toxicity, carcinogenicity, and relatively low density) must be the motivation to carry out this actively.

Fig. 10 represents a processes from propellant manufacturing to handling. In this process, we are greatly aware of the problems caused by the toxic portion of hydrazine. This particular handling is directly related to the cost of storage and waste disposal of the toxic propellant. On the other hand, low toxicity can be replaced by the word "green" propellant, and this propellant is intended to improve overall performance while having low pollution impact on the environment with the motto of low toxicity[17, 19, 20, 26-44].

The operation of low-toxic propellants has the following advantages in the long run:

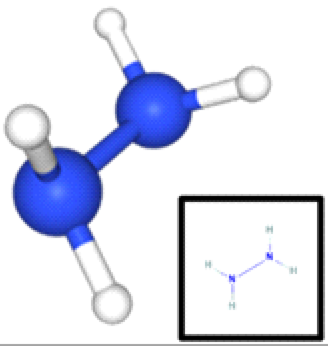
- The long-term storage capacity of propellants can significantly reduce the cost of logistics.
- Ground support equipment and personal safety are secured from a variety of problems that may occur in transport and charging situations of propellants, reducing ground operating costs and time.
- It is compatible with existing systems/interfaces, reducing additional R&D costs and time in manufacturing, assembly, integration and testing work.

Based on the low-toxic propellant-based technology, research is underway on applications from simple thrust control to space launch vehicles. Fuels such as 2-Dimethylaminoethylazide(DMAZ)[28-31], which have similar physical properties to Hydrazine, as well as Energetic oxidizers such as Ammonium dinitramide(ADN)[32-7, 58-66], Hydroxylammonium nitrate(HAN)[38-41, 43], and Hydrazinium nitroformate(HNF)[42-4], are being studied as candidates for hydrazine replacement. Toxicity evaluation of propellants based on candidate substances is given in Table 3 in which it is classified according to the Global Harmonized System (GHS) regulation [25]. Fig. 11 is classified according to the toxicity of these low-toxic propellants. As seen from the graph, while the toxicity and carcinogenicity of Hydrazine is very high, the toxicity and carcinogenicity of the low-toxic propellants currently being studied are significantly lower.

Various research institutions in the space propulsion field, such as the United States(NASA) and Europe(ESA), have conducted research demonstrations to replace hydrazine propellants. Fig. 12 is a summary of projects that have already performed or are currently performing performance verification on an actual environment. In the United States, HAN propellant-based Green Propellant Infusion Mission (GPIM) projects have been conducted to demonstrate propellants on the orbit, and in addition, they are focusing on research on the use of low-toxic propellants in various types [40, 41]. In Europe, ADN-based low-toxic propellants were developed and demonstrated in the space environment through the Prototype Research Instruments and Space Mission technology Advancement (PRISMA) mission [37]. Recently, Rheform is attempting to limit the use of hydrazine propellants [34, 35]. As seen from Fig 13, it aims to respond to a wide range of propulsion systems ranging from small thrusters to propellants for large projectiles [16].

Propellant	Major exhaust products
Liquid Oxygen(LO_X)/Liquid Hydrogen(LH_2)	H_2O, H_2
Liquid Oxygen(LO_X)/Hydrocarbon series	CO, CO_2 and other Hydrocarbons
Ammonium Perchlorate/Aluminum	HCl, H_2, H_2O, Al_2O_3, CO, CO_2, N_2
N_2O_4 or Dimethylhydrazine	N_2, NO_X, CO, CO_2, H_2O

Fig. 8 Major chemical species produced by the combustion of propellants applied to space launch vehicles



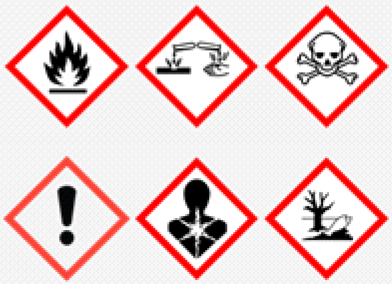
	HYDRAZINE
Chemical formula	N_2H_4
Molar mass	32.0452 g/mol
Density	1 g/cc
Boiling point	114
Melting point	2
GHS Hazard classification	

Fig. 9 Physical properties of Hydrazine

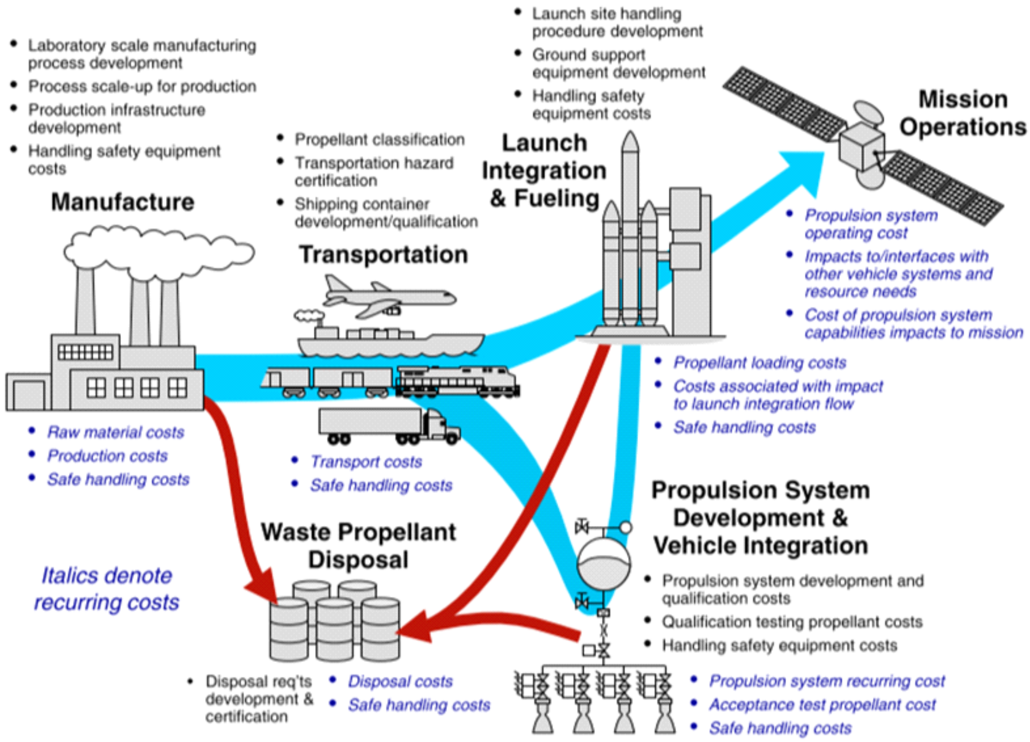


Fig. 10 Evaluation of life cycle cost from manufacture to mission operations of spacecraft propellants[27]

Table. 3 Classification of toxicity range values by experiments and estimation[25]

Exposure routes	Classification category or experimentally obtained acute toxicity range estimate	Converted acute toxicity point estimate
Oral (mg/kg body weight)	0 <Category 1 ≤ 5	0.5
	5 <Category 2 ≤ 50	5
	50 <Category 3 ≤ 300	100
	300 <Category 4 ≤ 2000	500
	2000 <Category 5 ≤ 5000	2500
Dermal (mg/kg bodyweight)	0 <Category 1 ≤ 5	5
	5 <Category 2 ≤ 200	50
	200 <Category 3 ≤ 1000	300
	1000 <Category 4 ≤ 2000	1100
	2000 <Category 5 ≤ 5000	2500
Gases (ppmV)	0 <Category 1 ≤ 100	10
	100 <Category 2 ≤ 500	100
	500 <Category 3 ≤ 2500	700
	2500 <Category 4 ≤ 20000	4500
Vapours (mg/l)	0 <Category 1 ≤ 0.5	0.05
	0.5 <Category 2 ≤ 2.0	0.5
	2.0 <Category 3 ≤ 10.0	3
	10.0 <Category 4 ≤ 20.0	11
Dust/mist (mg/l)	0 <Category 1 ≤ 0.05	0.005
	0.05 <Category 2 ≤ 0.5	0.05
	0.5 <Category 3 ≤ 1.0	0.5
	1.0 <Category 4 ≤ 5.0	1.5

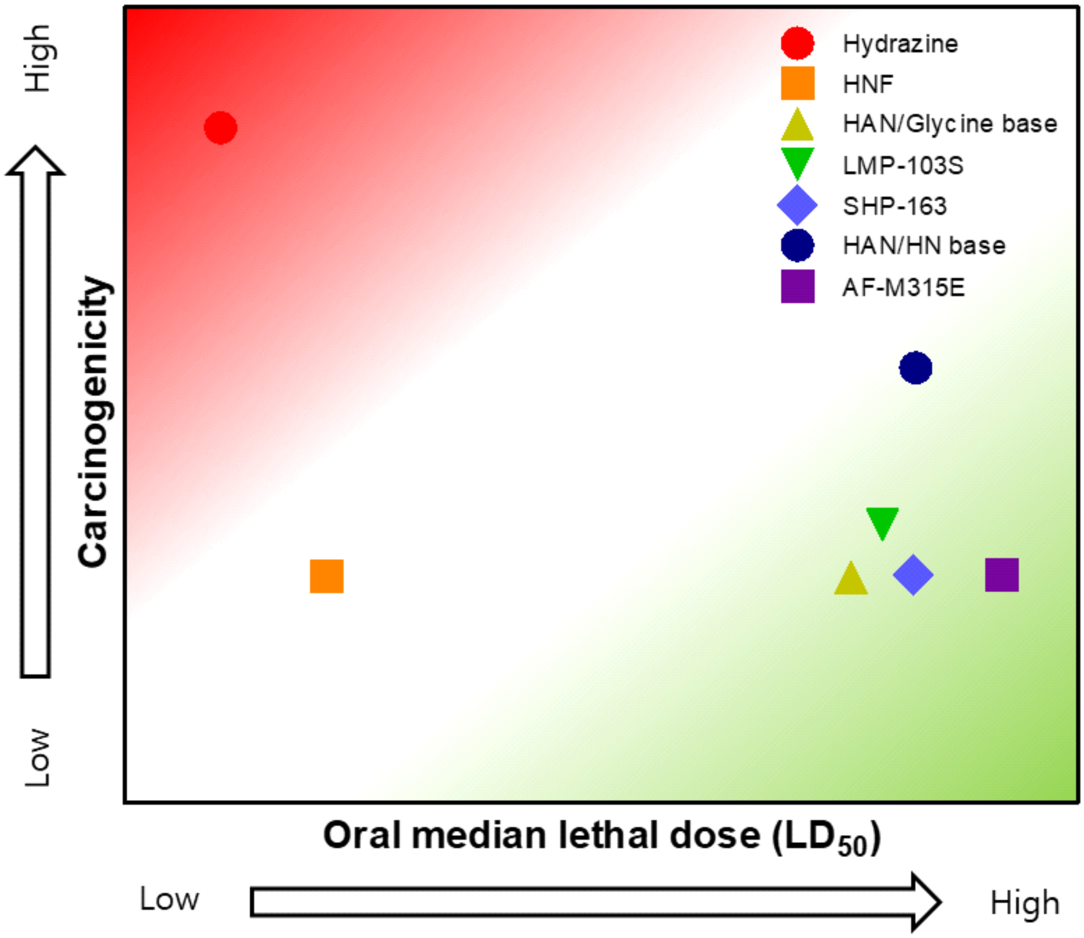
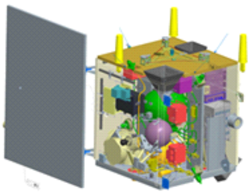


Fig. 11 Toxicity assessment of green propellants[39]



Prototype Research Instruments and Space Mission technology Advancement (PRISMA)	
Operator	Swedish Space Corporation (SSC)
Launch date	15 June 2010
Propellant	LMP-103S (ADN based Monopropellant)
Mission	• Flight demonstrations with new GN&C (Guidance, Navigation & Control) and sensors for future rendezvous and formation flying missions • Qualify the space propulsion system using ADN based propellant

Green Propellant Infusion Mission (GPIM)	
Operator	The National Aeronautics and Space Administration
Launch date	25 June 2019
Propellant	AF-315M (HAN based Monopropellant)
Mission	• Demonstrate the HAN(Hydroxylammonium Nitrate) blend propellant

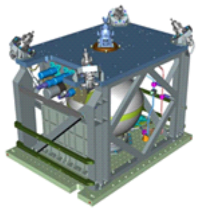


Fig. 12 Performance verification status of low-toxicity propellants

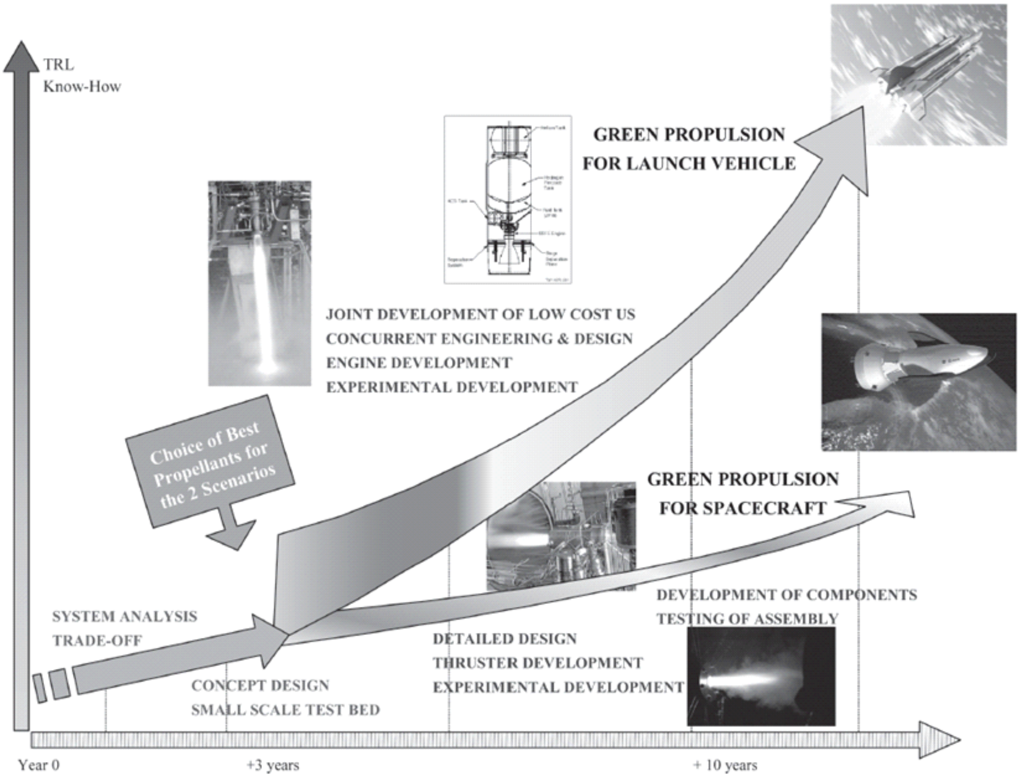


Fig. 13 Proposed roadmap for green propellants' development[16]

Table. 4 R&D status of low-toxicity propellant based on technical readiness level(TRL)

System test, launch, operation
System development
Technology demonstration
Technology development
Research to prove feasibility
Basic technology research

	TRL	국외			국내
		ECAPS (ADN)	NASA (HAN)	ETC	
성공적인 임무운용을 통한 실제 체계의 입증단계 (Actual system proven in operational environment)	9	○		△	
성능시한을 통한 실제 체계의 완성 및 입증 단계 (System complete and qualified)	8		□		
운용환경에서 체계 시제품의 성능시험 단계 (System prototype demonstration in operational environment)	7				▲
체계/부 체계 모델, 시제품 성능시한 단계(유사 운용환경) (Technology demonstrated in relevant environment)	6				
구성품 또는 조립체 성능 입증 단계(유사 운용환경) (Technology validated in relevant environment)	5				
구성품 또는 조립체 수준의 성능 입증 단계(실험실 환경) (Technology validated in lab)	4				■
주요 기능 분석/실험 또는 특성에 대한 개념 입증 단계 (Experimental proof of concept)	3				●
기술개념 형성 및 응용분야 식별 단계 (Technology concept formulated)	2				
기본 원리 이해 단계 (Basic principle observed)	1				

C. Suitable Catalyst in Propulsion System

A catalyst can be defined as a substance that accelerates change in a reaction. In the course of the reaction, it changes the speed of the reaction without consuming itself[12]. Fig. 14 represents a change in activation energy depending on the use of catalysis. Participation in the catalyst can affect the reaction rate. In general, reactions involving a catalyst occur faster than those that do not, because when the catalyst is used, the reactant reacts with the catalyst first, thereby reducing the activation energy of the reaction. This significantly lowers the ignition temperature required to trigger catalytic decomposition and at the same time increases the decomposition rate.

Currently, various types of catalysts are involved in the synthesis and production of chemicals. It is widely used throughout the chemical industry through properties such as increasing the speed of a chemical reaction or increasing the selectivity of a product by using a catalyst. It can be said that the utilization of the catalyst industry is almost as good as the overall chemical field, and its utilization is endless and the catalyst plays a very important role in the overall industry. The biggest contribution to human development through the use of catalysts is the synthesis of ammonia and fertilizer production through ammonia-synthetic iron catalysts, contributing to human food production and emphasizing the history of full-scale catalyst use. Since then, it has been used in crude oil refining and treatment in the petrochemical industry and has developed rapidly. The use of catalysts has not only been used for synthesis, but has also been used to counter environmental pollution such as removing contaminants. The scope of use of the catalyst is as follows.

Petroleum refining technology-This is a technology that refines low-quality crude oil into high-value added fuel through reforming such as cracking and reforming. In this technology, a facility using a catalyst is essential.

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Environmental and energy technologies-These are the most popular catalyst applications. In particular, studies are being actively conducted to prepare environmental impacts and measures against exhaust gas emissions from automobiles and factories. In addition, catalyst-related technologies have been studied in the field of fuel cells having a higher energy density compared to secondary batteries. Catalyst-related technologies are being studied from reforming fossil fuels to produce hydrogen, the main fuel for fuel cells, to electrodes for fuel cells.

In the aerospace sector, systems exist that actively utilize catalysts. In particular, thrusters used in space propulsion are installed for posture control. In the case of liquid propellant-based thrusters, thrusters in the form of a catalyst are often used. By using the catalyst, the energy consumed in decomposing the propellant can be reduced, which affects the life of the system.

The catalytic reaction is shown in Fig. 15 that there are homogeneous, heterogeneous and enzyme catalytic reactions. The homogeneous catalytic reaction refers to a reaction in which the reactant and the product are in the same state. At this time, all reactions of the gas phase belong, and the liquid phase is limited to a reaction that produces a solution, and there is no solid phase reaction. A non-homogeneous catalytic reaction refers to a reaction process in which a catalyst and a reactant exist in different phases (generally a solid catalyst and a liquid or gaseous reactant). Enzyme catalysis is a biological reaction in which an enzyme binds a substrate at an active site to promote a biochemical reaction.

The process of the catalytic reaction goes through a series of processes in which the product is produced by the reaction between the active site on the catalyst surface and the reactants. In order to be a good catalyst, three factors must be harmonized: fluid flow, reactivity and stability. These three factors affect the reactor design and the state of the catalyst bed according to the reaction. This element is usually related to the selection of active ingredients (catalyst precursors) and supports utilized in catalyst preparation. Most of the active ingredients are associated with catalytic reactivity. The type of catalyst and the supported amount have a great influence on the activation of the catalytic reaction.

The support provides a high specific surface area to induce the chemical reaction of the active ingredient. At this time, the surface area formed by the porous shape of the support affects fluid flow and pressure drop. In addition, the structural rigidity of the support also greatly affects the durability of the catalyst.

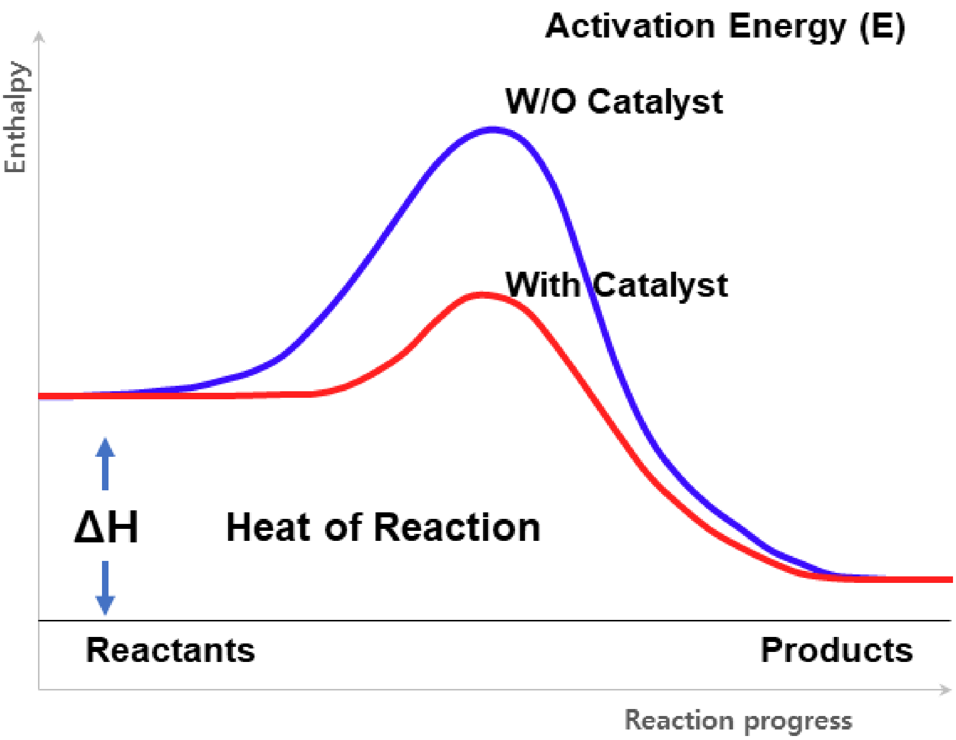


Fig. 14 Activation energy change by catalytic reaction

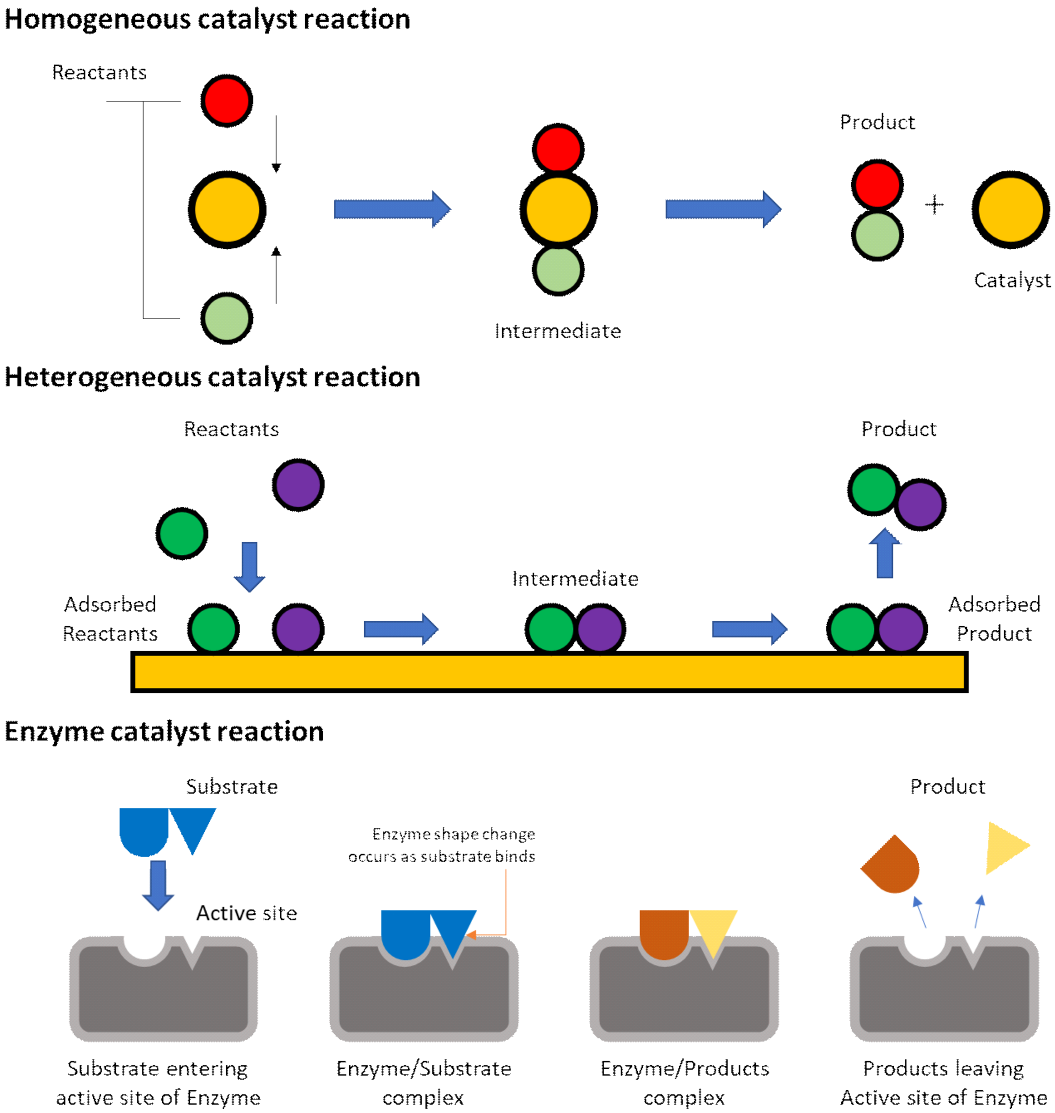


Fig. 15 Various catalytic reaction processes

D. Domestic Research of Low-toxicity Propellant based Propulsion System

There are several domestic research cases about the low-toxicity propellant based propulsion system[51, 52]. In 1997, Inha University conducted a study on the synthesis of ADN based on Bis(2-cyanoethyl)amine and Diethyl 4-azaheptanedioate[58, 59]. Since then, no major research and development issues have been reported. Recently, Kyunghee, Kongju, and Chosun University have conducted research on ADN-based propellants and technologies[60-7]. The study was initiated in accordance with the need to secure low-toxic propellant infrastructure technologies.

In the case of HAN, a performance test was conducted by synthesizing it as a propellant.[55-7]. The experiment was conducted with propellants based on HAN and methanol, but the results were not good. According to the results mentioned, it was confirmed that the decomposition efficiency of the propellant was lowered due to the low catalytic decomposition activation degree, and the thruster temperature was rapidly decreased during the experiment. For this reason, the performance of the propulsion system was not uniform. As a result, it was confirmed that higher preheating temperature is needed compared to hydrazine for smooth HAN propellant decomposition, and methods such as change of pressurization condition and increase of temperature inside catalyst bed were suggested as solutions to cope with instability. And the high decomposition temperature of the HAN propellant accelerates catalyst sintering and volatilization problems in catalyst-based systems.

Finally, there is a hydrogen peroxide-based propulsion system with the largest number of studies conducted in Korea[51-3]. High test peroxide(HTP) based monopropellant thruster and hydrogen peroxide/kerosine bipropellant thruster have been studied. In the case of the 50N thruster, performance evaluation was conducted in a high-altitude vacuum environment for mounting a Korea Space launch Vehicle(KSLV-II).

Nevertheless, there is a problem that the hydrogen peroxide-based propulsion system must solve. Due to the high risk of explosion due to the high concentration of hydrogen peroxide instability, care must be taken for handling. And although it has improved compared to the past, the long-term storage problem based on the nature of hydrogen peroxide is still a factor that limits the scope of application of hydrogen peroxide-based propulsion system. Finally, low thrust performance (−50s versus Hydrazine) is still a problem to be solved. These problems are not suitable for systems that require long-term storage.

As can be seen in the foregoing domestic case, the study of low toxicity propellants and catalysts has mostly been conducted individually for each element of catalysts and propellants. As a result, normal performance evaluation is not carried out due to reactivity problem between propellant and catalyst.

In this study, the research was conducted with the aim of developing integrated test evaluation criteria for low toxicity propellant-catalysts. Through this, the selected propellant-catalyst combination was applied to the actual thrusters to perform the combustion and thrust performance evaluation. It can easily respond to changes in the various propellant-catalyst combinations by providing simpler and easier application methods, especially in the process of verifying and screening complex and demanding decomposition characteristics. And one of the decomposition characteristics of low toxic propellants, the flameless phenomenon, confirmed the possibility of performance prediction through the decomposition flame observation of low toxic propellant thrusters.

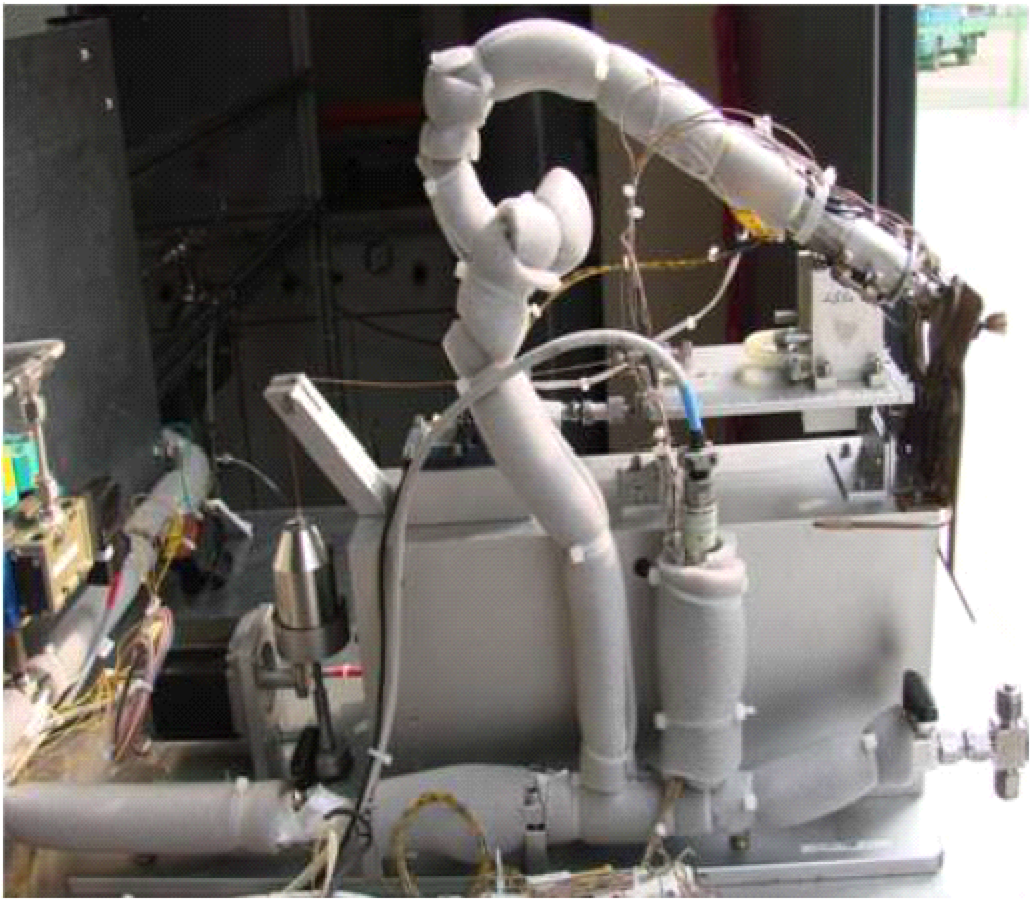
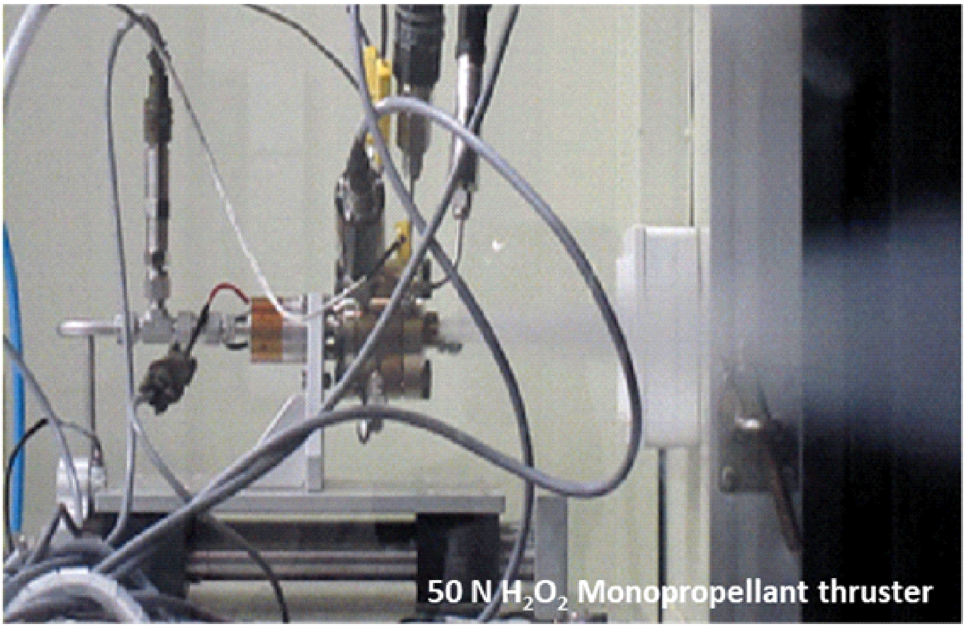


Fig. 16 HAN based propellant performance evaluation



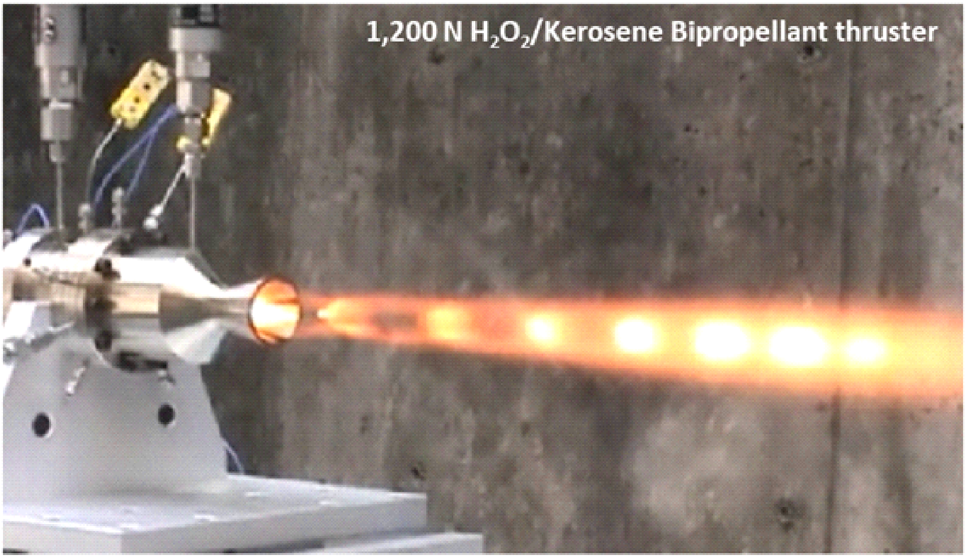


Fig. 17 High-Test Peroxide(HTP) base propulsion system test

II. Experiments

A. Propellant Synthesis

In this paper, the ADN-based low toxicity propellant was synthesized for the experiment. The dinitramide salt was reported in the United States in 1988, but the Russians had already used it in various missile programs in 1971. In particular, since dinitramide does not contain carbon or chlorine, combustion products are not harmful to the atmosphere. In addition, it has a 30% higher density and a 6% higher specific thrust value than hydrazine, and especially lethal dose(LD_{50}) is about 12 times higher than hydrazine, so it is getting the spotlight as a low-toxic propellant due to its advantages such as easy handling as a very safe material.

There are four general methods of synthesizing dinitramide salts[19, 33]. As shown in Eq. 1, The first synthesis method is to form a dinitramide anion by cutting off the R-N bond in the N and N-dinitro derivatives $R\text{-}N(NO_2)_2$.

$$R-N(NO_2)_2 \longrightarrow -N(NO_2)_2^- + R^+ \qquad \text{Eq. 1}$$

The second method of synthesis is based on an inorganic synthetic route such as Eq. 2 and is a synthetic route that directly causes nitration reaction of NH_3 or NH_2NO_2 through a powerful nitrating agent such as NO_2BF_4 or N_2O_5.

$$NH_3 \rightarrow NH_2NO_2 \longrightarrow HN(NO_2)_2 \xrightarrow{NH_3} NH_4N(NO_2)_2 \qquad \text{Eq. 2}$$

Same as Eq.2, the third method is a method of proceeding the nitrification reaction of an inactivated amine such as NH_2CONH_2 or NH_2COOCH_3 using a strong nitrating agent such as NO_2BF_4 or N_2O_5.

$$H_2N - X \xrightarrow{N_2O_5 \text{ or } NO_2BF_4} HN(NO_2)_2 \qquad \text{Eq. 3}$$
$$X: -CONH_2,\ -COOR$$

Finally, the fourth method like Eq. 4 is that it is synthesized by a double decomposition reaction between the dinitramide salts already synthesized and each metal salt.

$$MH(NO_2)_2 \xrightarrow[-MX]{M^{'}X} M^{'}N(NO_2)_2 \qquad \text{Eq. 4}$$
$$X: -OH,\ -Cl,\ -SO_4$$

In this paper, we synthesized ADN and ADN-based liquid monopropellant based on the following synthesis method.

1. Ammonium Dinitramide(ADN) Synthesis

It is diagram of ADN synthesis process such as Fig. 16. First, a strong acidic dinitramide aqueous solution is prepared by nitrating reaction of potassium sulfamate (KSO_3NH_2), high concentration nitric acid (HNO_3), and sulfuric acid (H_2SO_4). After neutralization with potassium hydroxide (KOH), solid ammonium dinitramide was extracted through an ion exchange reaction with ammonium sulfate [$(NH_4)_2SO_4$].

Subsequently, the ammonium dinitramide extracted as a solid phase was repeatedly separated using a carbon-based activated carbon adsorbent to recover high-purity ammonium dinitramide.

2. Synthesis of Liquid Monopropellant

Liquid phase monopropellant was prepared by mixing purified high-purity ammonium dinitramide (99%) as the main component with a solvent (H_2O), fuel (CH_3OH), and organic additives.

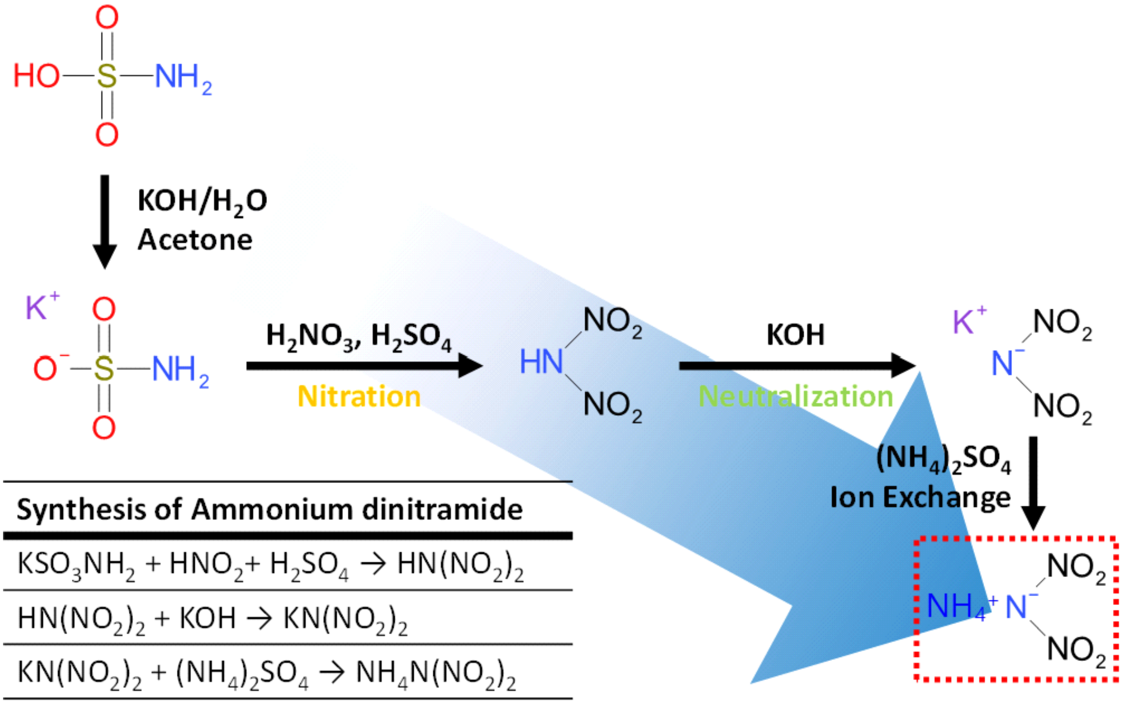


Fig. 18 Synthesis of ammonium dinitramide using Potassium Sulfamate

B. Catalyst Synthesis

In general, dispersion of a high specific surface area oxide-based support is one of four methods of coprecipitation, impregnation, adsorption, and ion exchange to prepare a catalyst[12]. As described, there are various synthesis methods, but the catalyst is prepared by impregnation method in this paper. The impregnation method is frequently used as a catalyst manufacturing method, and as one of the simple ways to increase the dispersion of active points on the support, the synthesis method is relatively simple compared to other synthesis methods. Fig. 17 shows the process of catalyst preparation through impregnation method. The impregnation method proceeds in the following order. In the process of preparing the impregnation method, it begins with the selection of catalyst support that has both porous structure and physical characteristics. Active substances are injected into a solution containing precursors. Effective interaction produces ion-exchange reactions between precursor ions in the solution and ions on the surface of the support during the impregnating process.

Metal catalyst precursor was used for ADN decomposition. This was impregnated with supports such as α-Al_2O_3 and γ-Al_2O_3. Generally, the specific surface area of γ-Al_2O_3 is high, but in the case of thermal conductivity, α-Al_2O_3 is high. The catalyst synthesis sequence is shown in Fig. 17.

1. 4 h calcining the alumina bead at 1200 ℃.
2. Prepare a metal precursor solution.
3. Alumina bead and precursor solution are put in a round flask, and a catalyst is prepared using a rotary evaporator.
4. The prepared catalyst is dried in an oven at 100° C. for 24 h, and then calcined at 500° C. for 3 h.

The synthesized catalysts were evaluated for performance through additional experiments.

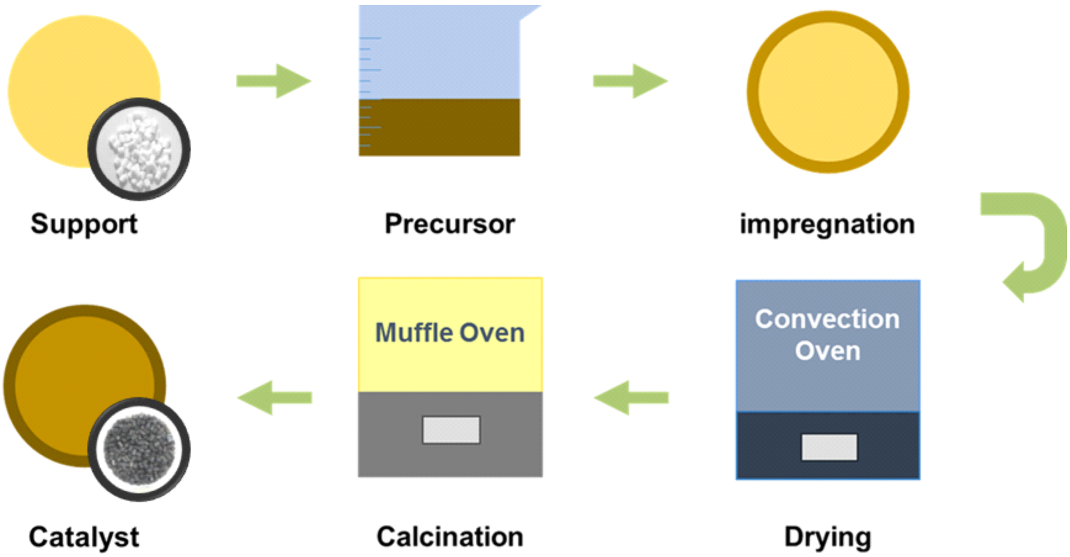


Fig. 19 Catalyst manufacturing process by impregnation method

C. Catalyst Screening through Isoconversional Method

The thruster used in this study is a catalyst decomposition based thruster. As the reactivity between the catalyst and the propellant greatly affects the performance of the thruster, it is necessary to analyze the decomposition properties between them. However, it is difficult to provide for judging the decomposition performance between the propellant and a catalyst through a simple experiment. Therefore, the evaluation was conducted through chemistry kinetics as a basis for judging and screening the performance of several catalyst candidate groups. The chemical reaction kinetics can be used as a basis for determining the reactivity with the catalyst through a correlation with the reaction rate with a change in the concentration of the reactants. In order to analyze the reactivity between the propellant and the catalyst, the decomposition mechanism for the catalytic reaction was grasped and the activation energy (E_a) and frequency factor (A) for each mechanism were compared to finally select the catalyst.

$$\ln K = \ln A - \frac{E_a}{RT} \qquad \text{Eq. 5}$$

In general, the Arrhenius equation is like Eq 5. Decomposition performance can be predicted through the equation, and the rate constant(k) is obtained experimentally. In addition, the mechanism in the intermediate process must be analyzed to finally determine the rate of the overall reaction. At this time, as the composition of the propellant is varied, the decomposition mechanism formed also varied, and the process of grasping the entire mechanism is very complicated and difficult. Since the low-toxicity monopropellant used in this study is also a pre-mixed monopropellant, it contains various species in the propellant, which has various mechanisms and requires great effort to compare and analyze its properties through the Arrhenius equation.

$$\ln \frac{\beta}{T_m{}^2} = \ln \frac{AR}{E_a} - \frac{E_a}{RT_m} \qquad \text{Eq. 6}$$

In order to analyze the decomposition performance of various catalysts in a low-toxic propellant, the catalytic screening was performed based on the isoconversional method to more easily and simply evaluate the suitability through activation energy and reaction factors[47-50]. The basic assumption of the isoconversional method is that in any given reaction range, the same reaction occurs at the same rate regardless of temperature. Unlike the Arrhenius equation, it is easy to predict decomposition performance without analyzing individual mechanisms. Fig. 18 is the result of the analysis obtained through equipment such as DSC/TGA. Based on these results, the heating rate (β) and peak temperature (T_m) are obtained. Activation energy and rate constant can be easily derived from the obtained values.

In this study, TGA analysis of low-toxicity propellant and catalyst was conducted to derive the results, and the reaction characteristics of each catalyst according to the decomposition of the propellant were analyzed through the isoconversional method. The decomposition performance of the low-toxicity propellant and catalyst was evaluated through the respective analysis results.

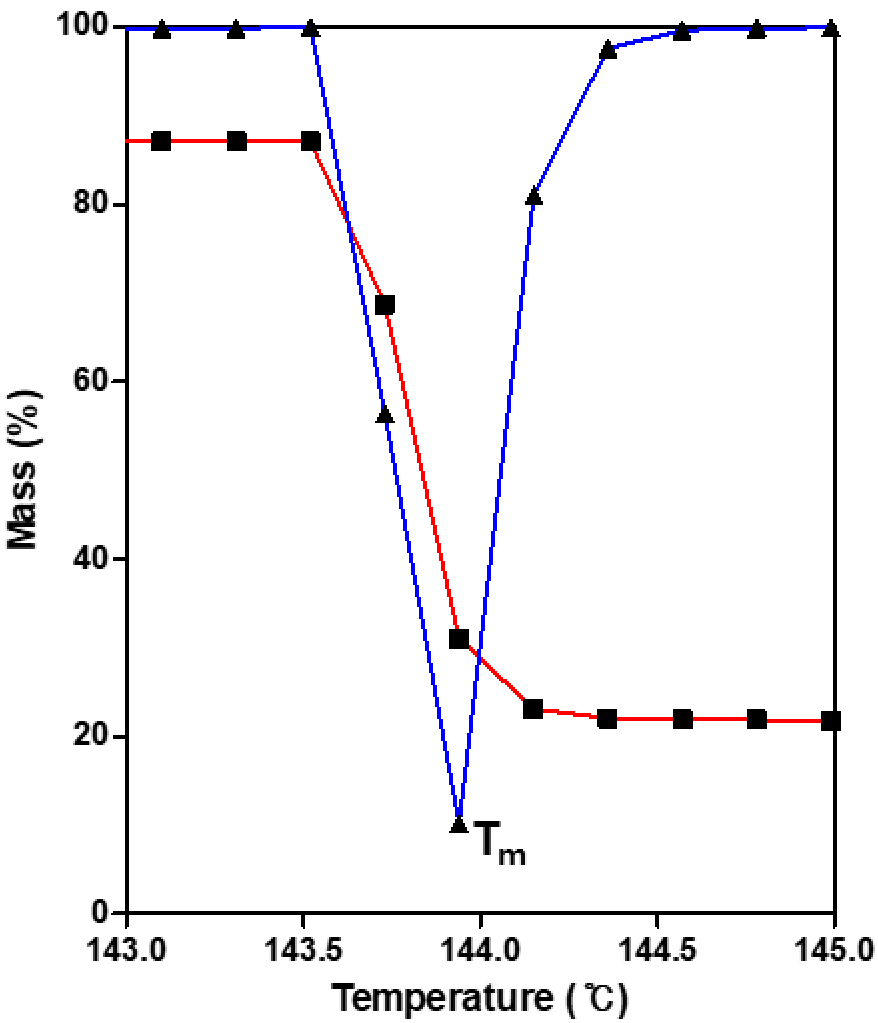


Fig. 20 Analysis of decomposition reaction through TGA of low-toxicity propellant and catalyst at specific heating rate

D. Thruster & Thrust Measurement System(TMS) Design

Prior to the design of the thruster, the specific impulse(I_{sp}) and the characteristic velocity (C^*) for the composition of the propellant were calculated through NASA Chemical Equilibrium with Applications (CEA)[13, 14]. In general, ADN-based low toxicity propellants are produced by premixing oxidizer ADN with other additives such as solvents and fuels and stabilizers. The composition of the synthesized propellant was calculated by setting ADN, H_2O, and CH_3OH to 63.4:25.4:11.2, respectively. And considering the actual operating range, the calculation result by setting the nozzle expansion ratio (ε=50) and supply pressure (20bar) is shown in Table. 5. Based on the results, the propellant supply amount of the thruster and the design of the thruster nozzle were performed. According to Eq.7 and Eq.8, the flow rate of propellant supplement and nozzle outlet diameter at the target thrust were obtained and the nozzle design was also performed. The mass flow rate of propellant supplement was calculated to be about 0.4g/s.

$$F = I_{sp} \cdot \dot{m} \cdot g_0 \qquad \text{Eq. 7}$$

$$\dot{m} = P_c \cdot \frac{A_t}{C^*} \qquad \text{Eq. 8}$$

Fig. 19 is the cross section of the thruster. The design consists of an injector, a catalyst bed, and a nozzle. First, the injector supplying the propellant has a shower head type, a collision type, and a swirl type. The injectors for the experiment adopted showerhead type injectors. The hole machining with injection angle of 30˚, diameter of 0.1 mm was carried out through super drill machining. Fig. 20 is the shape of an injector machined with a super drill. And Fig. 21 is the propellant injection test of the manufactured injector. The test confirmed that the propellant was atomized and injected. And a general converging-diverging nozzle was designed and manufactured. A nozzle shape having a nozzle neck diameter of 0.6 mm was produced at an expansion ratio ε=50.

Fig. 22 is a schematic diagram of thrust measuring system for collecting performance and status information of a thruster. For the propellant pressurization, an inert gas N_2 was used. At this time, N_2 is used not only for pressurizing the propellant, but also for purifying the working fluid of the pneumatic valve and the end of the experiment. And a real-time monitoring and control system was built. Before and after the supply of propellant and inside the catalyst bed where the catalytic decomposition reaction appears, each temperature and pressure sensor is installed, and data is collected through DAQ(Data Acquisition Device) for real-time monitoring. manual/automatic propellant supply control for the experiment was programmed. and algorithm for an emergency shut down event also programmed. In Fig. 23 and Fig. 24, the control module is programmed.

The thrust measurement system was designed as shown in Fig. 25. The system was manufactured in the form of a modular. In particular, by placing the thruster module on the spindle lift table, it was designed to prevent damage to the equipment by quickly removing it from the thrust measuring plate after stopping the experiment, as well as preventing damage to the load cell due to sudden impact. In addition, it is possible to increase the ease of modification and maintenance of the device through modularization.

There are various thrust measurement methods, but the target thrust is 1N class, so a pendulum-type thrust measurement system was designed for precise thrust measurement of thrusters. At this time, measurement errors may occur due to the physical properties of the thin plate connected to the stand. In order to correct the measurement error, an error was checked through a weight balance and a work was performed to correct it. Fig. 26 is a measurement result using a weight in a thrust measurement system and a measurement error correction operation was performed based on the result.

Table. 5 CEA code calculation result with ADN monopropellant

<table>
<tr><td colspan="2">Supply Pressure</td><td colspan="2">Nozzle Expansion Ratio</td></tr>
<tr><td colspan="2">20 bar</td><td colspan="2">50</td></tr>
<tr><td colspan="2">Reactants</td><td>Weight Ratio</td><td>Temperature</td></tr>
<tr><td colspan="2">CH_3OH</td><td>11.5</td><td rowspan="3">298.15 K</td></tr>
<tr><td colspan="2">H_2O</td><td>23.9</td></tr>
<tr><td colspan="2">ADN</td><td>64.6</td></tr>
<tr><td>I_{VAC}</td><td colspan="2">2496.4 Ns/kg</td><td>254.7s</td></tr>
<tr><td>I_{SP}</td><td colspan="2">2415 Ns/kg</td><td>246.4s</td></tr>
<tr><td>C^*</td><td colspan="3">1344.2 m/s</td></tr>
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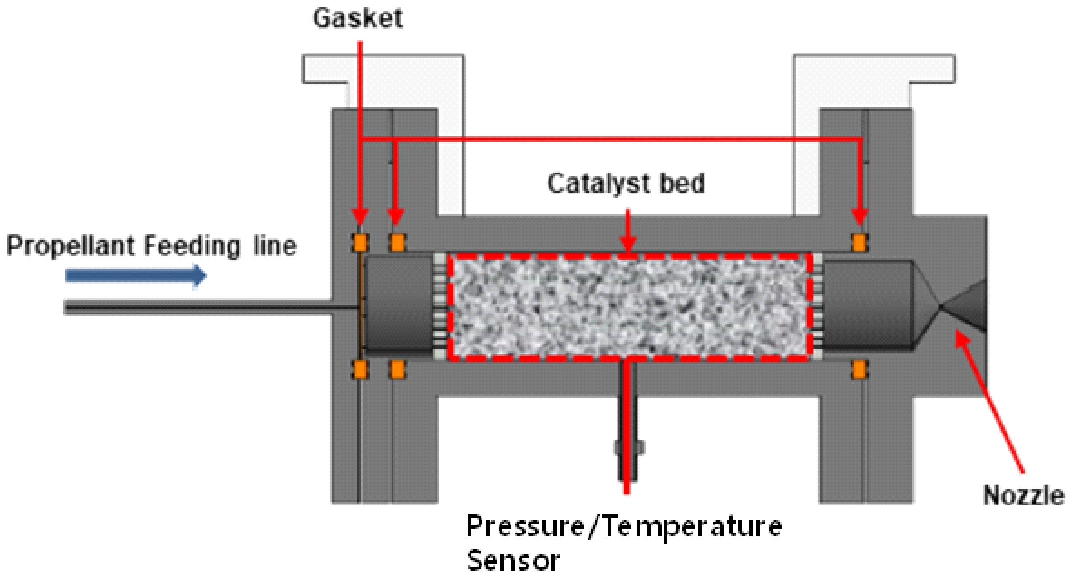


Fig. 21 Sectional view of 1 N class thruster for low-toxicity propellant/catalyst evaluation

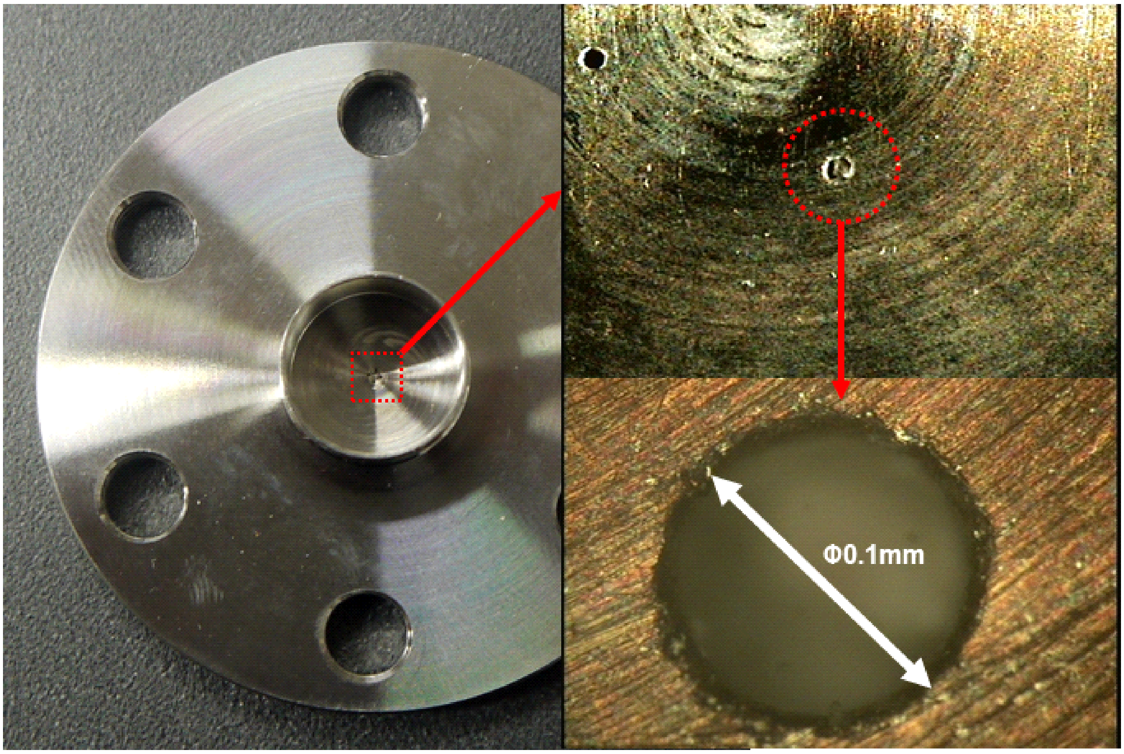


Fig. 22 Super drill machined showerhead type injector (Dia 0.1mm * 4 holes)

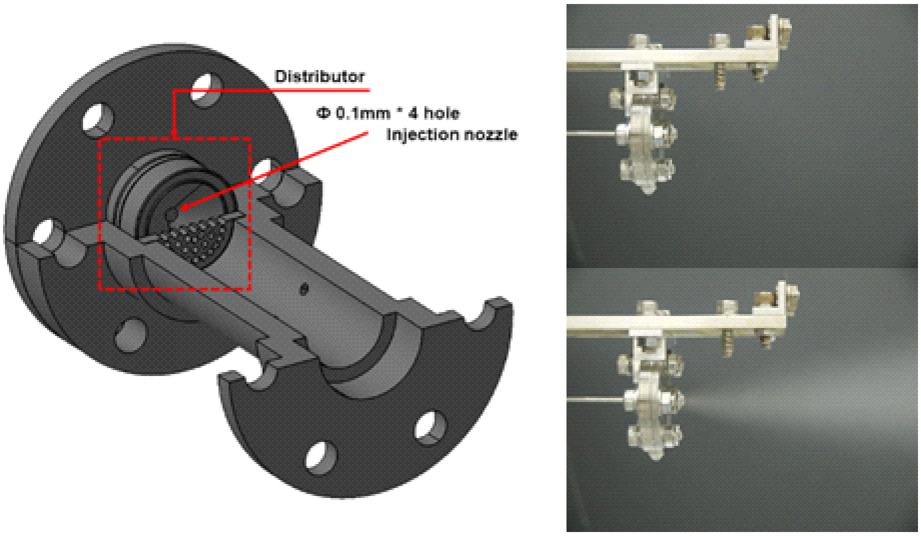


Fig. 23 Propellant injection test of showerhead type injector

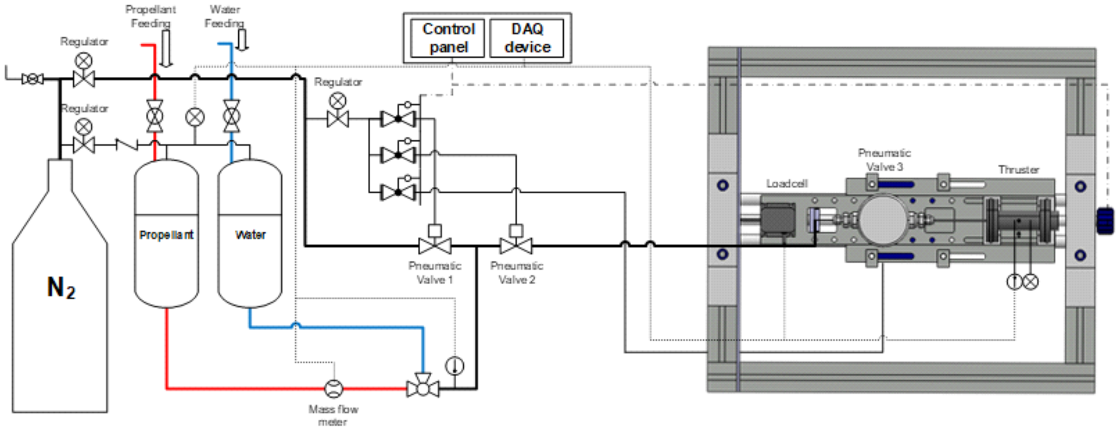


Fig. 24 Schematic of thrust measurement equipment for collecting thruster performance and condition

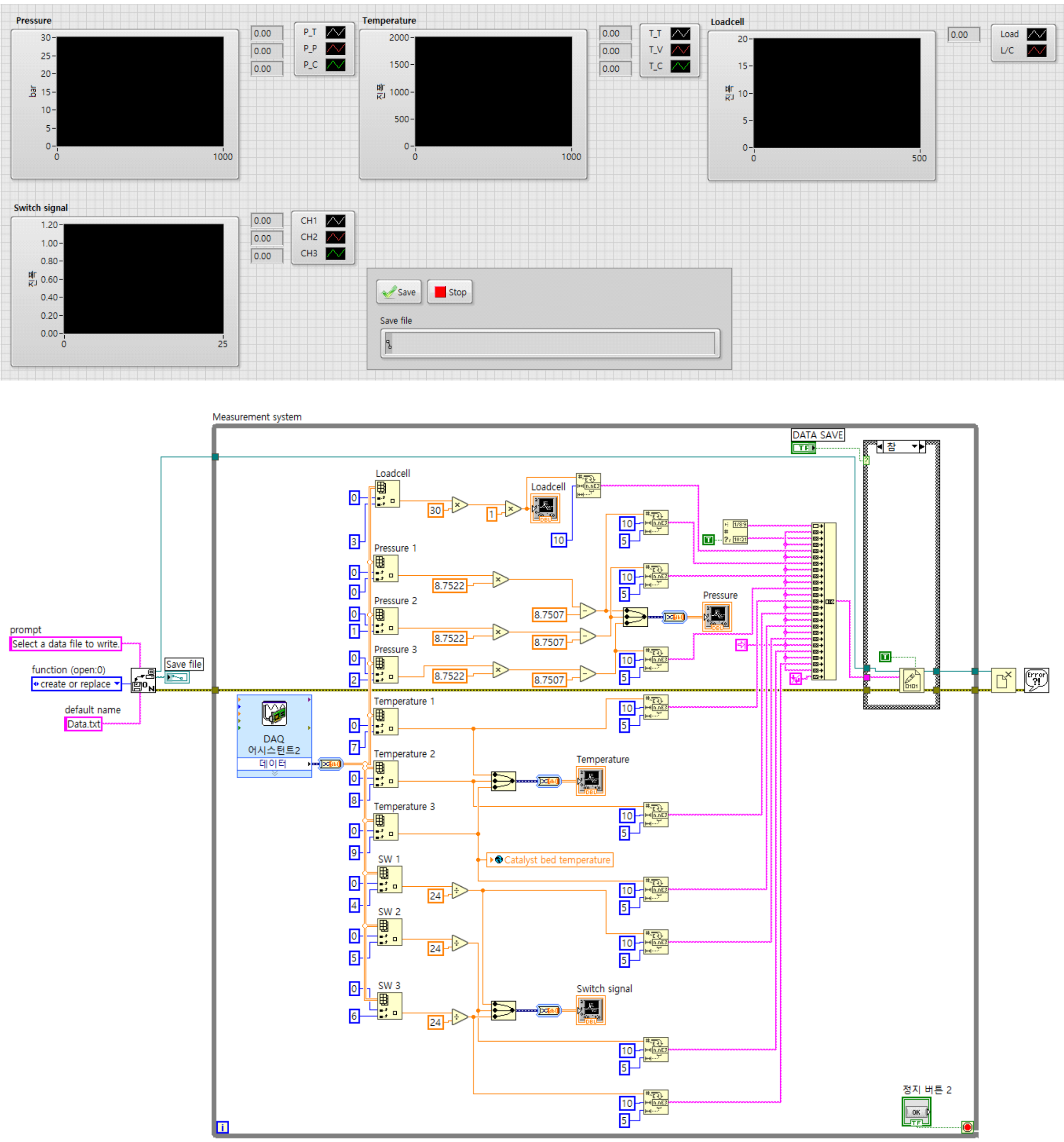


Fig. 25 Thruster condition monitoring system

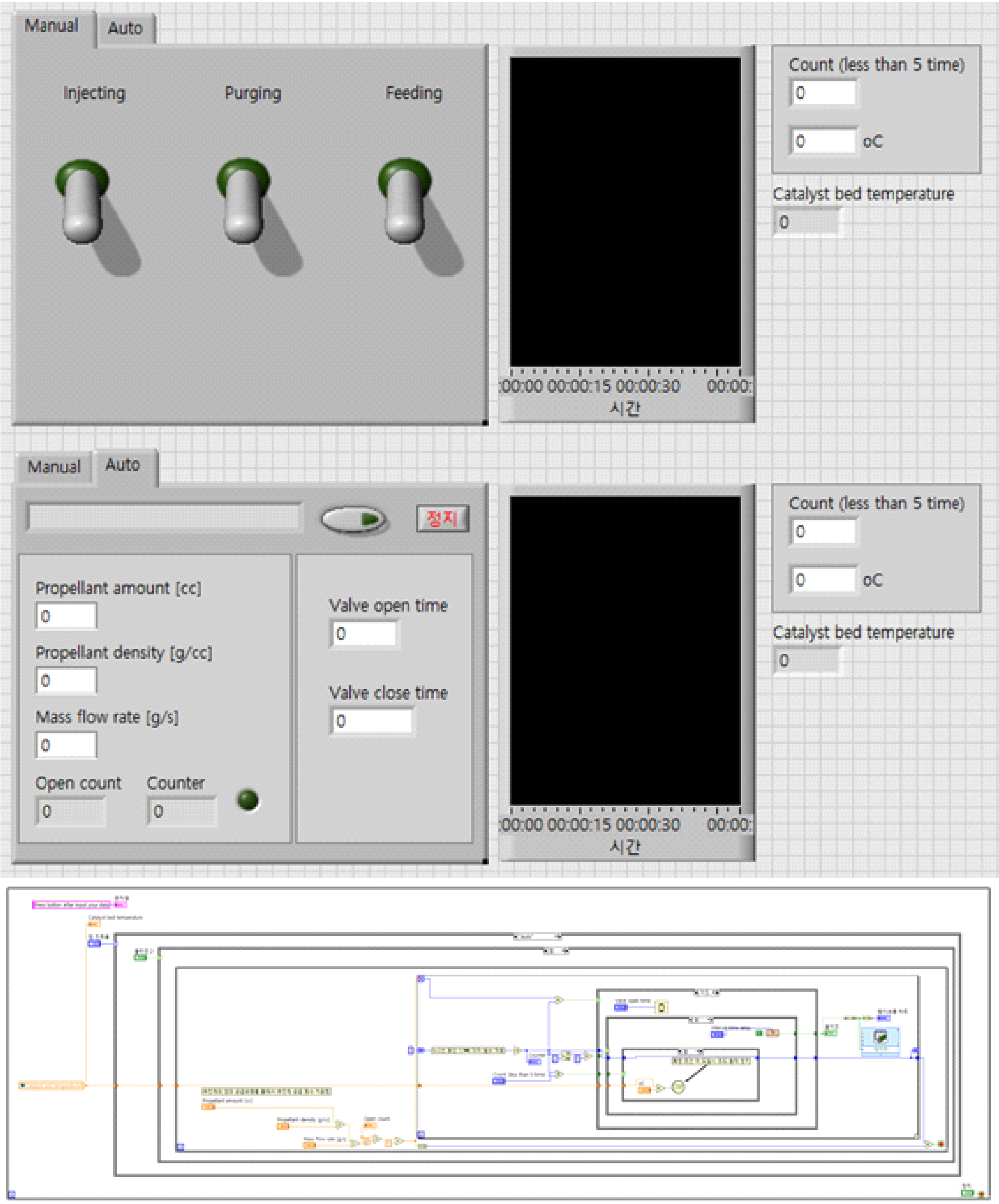
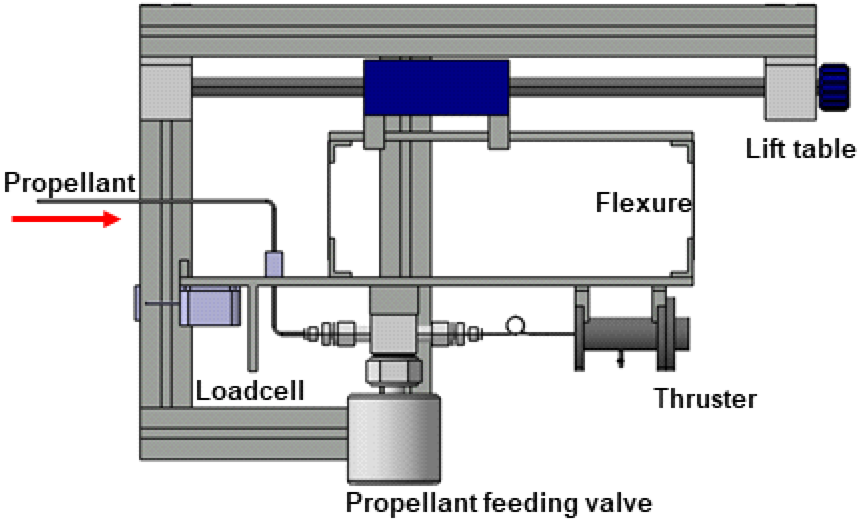


Fig. 26 Thrust experiment control system [A] manual mode, [B] auto operation mode



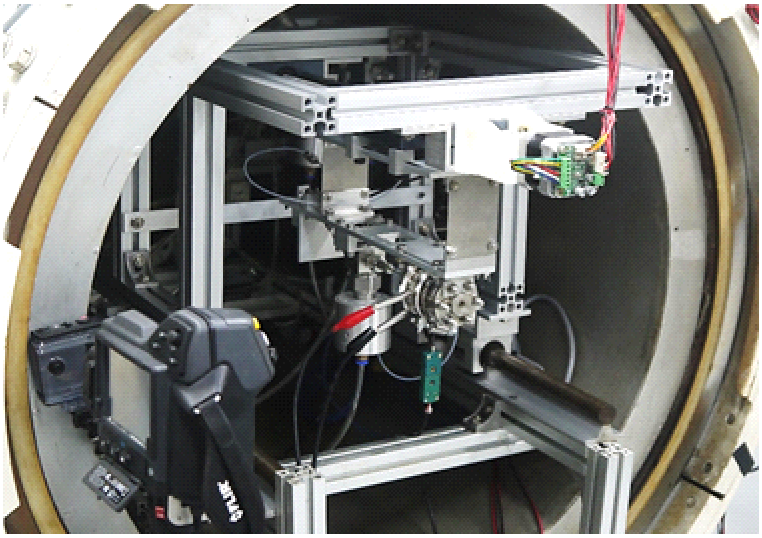
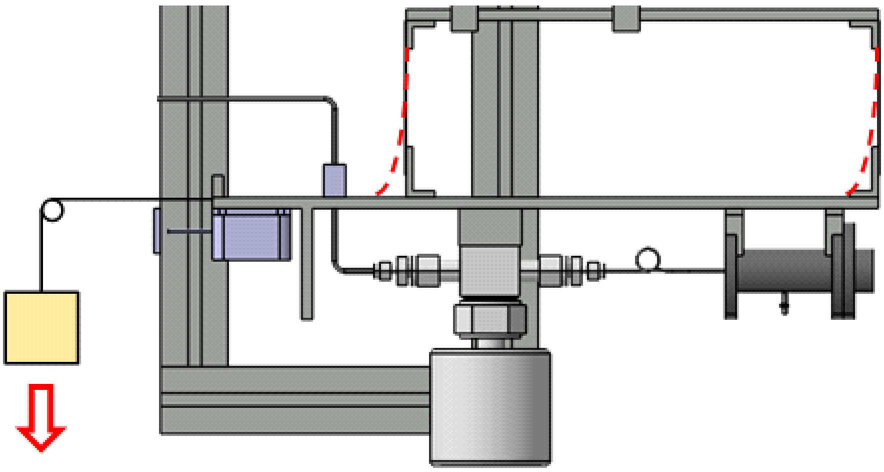


Fig. 27 Thrust measurement system produced through modular design



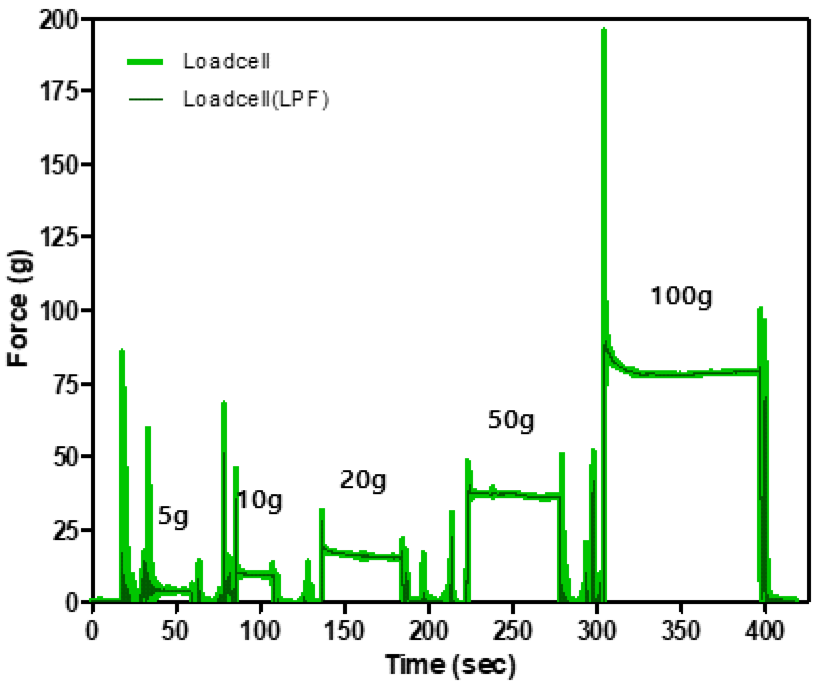


Fig. 28 Measurement error correction for pendulum-type thrust measurement system

E. Visualization of ADN Propellant Exhaust Plume through Mid-range Infrared Observation

In addition to thrust measurement system, a exhaust flame visualization equipment was provided. ADN is very difficult to visually determine the flame, so separate equipment is required for the observation and visualization of the propellant decomposition gas [36]. This is similar to HAN-based propellants as well as ADN. For the HAN-based propellant, the visualization of the flame through Schlieren was proceeded as shown in Fig. 27.[40]

The ADN propellant used in the experiment is in an pre-mixed state. Therefore, it was confirmed that different colors of smoke are formed depending on the conditions in abnormal decomposition as shown in Fig. 28. To observe this, the experimental apparatus was constructed like Fig. 29. Particularly, the flame observation was attempted through a Mid-range Infrared (IR) camera.

For the observation of the flame of the low-toxicity propellant by adding an observation equipment, while the possibility of visualization of the propellant-decomposed flame was confirmed through a mid-range IR camera, performance prediction was performed through observation results. From the observation results, it is intended to verify whether the propellant was decomposed such as Fig. 30. In addition, it was judged that the degree of decomposition of the propellant and the performance of the propellant could be predicted based on this, as the flame could observe the decomposition of the propellant. In this experiment, the catalytic decomposition and performance prediction of the propellant were conducted through the decomposition gas observed by visualization.

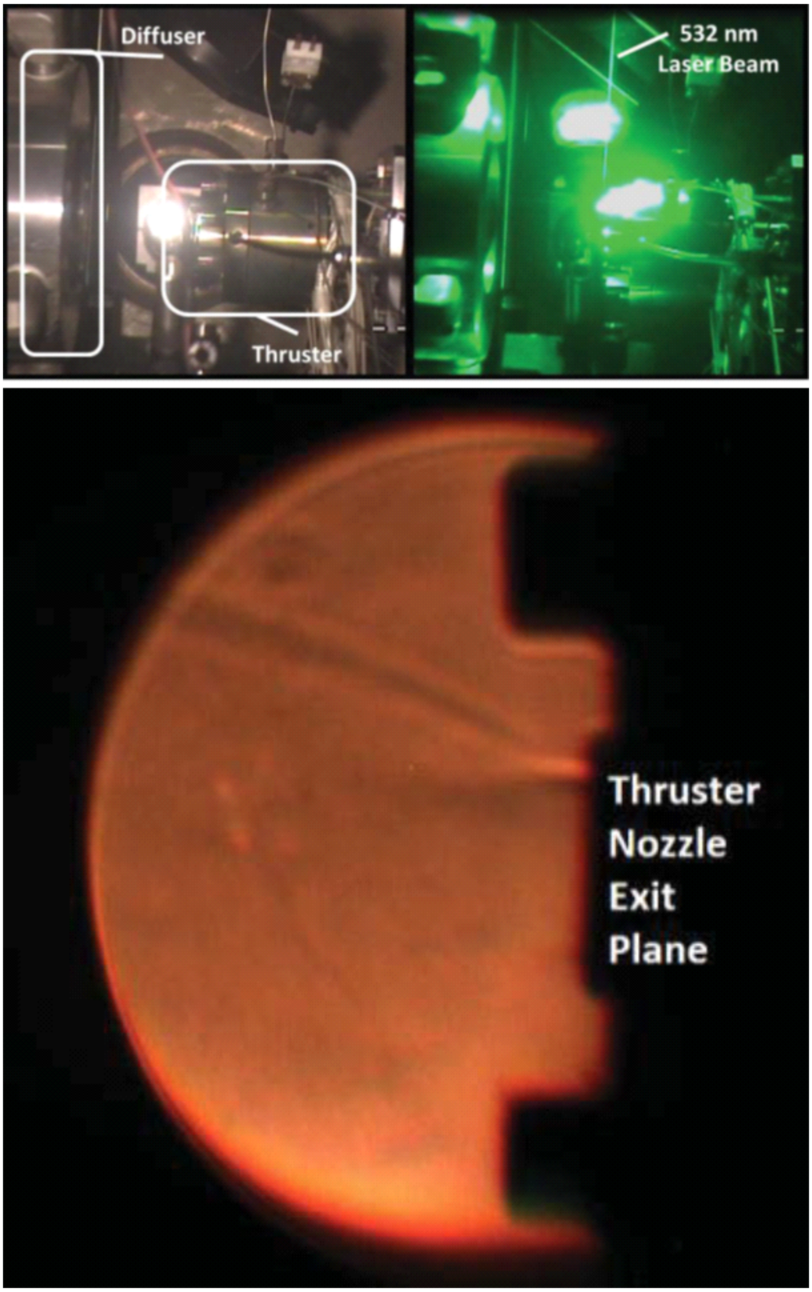


Fig. 29 Plume observation of HAN-based AF-315E propellant through Schlieren[40]

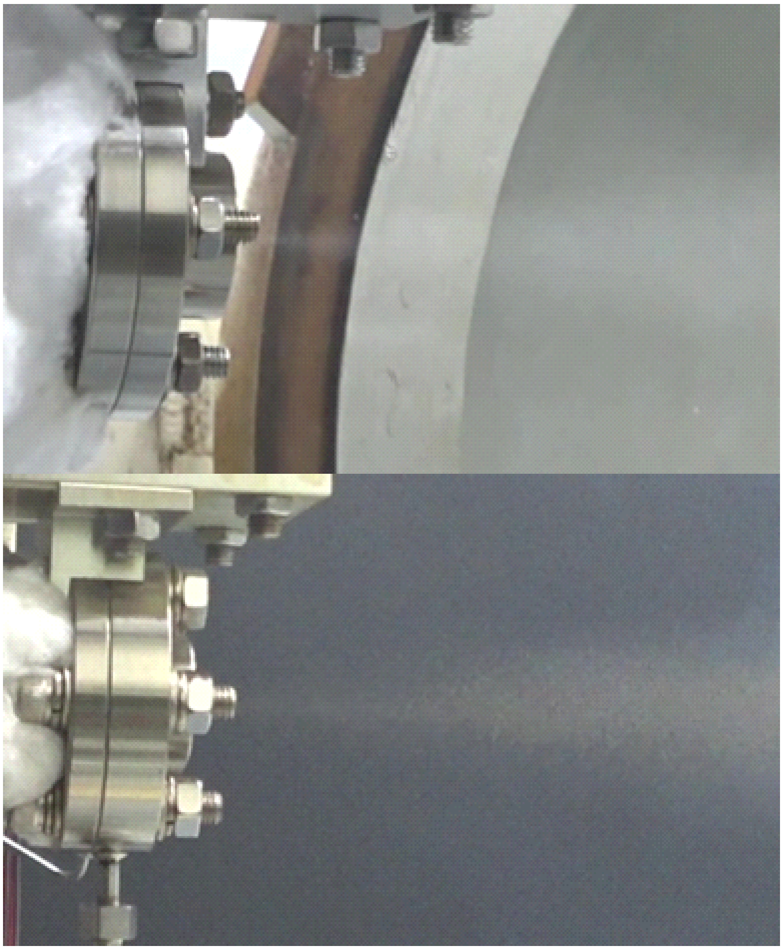


Fig. 30 Observation of smoke at the rear end of thruster due to abnormal decomposition

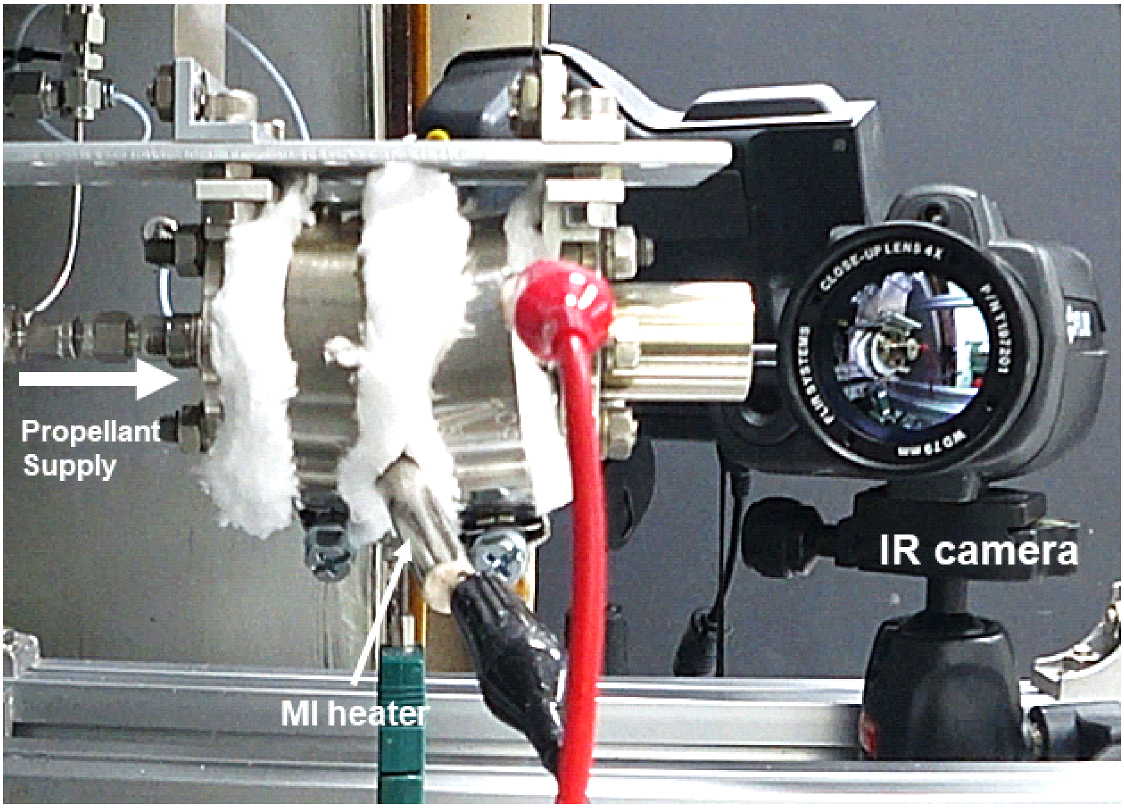


Fig. 31 Experimental equipment for flame observation of low-toxicity propellants

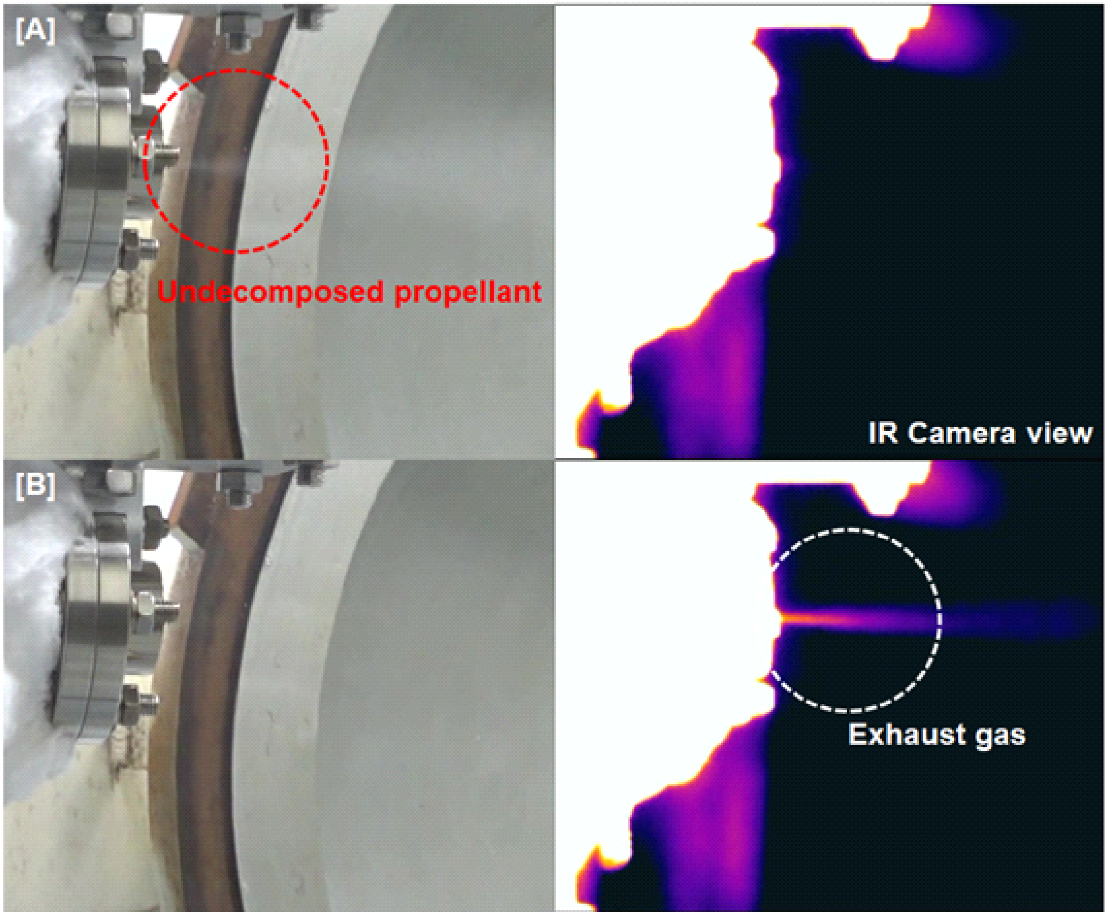


Fig. 32 Flame visible/infrared observation according to the decomposition condition of the propellant

III. Results and Discussion

A. Catalyst Selection through Isoconversional Method

First, the catalyst was selected through the isoconversional method. Through the TGA, the results of analysis of each catalyst (Hexaaluminate, Cu, Ir) to which the same propellant was applied were obtained. Analysis was carried out by changing the heating rate (β) for each sample from 2°C/min to 10°C/min. At this time, the peak temperature (T_m) was obtained through the first derivative of the obtained result.

Based on the results, applying Eq. 6 confirmed the results shown in Fig. 31. The activation energy of the Ir/Al_2O_3 catalyst was the lowest at 176.4 kJ/mol, indicating that the decomposition reaction was excellent compared to other samples.

In the result of Table. 6, It was not significantly different from the result when selecting the catalyst by the isoconversional method. As a result of experiments under the lowest reaction conditions, the Hexaaluminate catalyst showed a sudden drop in temperature at the same time as the supply of the propellant. On the other hand, in the case of the Ir/Al_2O_3 catalyst, decomposition was started at a lower temperature than the experimental conditions of the other catalysts, and it was confirmed that the temperature and pressure increased. Through this result, it was confirmed that the reactivity of the Ir-based catalyst was excellent in the actual thruster application test.

Based on these results, the experiment was conducted with Ir/Al_2O_3 catalyst.

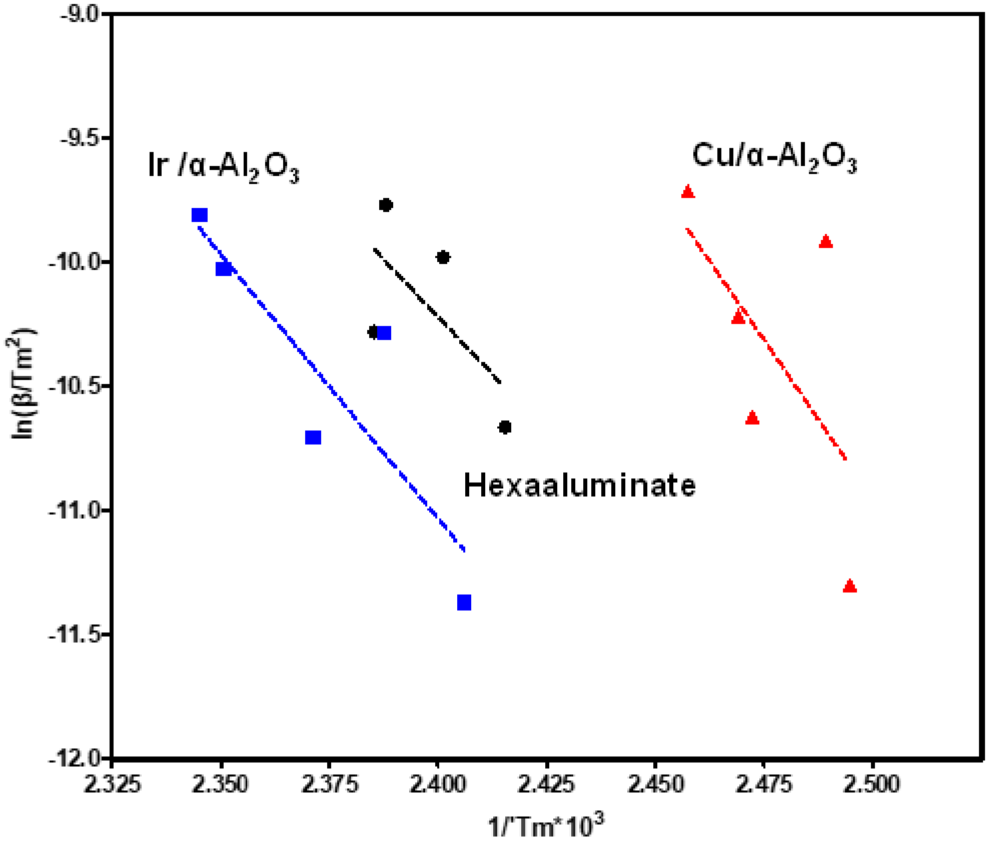


Fig. 33 Comparison of decomposition characteristics by catalyst through isoconversional method(Kissinger method)

Table. 6 Catalyst analysis result through isoconversional method and its lowest decomposition reaction result

	Hexaaluminate	Cu/Al_2O_3	Ir/Al_2O_3	(Unit)
E_a	272.5987	210.5575	176.4065	kJ/mol
A	2.03648E+31	1.39644E+24	4.49E+18	min^{-1}
T_{Min_decomp}	317.7	354.86	301.26	℃
ΔT	-126.9	1.75	3.42	℃
P_{Peak}	3.31	3.34	6.61	bar

B. Propellant/Catalyst Combustion Test

Next, a combustion test in an actual thruster was performed. Through the above analysis, the Ir catalyst was charged, and the experiment was started at the catalyst bed temperature reached 350° C.

Fig. 32 shows the combustion results according to the primary propellant supply. At the same time as the propellant was supplied, the pressure increased and the pressure remained stable for the duration of the propellant supply. In the case of temperature, the temperature rise was slightly slower due to the heat loss, but this seems to be due to the large heat loss compared to the small heat capacity due to the characteristics of the small thruster.

Fig. 33 shows the combustion results according to the supply of secondary propellants. However, in the second test, the temperature inside the catalyst bed was sufficiently preheated due to the decomposition of the propellant in the first test, resulting in a rapid temperature increase compared to the first test. In the second experiment, it was also confirmed that the pressure inside the catalyst bed was maintained stably after the sudden increase in pressure at the same time as the supply of the propellant.

Due to the internal decomposition reaction of the catalyst bed, the highest temperature inside the thruster was 1019.06℃ in the first experiment and 998.86℃ in the second experiment.

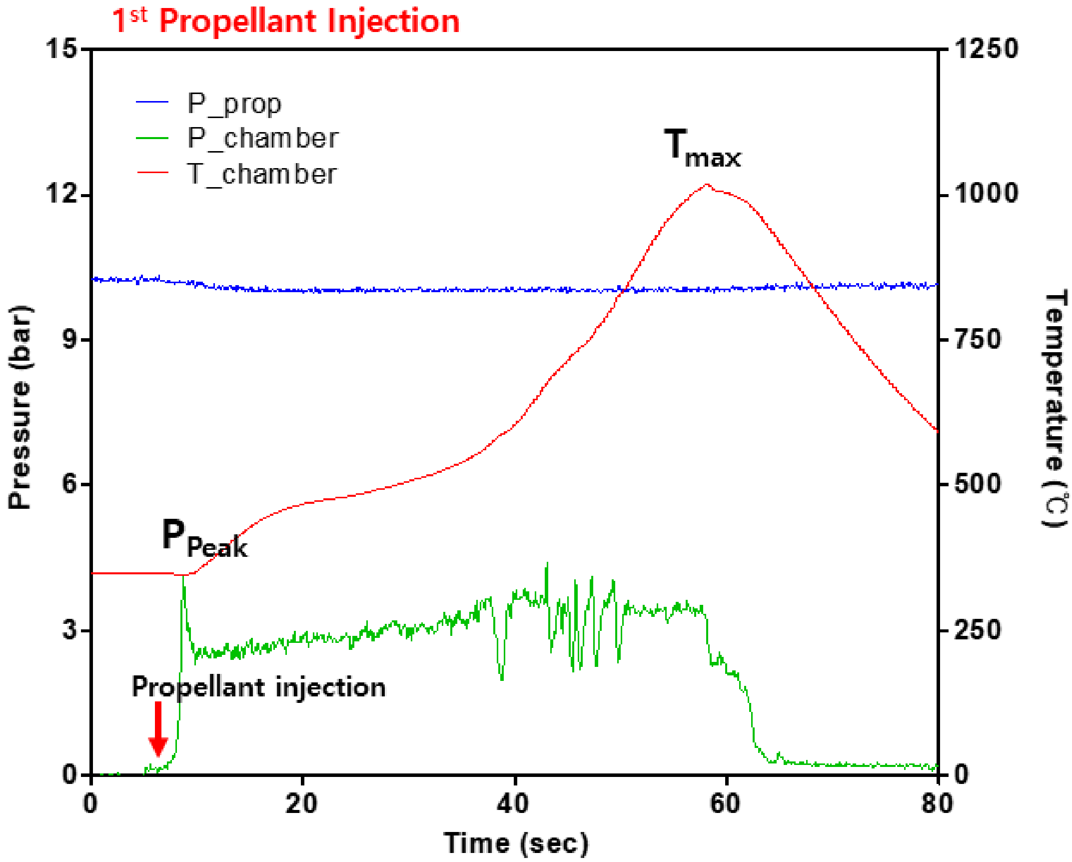


Fig. 34 1st low-toxicity propellant/catalyst combustion test evaluation result

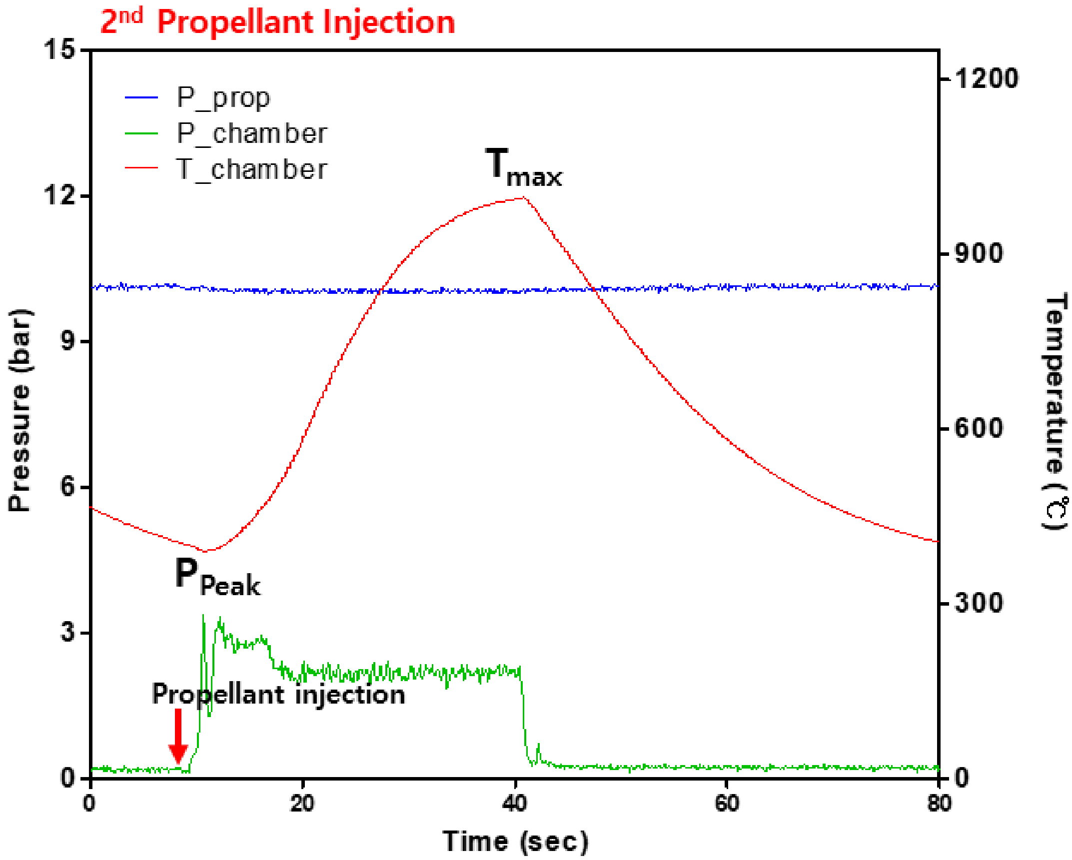


Fig. 35 2nd low-toxicity propellant/catalyst combustion test evaluation result

C. Catalyst Mechanical Strength Analysis

The thruster obtains a moment to control the object by injecting the gas generated from the decomposition of the propellant. At this time, the liquid propellant is sprayed on the surface of the catalyst, and decomposition occurs simultaneously, and at the same time as the decomposition, a rapid temperature/pressure increase occurs, thereby forming thermal shock and high pressure due to the rapid temperature rise in the catalyst bed. This environmental change causes various changes to the catalyst. Therefore, the durability of the catalyst is also closely related to the durability of the thruster.

The state of the catalyst before and after the experiment was compared due to the environmental change in the catalyst bed according to the supply of the propellant. In the experiment, precious metal catalysts for propellant decomposition were supported on α-Al_2O_3 and γ-Al_2O_3 supports, respectively. The catalysts before and after the experiment were collected and analyzed through a scanning electron microscope (SEM) and Brunauer–Emmett–Teller (BET).

In general, the specific surface area of γ-Al_2O_3 is higher than that of α-Al_2O_3, which can be considered to be related to the mechanical strength of the support. In the case of the γ-Al_2O_3 catalyst used in the experiment, an analysis for durability evaluation could not be performed due to the loss of the shape of the catalyst through the high temperature/high pressure environment generated during the experiment. These results indicate that the durability cannot be guaranteed in the high temperature/high pressure decomposition environment of γ-Al_2O_3. On the other hand, in the case of α-Al_2O_3, the shape of the support was maintained. As can be seen in Fig. 34, the surface of the catalyst confirmed through SEM can be seen in the form of particles after the experiment, unlike the catalyst layer evenly distributed on the surface before the experiment. This phenomenon means that the area where the catalyst precursor and the propellant are closed is reduced, and as a result, the catalyst efficiency may be lowered.

Table. 7 shows the BET measurement results. BET analysis was performed to confirm the change in specific surface area before and after the experiment. At the time of catalytic decomposition, it was confirmed that the change in specific surface area of the catalyst was reduced at the maximum temperature inside the thruster. Also, it was confirmed that the specific surface area decreased linearly with the change in the decomposition temperature of the propellant even in the change of the BET by the decomposition temperature of the propellant.

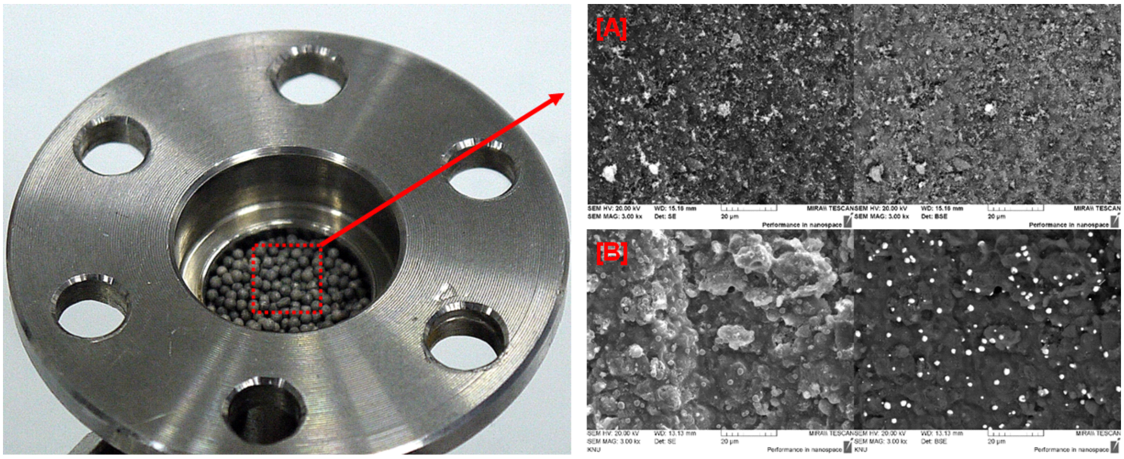


Fig. 36 Comparison of catalyst surface changes before and after the decomposition experiment

Table. 7 Catalyst BET change with various propellant decomposition temperature

State	Before Experiment	After Experiment		
Temperature (K)	-	828.932	998.303	1208.742
BET (m^2/g)	6.0639	4.7321	4.2821	3.8637

D. Thruster Test with Low-toxicity Propellant

While performing the performance evaluation on a 1N thruster, the abnormal test results were first checked to compare the test results. First, Fig. 35 confirms the abnormal decomposition results. Each time the propellant is injected, the temperature suddenly decreases, but the catalyst does not decompose normally and seems to have the effect of cooling the inside of the catalyst bed. And although the pressure or thrust increased slightly, it was because it had the same effect as a simple injection of a propellant like a cold gas thruster. As can be seen from the IR observation results shown in Fig. 36, nothing was found at the end of the thruster even after propellant injection. Due to this abnormal decomposition phenomenon, the thruster did not reach the target thrust, and the experiment was terminated because a sudden temperature drop could cause dangerous situations such as the accumulation of propellant inside the thruster and the sudden decomposition reaction.

Next, the following results were obtained in the case of normal decomposition according to the continuous supply of the propellant. First, the preheating temperature of the catalyst bed was 400~450℃, and the experiment was conducted at a supply pressure of 20 bar. Propellant supply times are 1,000 ms and 1,250 ms, respectively. Fig. 37 is a graph of the experimental results. The catalyst bed pressure according to the propellant injection reached a maximum of 14 bar and the temperature reached a maximum of about 740°C. As a result of the experiment, the thrust was measured to be 966.36 mN, I_{sp} 246.35 s, and 908.83 mN, I_{sp} 231.69 s. And in Fig. 38 and Fig. 39, a long flame was observed at the end of the thruster, unlike the previous abnormal decomposition experiment.

Next, the experimental results under the pulse supply condition of the propellant are shown in Fig. 40. Propellants were supplied at 200 ms duration. As a result, decomposition is activated for each propellant supply, and the thruster temperature and pressure gradually increase. During the experiment, the maximum catalyst bed pressure and temperature reached 9.46 bar and about 459.8°C, respectively. A maximum thrust of 1085.96 mN was obtained. Considering the dynamic characteristics, the maximum thrust was 932.67 mN as a result of filtering through the low path filter(LPF), which was converted to specific impulse and calculated as 237.9 s. In particular, propellants were supplied nearly twice as much as in continuous supply, but thrust values were similar despite relatively low temperature and pressure. The change in temperature and pressure between the two supply conditions is as follows. As shown in Fig. 41, it was confirmed that the size of the flame according to the thrust value was observed in the flame observation according to the injection of the propellant.

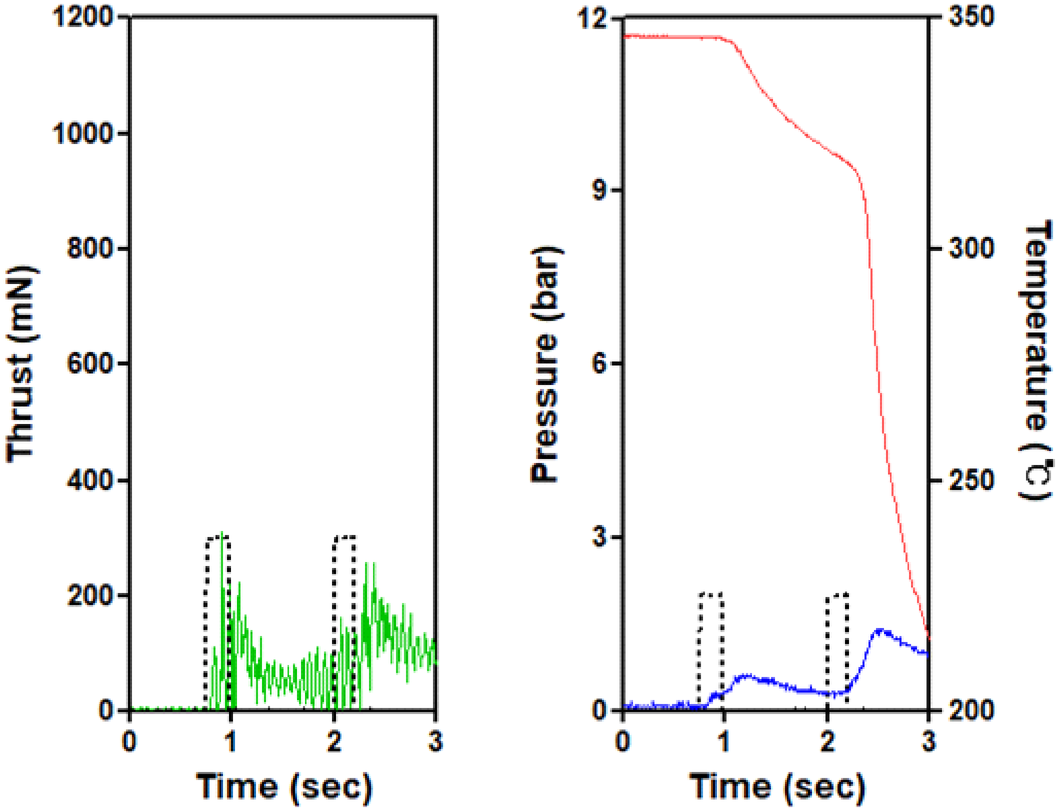


Fig. 37 Experimental results according to the abnormal decomposition status

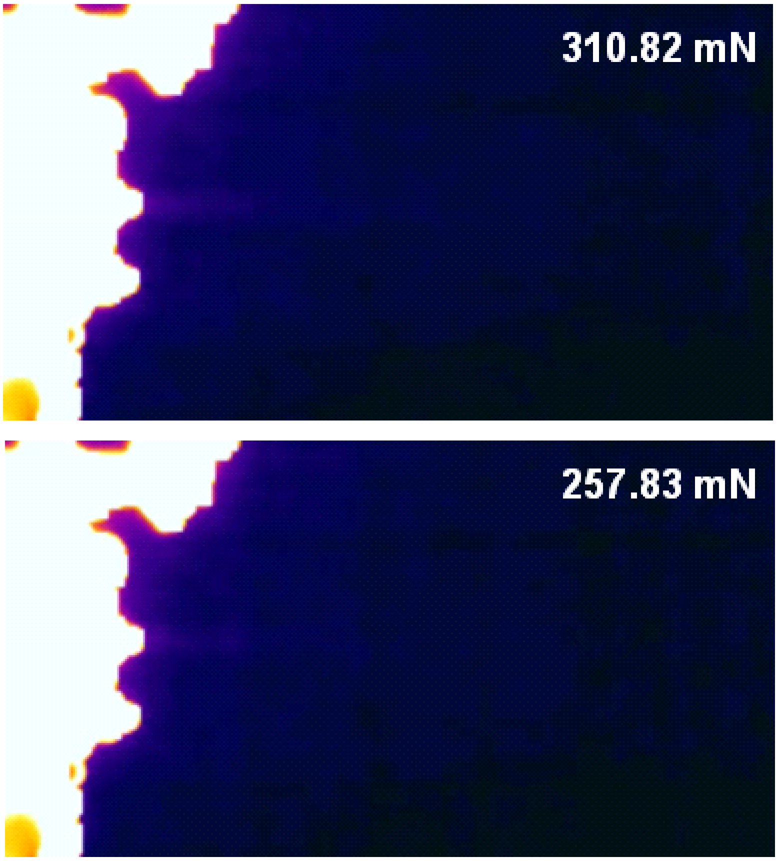


Fig. 38 Observation results according to the abnormal decomposition status

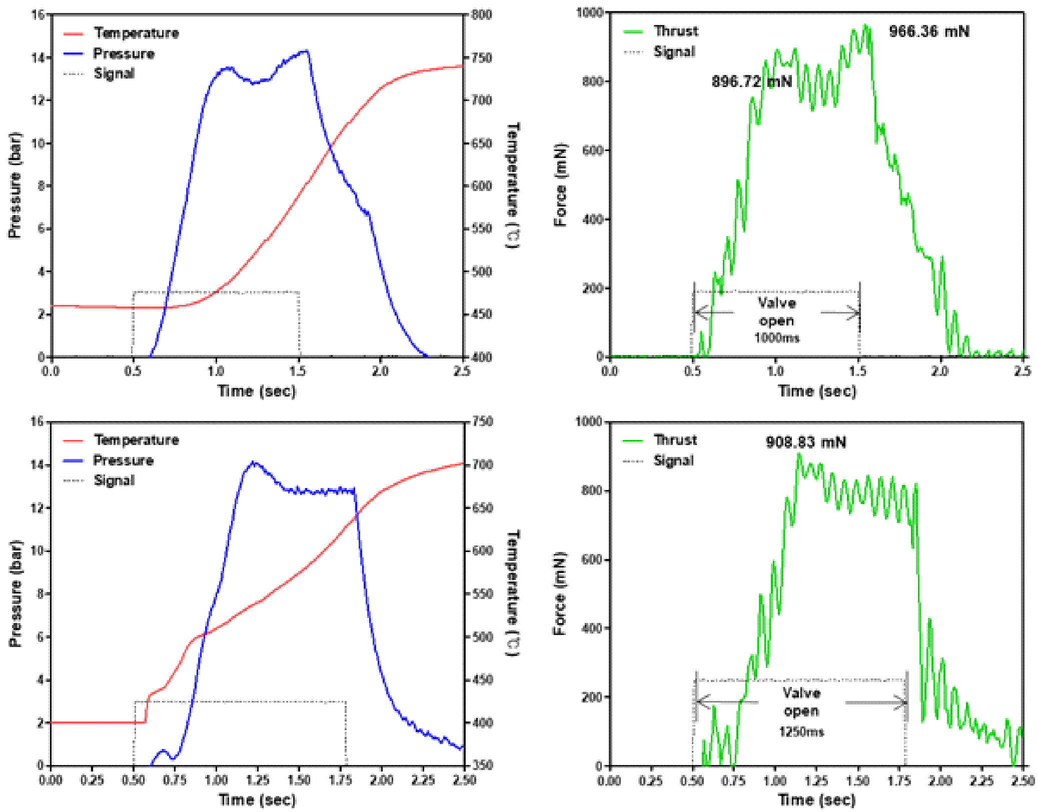


Fig. 39 Experimental results according to the normal decomposition status in continuous mode (1,000ms, 1,250ms propellant injection)

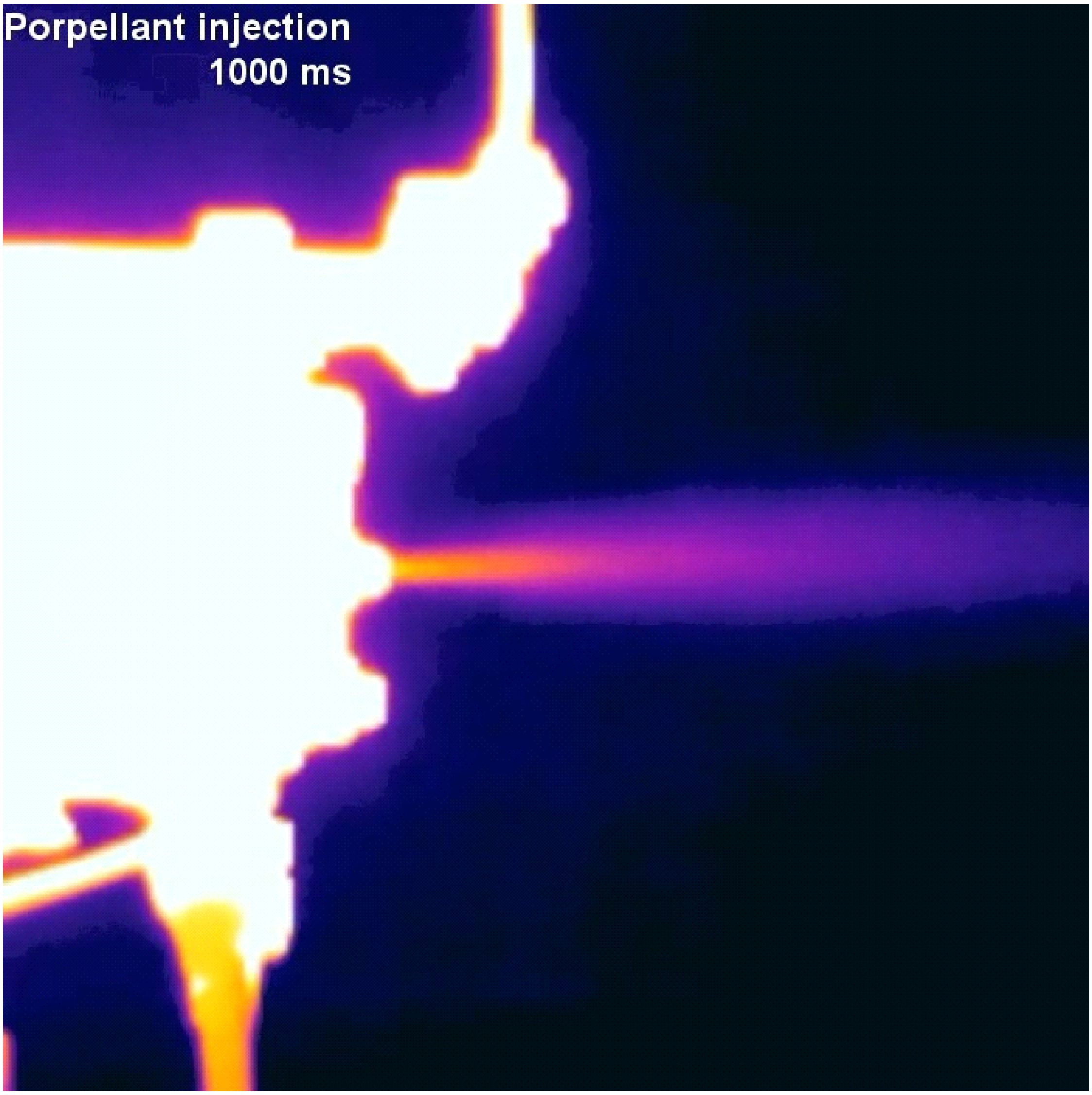


Fig. 40 Observation results according to the normal decomposition status in continuous mode (1,000ms propellant injection)

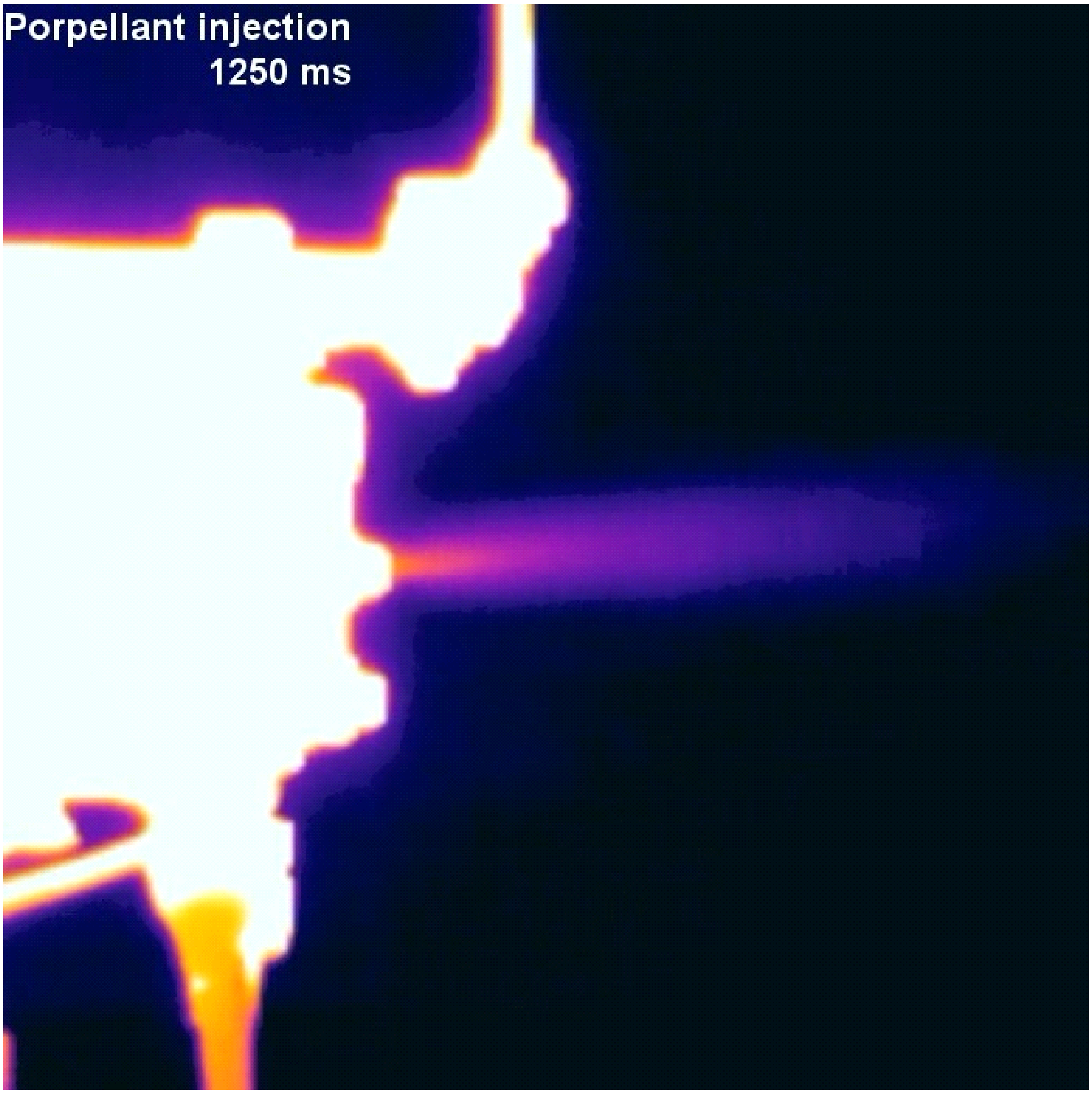


Fig. 41 Observation results according to the normal decomposition status in continuous mode (1,250ms propellant injection)

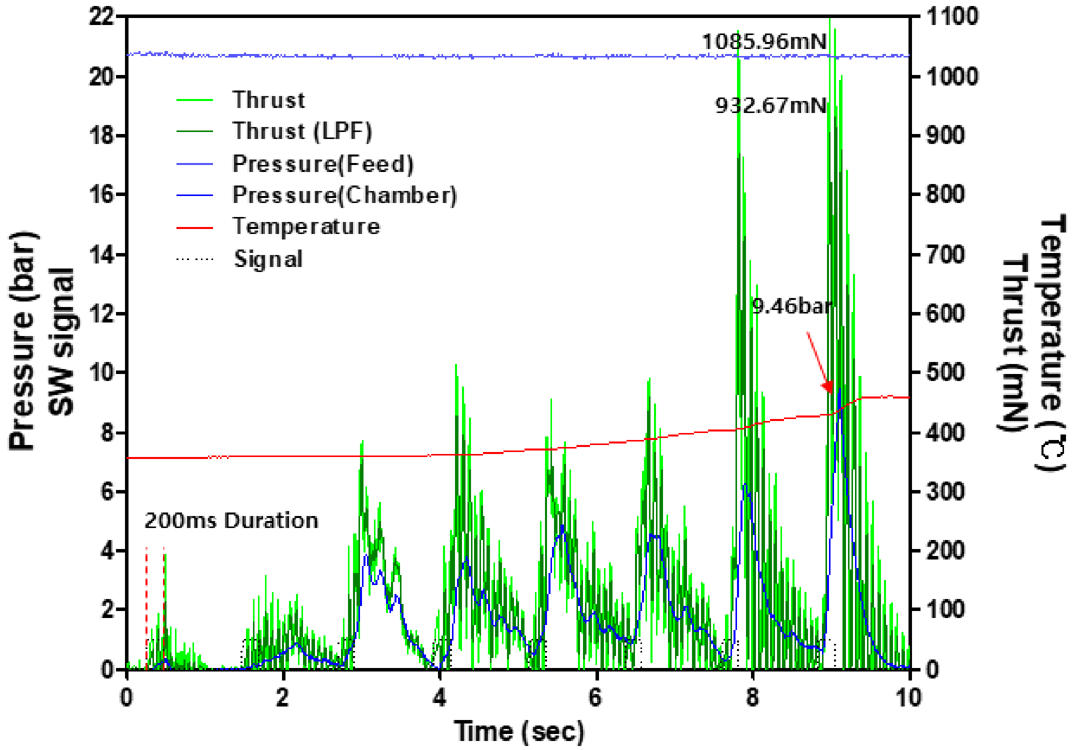


Fig. 42 Experimental results according to the normal decomposition status in pulse mode (200ms propellant injection duration)

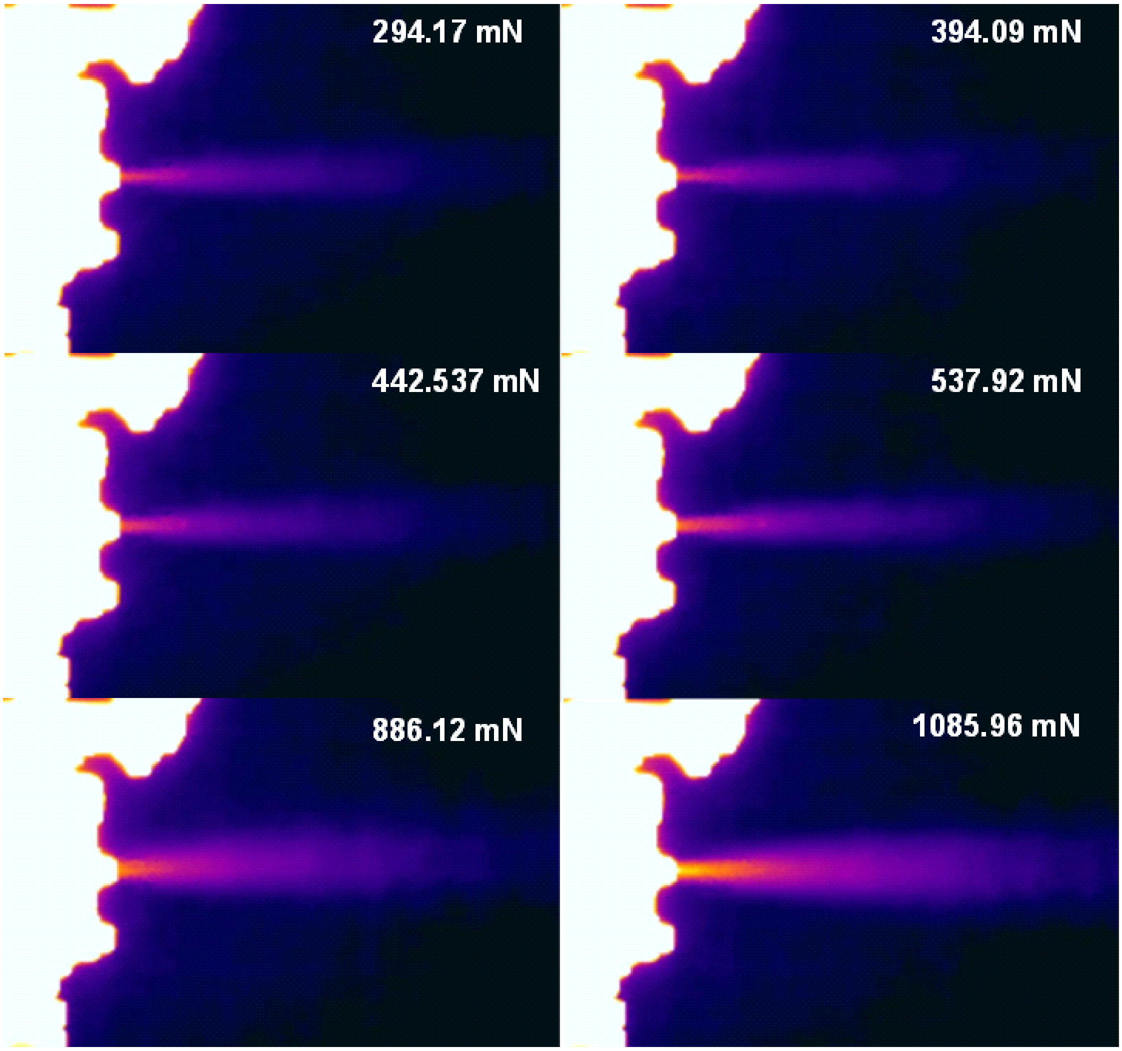


Fig. 43 Observation results according to the normal decomposition status in pulse mode (200ms propellant injection duration)

E. Visualization of Flameless Low-toxicity Propellant Combustion

Figure 42 shows the results of Mid-range IR observation of the decomposition flame in the pulsed state of the propellant. Performance prediction was conducted through observation results. A flame that appeared as a result of propellant decomposition at the thrust nozzle appeared at the time of propellant injection. The difference was more apparent when compared with the previous abnormal decomposition results. The change in the nozzle according to decomposition in a similar low thrust section was identified. Afterwards, in another thrust section, a clearer flame shape was observed at the end of the nozzle. In particular, in the 1085.96mN thrust section, a clearer flame was observed from the nozzle.

Based on this observation, the area of the decomposed gas by thrust value of the rear end flame of the thrusters was identified. The results of the area ratio of this to the maximum thrust section are as shown in Fig. 43. As a result of comparing the tendency with the thrust value based on the ratio of the flame area, the area ratio also increased as the thrust value increased, confirming the tendency between them. Through this method, it was confirmed that performance prediction is possible based on the observation results of the low-toxicity propellant-based thrust system decomposition gas in operation.

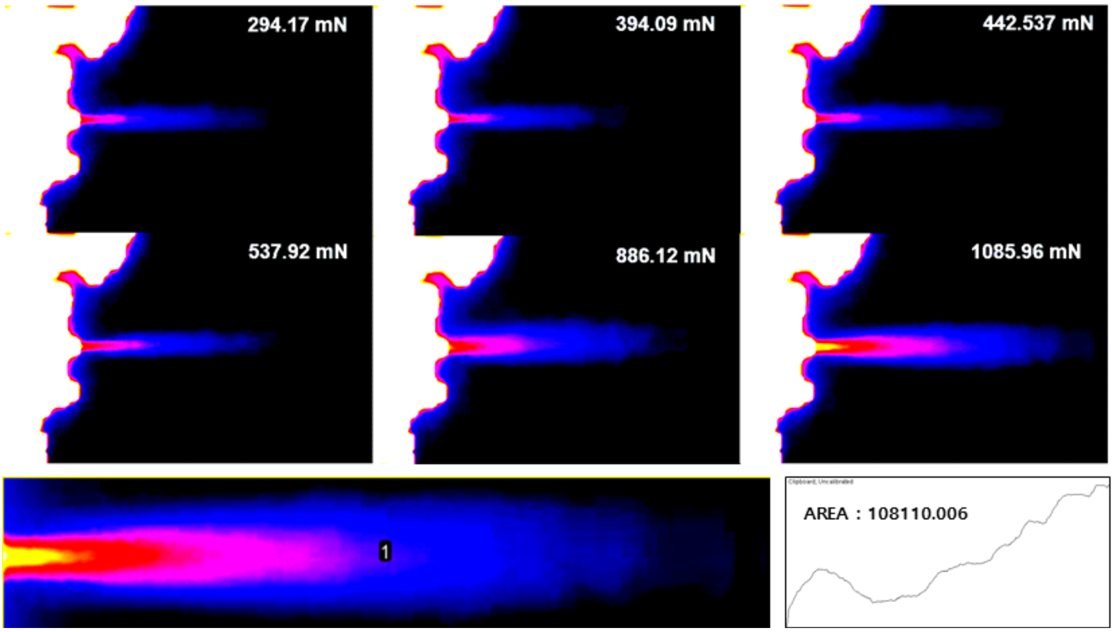


Fig. 44 Decomposition flame observation result filtering and flame area measurement

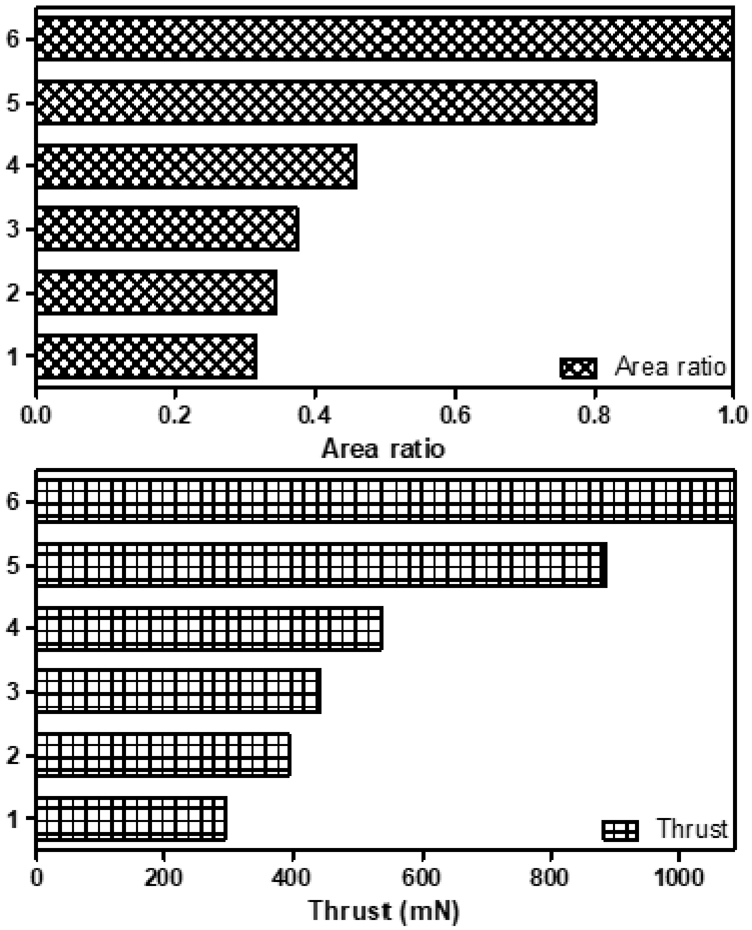


Fig. 45 Prediction of thrusters performance by comparing the area ratio and thrust of the observed flame

IV. Conclusion

As demand for space launch vehicle and satellites increases, so does their impact on environmental issues. This, in turn, has led to the need for replacement of hydrazine-based propellants, where environmental and toxicological problems have been presented. Several overseas space development institutions have completed the demonstration of technologies related to low-toxicity propellants, but the domestic research and technology maturity related to low-toxicity propellants to replace hydrazine propellants is low, and the research data related to them is also insufficient. In this study, the optimal combination through propellant, catalyst synthesis, and performance evaluation was selected and the design and fabrication of 1 N class thrusters and thrust measurement systems were performed to evaluate them. Performance evaluation was performed through experiments. Prior to the experiment, a low toxicity propellant, catalyst synthesis and test evaluation system were constructed. The thruster was designed based on the numerical value obtained by calculating the chemical balance, and the thrust measurement system was designed in a pendulum manner to perform precise thrust measurement.

- Evaluation/screening of propellant/catalyst decomposition properties

In this study, the test was conducted by synthesizing the low-toxicity propellant based on ADN. And for catalytic decomposition, the metal precursors were immersed through the impregnation method to carry out catalyst synthesize. The decomposition characteristics between propellants/catalysts were compared by chemical kinetics theory. The characteristics of decomposition between propellants/catalysts were evaluated through isoconversional methods, such as not the commonly used Arrhenius equation. The isoconversional method can easily and simply obtain activation energy and frequency factors according to the decomposition of propellants and catalysts compared to the Arrhenius equation. The results of the analysis showed excellent Ir-based catalyst performance, and the results of the actual experiment also showed that propellant decomposition took place under much lower conditions.

- Catalyst durability evaluation

The combustion test of the propellant was carried out based on the selected catalyst. With the supply of propellants, the temperature and pressure increased and the normal combustion process could be seen. The catalyst status before and after the combustion test was compared through SEM and BET analysis. The thrust is formed by catalytic decomposition of propellants, and the change in the state of the catalytic bed in the process of decomposition affects the durability of the catalyst, which has a great connection with the performance of the thrusters. The results of the experiment were analyzed to confirm the change in the catalytic layer and the specific surface area due to the increase in catalyst bed temperature following catalytic decomposition. The result is a reduction in the area of the reaction between the active point of the catalyst and the propellant. Therefore, it was confirmed that the long-term durability of catalysts is needed through the development of catalysts with high-temperature durability or control of the decomposition environment of catalysts.

- Low toxicity propellant flame visualization and performance prediction

The results of the decomposition state during thrust experiments were compared. First of all, at the time of abnormal propellant decomposition, the temperature of the propellant supply decreased, and the additional supply of propellant reduced the temperature inside the catalyst bed. This caused the additional supply of propellants to cool the inside of the thruster. On the other hand, during normal decomposition, temperature, pressure, and thrust could be seen to rise with the propellant supply.

In addition, decomposition status and performance prediction were performed through flame observation of the thruster. Differences in decomposition in low thrust sections were evident. Although the thrust level was similar, the flame was not observed through the IR camera in the abnormal decomposition, but the flame was observed in the rear end of the thruster in the normal decomposition. It seems that the CO_2 generated by the decomposition of the propellant corresponds to the range of the observed spectrum of the mid-range IR camera, which is why it was observed at the thruster nozzle[23].

Depending on the degree of decomposition of the propellant, the final amount of CO_2 produced will be different. And the thrust value will change depending on the degree of decomposition of the propellant. Through this, it was judged that the relationship between the size of the observed flame and the thrust could be predicted. Therefore, analysis was performed as a result of the thrust test in pulse mode. First, the observation result of the flame was filtered, and then the area of the decomposed flame for each thrust value was obtained. This was compared with the area ratio of the maximum thrust section. As a result of comparing the area ratio of the flame at the end of the thruster, the area ratio also increased as the thrust value increased, confirming the tendency between them. Through this, it was confirmed that the performance of the thruster can be predicted only by observation of the decomposition gas of the thruster in operation.

Most of the existing propellant/catalyst evaluations are made individually or researched on an extension of existing experimental and evaluation results. This research process makes it difficult to establish universal performance evaluation and standards for propellants and catalysts. As a result, it is difficult to uniformly apply the performance evaluation method according to the composition change of the new material. However, by applying the isoconversional method, it was easy to compare the performance between the low-toxic propellant/catalyst.

It was also confirmed that performance prediction is possible based on simple observations in situations where thrust must be specified without thrust measurement equipment. Flame from a typical thruster is observed in the visible light area and rarely used as a performance data for thrusters. On the other hand, flameless low-toxic propellants were able to correlate these observations with the decomposing and thrusting performance of propellants because only the wavelength bands of a particular substance were observed and utilized in reversely. And the results obtained through these processes were applied to the actual thrusters to ensure stable performance results.

In this study, the focus was on configuring how catalysts and propellants are evaluated in an integrated manner and how performance is predicted. Procedures and techniques were developed to assess the propellants and catalysts, which were generally evaluated individually. It also provided a basis for performing performance analysis relatively easily despite changes in the composition of propellants or catalysts. It is hoped that this study could be a good reference for evaluating a series of new materials and low-toxic propellants and catalysts that will be studied in the future.

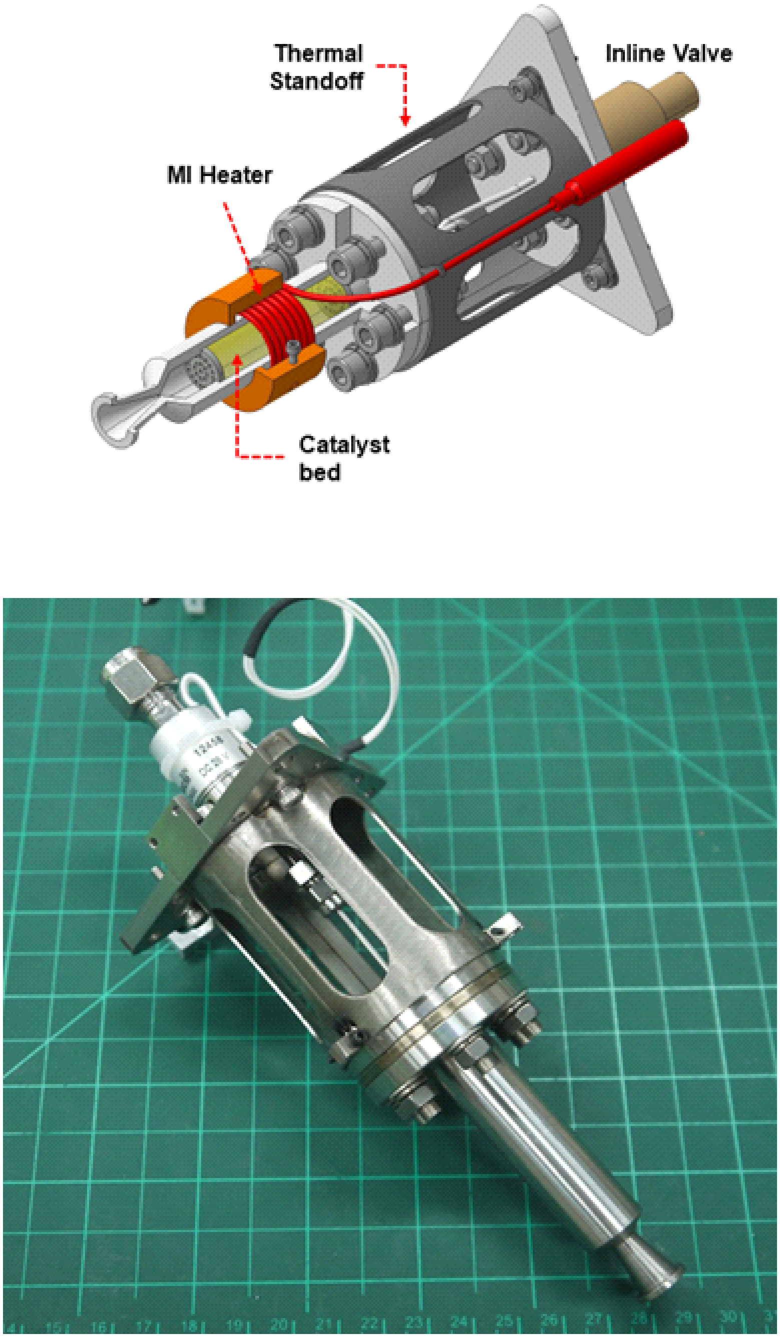


Fig. 46 Assembled EM(Engineering model) low-toxicity propellant thruster

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