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Propulsion Systems, Propellants, Green Propulsion Subsystems and their Applications: A Review

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Abstract

A wide range of propellants, and propulsion systems in space exploration by aircrafts or space vehicles was studied, developed, investigated, and commercialized. Liquid, solid, or hybrid propellants have been used for rocket's launches. In this review, a consistent definition of space propulsion systems, including solid, liquid and hybrid has been given with up-to-date state of developments. A comparison of their performances was made by theoretical and experimental specific impulses. On the other hand, ammonium perchlorate and hydrazine were used as propellants for rocket's launches and for satellite's maneuverings; respectively. However, their high toxicity and their storage problem pushed researchers and scientists to investigate and develop other eco-friendly, propellant systems, so called "green propellants", for launch or reaction control systems of satellites.

Abbreviations

I_{sp}	Specific impulse
RP-1	Refined kerosene
LOx	Liquid oxygen
LH_2	Liquid hydrogen
MMH	Monomethyl hydrazine
UDMH	Asymmetrical dimethylhydrazine
NTO	Nitrogen tetroxide
MON	Mixed oxides of nitrogen
OMS	Orbital maneuvering system
RCS	Reaction control system
FLOx	Liquid fluorine and liquid oxygen
PBAN	Polybutadiene acrylic acid acrylonitrile
HTPB	Hydroxy-terminator polybutadiene
PAM	Payload Assist Module
IUS	Inertial Upper Stage
CEA	Chemical equilibrium applications
AP	Ammonium perchlorate
HAN	Hydroxylammonium nitrate
ADN	Ammonium dinitramide
AN	Ammonium nitrate
HA	Hydroxylamine
HN	Hydrazinium nitrate
H_2O_2	Hydrogen peroxide
N_2O	Nitrous oxide
EILS	Energetic ionic liquids
HEHN	Hydroxyethyl hydrazinium nitrate
TEAN	Tetraethyl ammonium nitrate
SHP163	Sei-Hori propellant
AF-M315E	Air Force propellant
REACH	Registration, evaluation, authorization and restriction of chemicals

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1. Introduction

A propellant is a chemical compound composed of fuel and an oxidizer that is burnt to create thrust in rockets. In space propulsion systems, fuel is the substance that burns when combined with oxygen-generating gas. An oxidizer is a substance that, when combined with fuel, produces oxygen. The mixture ratio is the proportion of fuel to the oxidizer. Propellants can be classified as liquid, solid, or hybrid [1]. Specific impulse, measured in seconds, is used to evaluate the efficacy of rocket propellants. It indicates the amount of thrust generated by one kilogram of fuel in 1 s. Even though the type of propellant affects specific impulse, its exact value depends on how the rocket engine is built and how it is used [2].

An excellent liquid propellant has a high specific impulse and a fast exhaust gas ejection speed. This means that exhaust gases have a low molecular weight and a high temperature of combustion. Nevertheless, there is one more important thing to think about, namely the density of the propellant. Low-density propellants need larger storage tanks, which makes the launch vehicle heavier. The temperature of the storage area is indeed vital [1]. For a propellant that needs to be stored at a low temperature, such as cryogenic, thermal insulation is needed, which adds to the weight of the launcher. Also, the handling, transporting, and storing of those highly toxic compounds is a very important issue and needs special procedures. Also, some propellants are very corrosive, but materials can be used to build rockets that are resistant to certain propellants [2]. Currently, propulsion systems aboard satellites and spacecraft use propellants such as hydrazine and its derivates due to their excellent performance with a theoretical specific impulse of 230 s. However, compared to less dangerous propellants, their high toxicity and potential to trigger cancer make them expensive to handle, store, and transport. Because of this disadvantage, people are becoming interested in "green" or less toxic propellants [3]. There is presently no generic replacement for hydrazine. As a result, a variety of alternatives is being investigated. In general, the division of mission requirements depends on whether they are short-term or long-term in nature. Short-term missions usually involve operations that last no more than a few days, often related to launcher propulsion. On the other hand, long-term missions may have lifetimes that extend up to or even beyond 15 years, and are typically associated with satellite propulsion [4].

In this review, we present an up-to-date stae-of-the-art overview of different propulsion systems and the possible replacement by so-called eco-friendly or "green" environmentally propellants.

2. Propulsion systems

Propulsion systems or propellants are divided into two categories: mono- and bi-propellant systems. The monopropellant system contains oxidizer and fuel in the same tank. However, bipropellant system contains oxidizer and fuel in two separated tanks. Figure 1 illustrates both systems.

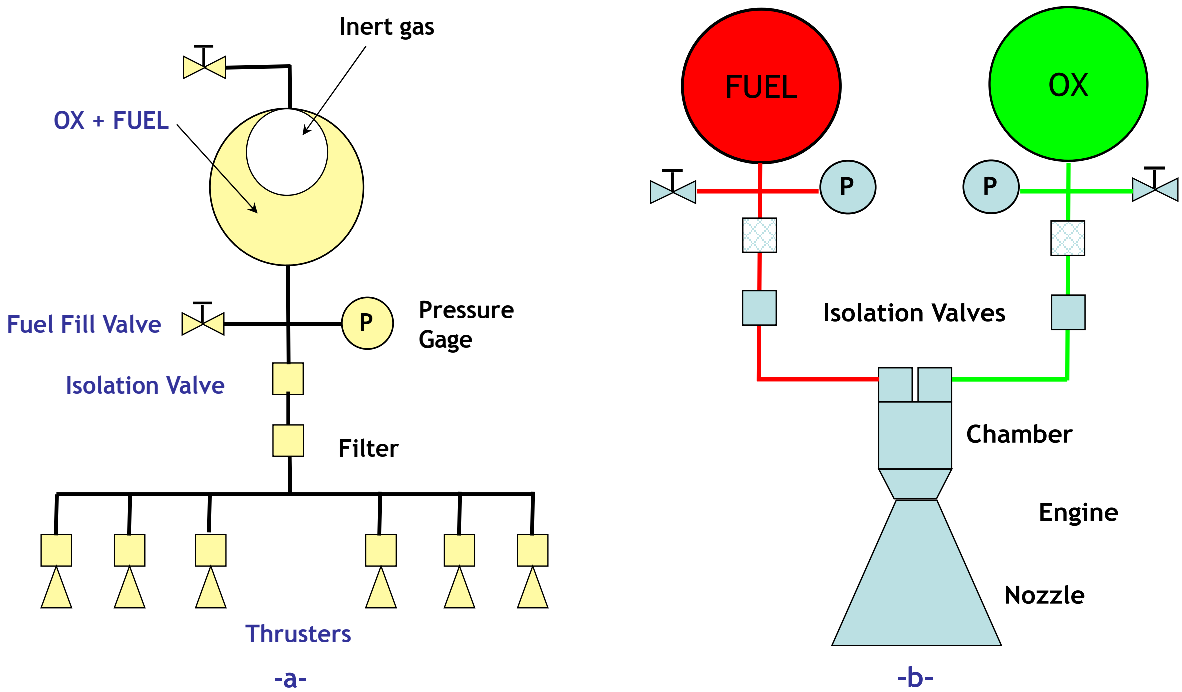

Fig. 1. (a) Mono- and (b) bipropellant systems.

Table 1
Properties of rocket propellants [5]

Compound	Chemical formula	Molecular weight (g mol^{-1})	Density (g cm^{-3})
Liquid oxygen	O_2	32.00	1.14
Liquid Fluorine	F_2	38.00	1.5
Nitrogen Tetroxide	N_2O_4	92.01	1.45
Nitric Acid	HNO_3	63.01	1.55
Hydrogen Peroxide	H_2O_2	34.02	1.44
Nitrous Oxide	N_2O	44.01	1.22
Chlorine Pentafluoride	ClF_5	130.45	1.9
Ammonium Perchlorate	NH_4ClO_4	117.49	1.95
Liquid Hydrogen	H_2	2.016	0.071
Liquid Methane	CH_4	16.04	0.423
Ethyl Alcohol	C_2H_5OH	46.07	0.789
n-Dodecane (Kerosene)	$C_{12}H_{26}$	170.34	0.749
RP-1	$C_nH_{1.953n}$	175	0.82
Hydrazine	N_2H_4	32.05	1.004
Methyl Hydrazine	CH_3NHNH_2	46.07	0.866
Dimethyl Hydrazine	$(CH_3)_2NNH_2$	60.1	0.791
Aluminum	Al	26.98	2.7
Polybutadiene	$(C_4H_6)_n$	3000	0.93

Different propellant systems with their formulas, molecular weights and densities were summarized in Table 1 [5].

2.1. Liquid propulsion

There are three types of liquid propellants used in rocketry: petroleum, cryogens, and hypergolic.

2.1.1. Petroleum

Fuels are a combination of complex hydrocarbons and organic chemicals that solely contain carbon and hydrogen and are processed from crude oil. In the United States, rocket fuel is a form of highly refined kerosene known as RP-1. The most common way to use petroleum fuels is with liquid oxygen as an oxidizer. Kerosene has a lower specific impulse than cryogenic fuels, but it outperforms hypergolic propellants in general. When the need for a clean-burning petroleum rocket fuel was identified in the United States in 1957, specifications for RP-1 were first established [6]. Experiments with jet fuels in the past resulted in tarry residue in the cooling channels of the engine and excessive soot, coke, and other deposits in the gas generator. Even with the more severe criteria, kerosene-burning engines still create enough residue to restrict their operating lives. The Atlas and Delta II launch vehicles' first-stage boosters employ liquid oxygen and RP-1 as propellants [7]. The early stages of the Saturn 1B and Saturn V rockets were also powered by it.

2.1.2. Cryogenic

Cryogenic propellants are made of liquid gases that are kept at very low temperatures. They usually have liquid hydrogen (LH_2) as the fuel and liquid oxygen (LO_2 or LOx) as the oxidizer. At -253 °C, hydrogen stays liquid, whereas oxygen remains liquid at -183 °C. Cryogenic propellants are difficult to store for an extended period due to their low temperatures [6]. As a result, they are less suited for military rockets that need to be ready to fire for months at a time. Furthermore, because liquid hydrogen has a relatively low density (0.071 g cm^{-3}), it requires a much larger storage volume than other fuels [5]. However, the high efficiency of liquid oxygen/liquid hydrogen

[5] makes it worth dealing with them when reaction time and storage are not very important. Most other rocket fuels have about 40% less specific impulse than liquid hydrogen. The high-efficiency main engines of the Space Shuttle use LOx and LH_2 as fuel. They were also used to make energy in the upper stages of the Saturn V and Saturn 1B rockets, as well as in the upper stage of the Centaur rocket, which was the first LOx/LH_2 rocket built in the US in 1962 [8].

Another cryogenic fuel with favorable qualities for space propulsion systems is liquid methane (-162 °C). Methane burns better than state-of-the-art storable propellants when mixed with liquid oxygen, but it does not increase the volume as LOx/LH_2 systems do. This means that, compared to typical hypergolic propellants, the total weight of the vehicle is lower. LOx/methane is also safe and burns cleanly. Future missions to Mars will probably use methane as fuel, which can be made mostly from materials already on Mars. LOx/methane has never flown, and there have only been a few tests on the ground [9]. Engineers have also made and tested engines that burn liquid fluorine at -188 °C. Fluorine is not only very harmful, but it is also a super oxidizer that reacts violently with almost everything except nitrogen, lighter noble gases, and fluorinated compounds. Even though fluorine has all these disadvantages, it is a very good fuel for engines. It can also be mixed with liquid oxygen to improve the performance of engines that burn LOx. This mixture is called FLOx. Because fluorine is so dangerous, most countries that go into space have stopped using it [6].

2.1.3. Hypergolic

Hypergolic propellants consist of fuels and oxidizers that spontaneously ignite upon contact and do not need an ignition source. Hypergolics are advantageous for spacecraft maneuvering systems because they are easy to initiate and restart. Hypergolics also do not have the same storage issues as cryogenic propellants because they stay liquid at room temperature. They are very hazardous and should be handled with caution [10]. Hypergolic fuels include hydrazine, monomethyl hydrazine (MMH), and asymmetrical dimethylhydrazine (UDMH). The most sophisticated rocket fuel is hydrazine, but it cannot be utilized as a coolant because of its high freezing point and instability. When the freezing point is a problem, as in spaceship propulsion applications, MMH is more stable and provides optimum performance. The lowest freezing point and sufficient thermal stability make UDMH ideal for use in large regeneratively cooled engines. As a result, UDMH, although being the least efficient of the hydrazine derivatives, is frequently utilized in launch vehicle applications. Blended fuels, such as Aerozine 50 (or "50-50"), which is a 50/50 combination of UDMH and hydrazine, are also widely utilized. Aerozine 50 is almost as stable as UDMH and provides better performance [11]. As oxidizers, nitrogen tetroxide (NTO) or nitric acid are often used. The most frequent nitric acid formulation in the United States is type III-A, also known as inhibited red-fuming nitric acid (IRFNA), which is composed of HNO_3 + 14% N_2O_4 + 1.5–2.5% H_2O + 0.6% HF (added as a corrosion inhibitor) [6]. Nitrogen tetroxide is less corrosive and performs better than nitric acid, although it has a higher freezing point. When the freezing point is not a problem, nitrogen tetroxide is normally the oxidant of choice. However, the freezing point can be decreased by adding nitric oxide, giving mixed oxides of nitrogen (MON). The number in the description, such as MON-3 or MON-25, indicates the proportion of nitric oxide by weight. While pure nitrogen tetroxide has a freezing point of around -9 °C, MON-3 and MON-25 have freezing points of -15 °C and -55 °C, respectively [5]. The initial military requirements for IRFNA were issued in 1954, followed by UDMH standards in 1955. NTO/Aerozine 50 propellant is used in the Titan launch vehicle family and the Delta II rocket's second stage. The orbital maneuvering system (OMS) and the reaction control system (RCS) of the Space Shuttle orbiter both use NTO/MMH. IRFNA/UDMH is frequently employed in tactical missiles, such as the ones used by the US Army (1972) [12] Hydrazine is often used as a monopropellant in catalytic decomposition engines. In these engines, liquid fuel decomposes into hot gas in the presence of a catalyst. The decomposition of hydrazine produces temperatures of up to 1100 °C and an impulse lasting between 230 and 240 s. It is possible to decompose hydrazine into hydrogen and nitrogen or ammonia and nitrogen.

2.2. Solid propulsion

Solid-fuel rocket engines are the most fundamental kind of rocket engine. They consist of a steel casing containing a mixture of solid compounds (fuel and oxidizer) that burn fast and generate thrust by expelling hot gases via a nozzle.

When a solid propellant is discharged, it consumes the case from the center outward. The shape of the center channel determines the rate and pattern of the burn, enabling thrust to be adjusted. Solid propellant engines, unlike liquid-propellant engines, cannot be shut off. Once lit, they will burn until the propellant is exhausted [13]. There are two types of solid propellant families: homogenous and composite. Both are dense, stable at average temperatures, and simple to store. Simple base or double base homogeneous propellants are available. A basic base propellant consists of a single component that can both oxidize and reduce, often nitrocellulose. In double-base propellants, nitrocellulose and nitroglycerine are often mixed with a plasticizer. Typically, homogeneous propellants do not have particular impulses for longer than around 210 s. The fact that they do not generate traceable vapors is their primary benefit, making them suitable for tactical weaponry. Furthermore, they are commonly used for supplementary activities such as discarding waste components or separating a rocket stage [14, 15].

Modern composite propellants are composed of heterogeneous powders, or mixtures, that contain a crystalline or finely broken mineral salt as an oxidizer, such as ammonium perchlorate, which can make up anywhere from 60 to 90% of the propellant's mass. Aluminum is commonly used as fuel. A polymeric binder, like polyurethane or polybutadiene, serves to hold the propellant together and is also utilized as fuel. Sometimes, additional chemicals such as a catalyst to enhance the burning rate or other agents to simplify the production of the powder are added. The finished product looks and feels like a solid rubber eraser. Composite propellants are typically classified based on the type of polymeric binder used. The two most common binders are PBAN and HTPB. PBAN formulations offer slightly greater specific impulse, density, and burn rate than HTPB formulations with identical characteristics. However, PBAN propellant requires higher curing temperatures and is more difficult to mix and produce. The HTPB binder is more flexible and durable than the PBAN binder. Both PBAN and HTPB formulations produce propellants with high performance, excellent mechanical properties, and the ability to burn for extended periods. Solid propellant motors have broad applications. Small solids are commonly used to boost payloads to higher orbits or to power the final stage of a launch vehicle. Medium solids, like the Payload Assist Module (PAM) and the Inertial Upper Stage (IUS), provide spacecraft with additional thrust needed to enter geosynchronous orbit or travel along planetary paths. Strap-on solid propellant rockets are used on the Titan, Delta, and Space Shuttle launch vehicles to enhance the liftoff power. The Space Shuttle uses the largest solid rocket engines in the world. Each rocket has a thrust capacity of 14,680,000 newtons (3,300,000 pounds) and a propellant capacity of 500,000 kg (1,100,000 pounds) [14].

The following are some important properties to note:

- The theoretical maximum impulses assume 100% efficiency, but actual performance may be lower.
- Unless otherwise stated, all mixture ratios are optimized for the given operating pressures.
- The oxygen liquid/hydrogen liquid (LO_2/LH_2) and fluorine liquid/hydrogen liquid (LF_2/LH_2) mixture ratios are higher than optimal to enhance density impulse.
- The liquid fluorine and liquid oxygen (FLOx). Formulations of 70% liquid fluorine with liquid oxygen (FLOx-70).
- In cases where kerosene is mentioned, the calculations are based on n-dodecane.
- Solid propellant formulation (a) is made up of 68% ammonium perchlorate (AP), 18% Aluminum (Al), and 14% hydroxy-terminator polybutadiene (HTPB).

Solid propellant formulation (b) is made up of 70% AP, 16% Al, 12% polybutadiene acrylic acid acrylonitrile (PBAN), and 2% epoxy curing agent.

2.3. Hybrid propulsion

Hybrid propellant engines are a type of engine that falls between solid and liquid propellant engines. One of the components is solid, which is generally the fuel, and the other is liquid, which is usually the oxidizer. The liquid is injected into the solid, which also functions as the combustion chamber due to its fuel reservoir. The fundamental advantage of such engines is their superior performance. The latter is similar to solid propellant engines, except combustion may be controlled, stopped, and resumed. Due to the difficulty of applying this theory to a broad variety of thrusts, hybrid propellant engines are seldom manufactured [16].

3. Green propulsion systems

An import question here is why green propellants? Or, why green propulsion systems?

In this section, we are considering green alternatives of hydrazine as a liquid propellant and eco-friendly solid propellants for the replacement of the toxic solid propellant ammonium perchlorate (AP).

3.1. Substitution of hydrazine

Firstly, a hydrazine thruster (Fig. 2) has been shown to be a powerful monopropellant in terms of performance, with a theoretical specific impulse of 230 s [17].

Based on practical performance data, nonetheless, its toxicity is its main disadvantage. Despite its performance, the space community has recognized that the extra costs involved with managing hydrazine and its derivatives may outweigh the performance loss afforded by certain green propellant options in this area. The major characteristics of a green propellant are lower prices, less complexity, and fewer emissions. The ESA appears to have approved the following definition of a green propellant, which is followed by a more extensive explanation in [18]. In fact, "A green propellant refers to a type of fuel that has the ability to meet mission requirements while potentially having a reduced adverse impact, such as lower toxicity, on the environment or personnel who may come in contact with it". The space community has found a number of green propellants, including hydrogen peroxide. Others include HAN, ADN, HNF (ionic liquids), nitrous oxide, and, more broadly, electric propulsion. With an I_{sp} of 251 s, the performance of ADN outperforms that of hydrazine [18]. Gordon and McBride [19] constructed the NASA-CEA code, which was used to determine this number. It uses a pressure of 10 bars, chemical equilibrium frozen in the throat, and a 40:1 nozzle expansion ratio. However, compared to hydrazine systems, ADN has certain disadvantages, including greater propellant and thruster costs, a constrained supply chain (produced under license by a single European provider), and little flying history. However, compared to hydrazine, the high system cost is mitigated by considerable savings in ground handling of this low toxicity propellant, resulting in a cheaper system overall. Based on the parameters mentioned above, nitrous oxide has a theoretical performance of 198 s [20], estimated using the NASA CEA algorithm. Again, the lower system cost compensates for the lower performance. However, because of the high decomposition temperature of 1640 °C, finding or creating suitable catalysts has been difficult. As far as we know, nitrous oxide has never been used as a rocket propellant. NOFBXTM (a nitrous oxide fuel blend) is a nitrous oxide version with a stated I_{sp} of 325 s [18], but no flying history. Finally, electric propulsion has a particularly high specific impulse. Electric propulsion can take many forms, but they all function by creating an ionized gas that is propelled by an electric field. The I_{sp} of the QinetiQ T6 ion thruster is 4120 s at 143 mN [21]. They are, however, quite costly and demand a large quantity of electrical energy (4.5 kW), which has a considerable influence on spacecraft design. Because of its modest thrust, it must operate for long periods, making it unsuitable for maneuvers like debris avoidance.

Despite their individual advantages, the identified drawbacks of each of these green alternatives have rendered them unsuitable for further investigation. In a toxicity study, hydrazine, MMH, and UDMH were compared against hydrogen peroxide and several other propellants. One of the findings of this study was that there is no widely agreed technique of toxicity evaluation and that essential toxicity data are occasionally unavailable [22]. However, the qualitative rating of propellants according to their toxicity is more or less consistent throughout all of the examined toxicity studies used in this evaluation.

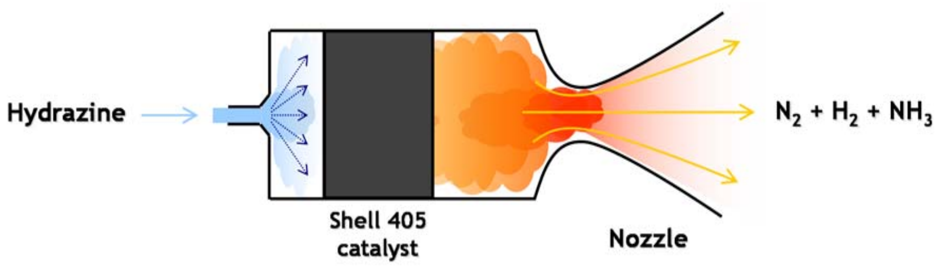

Fig. 2. Conventional hydrazine thruster.

Table 2
R-phrase categories for hydrogen peroxide, hydrazine and MMH22

R-values	H_2O_2	Hydrazine	MMH	UDMH
R23-28	0	1	1	1
R39	0	0	0	0
R48-68	0	0	0	0
R45-49	0	1	1	1
R46	0	0	0	0
R60-61	0	1	1	1
R50-53	0	0	0	0
R54-59	0	0	0	0
Total	0	3	3	3

The assessment incorporates data on toxicity under normal working settings as well as data on toxicity in the event of a spill or inadvertent contact with the propellant. The former is known as working toxicity data, whereas the latter is known as acute toxicity data. Working toxicity data are evaluated using R-phrases (Risk phrases), as established by European Union Directive [23]. The assessment employed the following categories with corresponding R-values:

- R23 through to R28: toxic in contact or if swallowed;
- R39: danger of very serious irreversible effects;
- R48: Danger of serious damage to health by prolonged exposure;
- R68: possible risk of irreversible effects;
- R45, R49: carcinogen categories 1 and 2;
- R46: genic damage;
- R60: may impair fertility;
- R61: may cause harm to the unborn child;
- R50-R53: toxic to aquatic organisms and environmental;
- R54-R57: toxic to flora, fauna, soil organisms and bees, respectively;
- R58 and R59: effects in the environment and dangerous for the ozone layer, respectively.

The findings for R-phrases for hydrogen peroxide, hydrazine, and hydrazine derivatives are summarized in Table 2. If a chemical has an R-phrase in one of the categories, it is assigned the number 1. If no R-phrases are supplied for that category, it gets a value of zero.

Some examples of candidates those can replace hydrazine for a reaction control system (RCS) are listed.

3.1.1. HAN system

Hydroxylammonium nitrate (HAN) is a liquid monopropellant. It is an ionic solution with the chemical formula $[NH_3OH^+][NO_3^-]$, falls within the grouping of energetic ionic liquids (EILS) that recently have received full attention due to their high density, smooth combustion characteristics and considerable $I_{sp} \times d$ values [24–29]. In addition, the HAN energetic compound possesses low toxicity and is easy to manipulate (Fig. 3).

Therefore, this eco-friendly chemical propellant has emerged as a substitute for hydrazine. Table 3 compares the toxicity of various chemical propellants to that of sodium chloride, a frequently used ionic salt.

Furthermore, HAN propellant offers several additional advantages over hydrazine, including high chemical stability, enhanced performance, and low development costs. These features make HAN particularly well-suited for small spacecraft and satellites [30–32]. Table 4 summarizes the performance comparison of various chemical propellants.

Table 3
Toxicity comparison of different propellants against sodium chloride [8, 9]

Propellants	LD_{50} oral (mg kg^{-1})	LD_{50} skin (mg kg^{-1})
Hydrazine	59	91
HNF	128	-
HAN	325	-
AF-M315E	550	-
LP1846 (XM46)	815	-
SHP163	500-2000	> 2000
ADN	832	-
NaCl	3750	-

Fig. 3. HAN-based propellant developed by Hosoya Corporation (Japan).

Table 4
Performance comparison between different propellants [29]

Parameter	Hydrazine	H_2O_2	HAN	SHP163
Density (g cm^{-3})	1.00	1.38	1.63	1.4
Freezing point (°C)	1.4	-0.43	-30 (75 wt.%)	-68
Theoretical I_{sp} (s)	233	144	257	276
Density × I_{sp} (×10^3 kg.s m^{-3})	233	198.72	470.3	386

Table 5
Formulations of HAN-based monopropellants [35–37]

Propellants	Ingredients (wt.-%)		
LP1845	HAN(63.2)	TEAN(20)	H_2O(16.8)
LP1846	HAN(60.8)	TEAN(19.2)	H_2O(20)
HAN269MEO15	HAN(69.7)	MeOH(14.8)	AN(0.6)-H_2O(14.9)
SHP163	HAN(73.6)	HEHN(44.5)	AN(3.9)-H_2O(6.2)
AF-M315E	HAN(44.5)	HEHN(44.5)	H_2O (11)

Table 5 provides a summary of various HAN-based propellant formulations that have been developed since the 1970s. These formulations are composed of different individual ingredients, and their respective weight percentages are included in the table. The US Air Force Research Laboratory developed one of the more recent formulations, known as AF-M315E. AF-M315E consists of HAN, 2-hydroxylethylhydrazinium nitrate (HEHN), and water, as previously described [33, 34].

This work presents a review of the development of HAN86-based liquid propellants for use in propulsion applications, building upon the background information provided earlier. A brief comprehensive and systematic information of the developments related to HAN is presented, including a discussion of various parameters that affect the decomposition behavior of the propellant. The review also highlights the current progress related to the use of HAN in propulsion systems, which have attracted the attention of researchers worldwide and are being considered for pilot-scale-up missions.

3.1.2. ADN system

In recent decades, ADN-based monopropellants have emerged as a new type of environmentally-friendly "green" propellant, possessing high specific impulse, safety, and low maintenance costs. These characteristics make them particularly suitable for use in low pollution space shuttle propulsion systems and transportation power systems [38, 39]. Development and research into ADN monopropellant formulations have been ongoing since 1997, with the Swedish Space Company (SSC) and FOI leading the efforts, as shown in Fig. 4.

Fig. 4. ADN-based liquid propellant developed by SSC and FOI Corporations.

On the basis of a selection process, different additives were retained as fuels for ADN-based propellants: glycerin, glycine, and methanol. The formulation performance of these monopropellants is shown in Table 6 [40–42].

It appears that a solution of LMP-101 has excellent explosive performance. However, poor thermal stability was shown. A solution of FLP-106 has good explosive performance and thermal stability and has been listed as a candidate formulation of monopropellants by the FOI. The thermal stability of LMP-103 is poor and too low to be considered a promising solution, but it can be improved by adding a loading of 3–5% of ammonia. The SCC selected an enhanced version of LMP-103, known as

Table 6
Formulation performance of ADN-based monopropellants and comparison of them with hydrazine and hydroxylamine nitrate (HAN) [43]

Propellants	Propellant compositions (%)			Specific impulse[a] (s)	Density[b] (g cm^{-3})
	ADN	Fuels	Water		
LMP-101	61.0	13.0 (Glycerin)	26.0	248	1.420
LMP-102	58.0	16.0 (Glycine)	26.0	214	1.390
LMP-103	63.4	11.2 (Methanol)	25.4	254	1.310
FLP-105	65.7	20.7 (F-5)[c]	13.6	261	1.405
FLP-106	64.6	11.5 (F-6)[c]	23.9	255	1.357
FLP-107	65.4	9.3 (F-7)[c]	25.3	256	1.351
Hydrazine	-	-	-	233	1.004
HAN	-	Methanol	26.0	234	1.320
HAN	-	Glycine	26.0	200	1.330

[a]The calculation condition of specific impulse is that the combustion chamber pressure P = 2 MPa and the expansion rate ε = 50. [b]The density test condition of ADN-based monopropellants is 25 °C.
[c]F-5, F-6 and F-7 refer to the code name of fuels.

LMP-103S, as the monopropellant candidate for use in satellite propulsion systems. LMP-103S was successfully employed in the Prisma satellites in 2010 [44–46]. The composition and properties of LMP-103S are detailed in Table 7.

Kim et al. [47] created two new monopropellants, KMP-4 and KMP-9, to enhance the energy level of the preheating temperature and vapor pressure of LMP-103S propellant. These new monopropellants contain ADN, water, and Tetraglyme fuel as their main components. It is worth noting that the KMP-9 propellant has a much lower adiabatic decomposition temperature compared to LMP-103S propellant, as shown in Table 8.

Table 7
Formulation composition and performance of LMP-103S [43]

Formulation composition and performances	LMP-103S
ADN (%)	63.0
Methanol (%)	18.4
Water (%)	14.0
Ammonia (%)	4.6
Freezing point (°C)	-7
Unstable transition temperature (°C)	165
Stable operating temperature (°C)	10-50
Density (g cm^{-3})	1.30
Specific impulse (s)	255
Density impulse (s)	332

Table 8
Formulation composition and performance of ADN/Tetraglyme monopropellants and comparison of them with LMP-103S propellant [43]

ADN (%)	63.0	60	60
Methanol (%)	18.4	-	-
Water (%)	14.0	30	30
Ammonia (%)	4.6	-	5
Tetraglyme (%)	-	10	5
Total (%)	100	100	100
Density (g cm^{-3})	1.30	1.32	1.29
Theoretical $I_{sp\ vac}$[a] (s)	255	249	247
Density.$I_{sp\ vac}$ (g s cm^{-3})	332	329	319
T_{ad}[b] (°C)	1645	1972	1604

[a]Chamber pressure is set at1.0 MPa in vacuum condition with frozen flow assumption, the nozzle
[b]T_{ad} refers to adiabatic decomposition temperature of the propellant.

Nevertheless, KMP-9 propellant has a lower specific impulse than LMP-103S propellant. However, the effective resolution of this issue would make KMP-9 the most favourable option.

3.1.3. H_2O_2 system

The use of hydrogen peroxide (H_2O_2) as monopropellant system has been tested in different space applications since 1938 [3, 48]. H_2O_2 is a type-clas-

Table 9
Physical and chemical properties of H_2O_2 propellant with different concentrations [3]

Properties	H_2O_2 Propellant Classification			
	Type 70 Grade ES	Type 85 Grade ES	Type 90 Grades ES-HP	Type 98 Grade HP
Concentration %	71.0-73.0	85.0-87.0	90.0-91.5	98.0-99.0
Density (g cm^{-3})	~1.29	~1.34	~1.40	~1.43
Freezing point (°C)	-40	-17	-12	-2
Boiling point (°C)	125	137	140	147

ES: Extra Stabilizers; HP: High Purity

sified molecule according to its high concentration in an aqueous solution and grade-classified according to the concentration of stabilizers, impurities and so on [49], as shown in Table 9.

H_2O_2 that contains over 85% as mass percentage is referred to as HTP or high-test peroxide. Rocket grade HTP, which is typical of 98% concentration [50], is utilized in space propulsion for low and medium thrust applications. Its high density (~1.43 g cm^{-3}) and nontoxic nature make it a compelling candidate for storage in propulsion systems. In monopropellant systems, it can catalytically decompose and reach temperatures up to 1222 K [50]. The performance of HTP 98% in monopropellant systems is about 20% less than that of hydrazine [51], with a specific impulse of approximately 186 s (at 1 MPa and 50:1 expansion conditions). Recently, H_2O_2 has exhibited excellent thrust pulses for reaction control of attitudes [52–61].

3.1.4. N_2O system

Nitrous oxide (N_2O) has been considered as a promising alternative for hydrazine for some time now [62–64]. After conducting a trade off study in the EUFBD program, Mayer et al. [1, 65] found that the combination of N_2O/ethanol was the preferred mixture. Ethanol, among hydrocarbons, demonstrated better ignitibility and moderate flame temperature, and it was found to be stable and miscible with nitrous oxide. The selected mixture had a saturated liquid density of 0.892 g cm^{-3} with a stoichiometric O/F ratio of 5.73 (~14.86% fuel). The critical point was 36.45 °C and 6.3 MPa, and the vapor pressure at the bubble point was 2.6 MPa. Theoretical specific impulse I_{sp} was reported to be 331 s, and the combustion temperature was 3093 K [65]. A 600 N thruster was used in a test campaign, and a specific impulse of 259 s was achieved [66]. However, some drawbacks were reported during the study, including the high combustion temperature requiring complex engine design and an active cooling system, expected incompatibility of the nitrous oxide fuel blend with titanium, the possibility of flammable vapors in the propellant tank, and low density at practical storage temperatures [65, 66]. Nonetheless, the advantages of nitrous oxide fuel blends, like most green monopropellants, include their nontoxic and noncarcinogenic nature, low freezing point, higher specific impulse than hydrazine, and the most prominent advantage,

Table 10
Performances and physical properties of the liquid NOx monopropellants [3, 67–70]

Propellant	Theoretical I_{sp} (s)	Density (g cm^{-3})	Density.I_{sp}	Tc (°C)
N_2O^a (liquid)	206	0.745	153.3	1640
Nitromethane^b	289	1.1371	328.6	2175.85
NOFBX$^{TM\ c}$	350	0.700	245	2926.85
$HyNO_x^d$ (Ethene)	303	0.879	266.3	2990.85
NO_x/Ethanol^e	331	0.892	295.3	2819.85

^{a}Pc = 0.3 MPa, ^{b}Pc = 1 MPa, ^{c}Pc = 0.7 MPa and stoichiometric O/F = 3, ^{d}Pc = 2.5 MPa and stoichiometric O/F = 6, ^{e}Pc = 1 MPa and stoichiometric O/F = 5.73

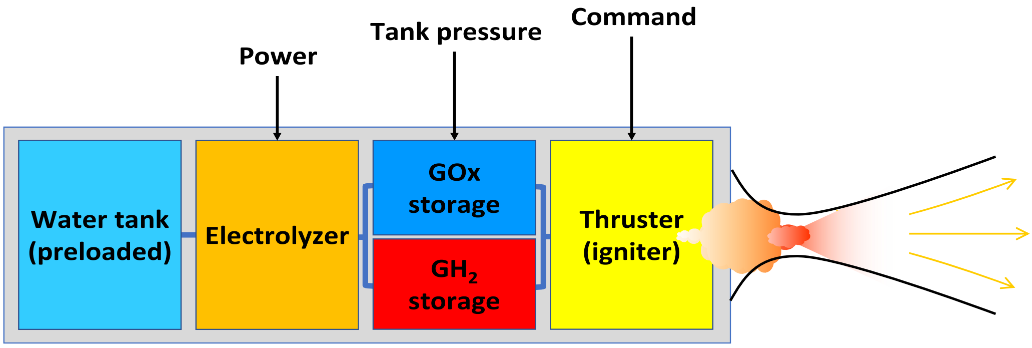

Fig. 5. Water propulsion by electrolysis [71].

their self-pressurization capabilities, allowing for a simple feed-system and tank-pressurization system design. The performance and properties of liquid NO_x monopropellants are presented in Table 10.

3.1.5. Water electrolysis system

Recently, water propulsion, also known as electrolRecently, water propulsion, also known as electrolysis propulsion, has gained attention due to its various benefits such as high performance (I_{sp} > 300 s), low cost, and use of a green propellant with no potential future limitations. This propulsion system can be preloaded at the manufacturer and high gas pressure is only generated when electrical power is applied. If combined with a fuel cell, it can even be used as an efficient battery [71, 72]. This system is based on using water as a stored propellant that is decomposed into gaseous oxygen and hydrogen via an electrolyzer in orbit. The resulting gases are then combusted exothermically to generate thrust. It is semi-electric propulsion where the propellant is generated over a longer period with low power and is then used during a short boost. The operational sequence of the system includes switching on the water feed from the water tank into the electrolyzer, producing GOx and GH_2 via electric power at high pressure, storing the propellants in gas tanks for later use, and utilizing the gases either in the thrusters to generate thrust or by a fuel cell to generate electric power when needed. Once the gas tanks are empty, the process starts again with the water feed being switched on.

The following figure shows the generic system layout and the operational sequence.

Several water electrolysis propulsion systems have been created specifically for CubeSats. Cornell University and Tethers Unlimited are among the institutions that have successfully designed and manufactured such systems [73–75]. In January 2021, NASA launched the CubeSat PTD-1 to demonstrate the water electrolysis propulsion technology. PTD-1 is the world's first space vehicle equipped with a water electrolysis system, and actual operational data have been obtained and presented [76, 77].

To conclude, the Table 11 represented comparative characteristics of green propellants such as HAN, ADN, H_2O_2 and N_2O.

In this Table, the green propellant systems have promising characteristics due to their high specific

Table 11
Comparative properties of green propellant used for reaction control systems (RCS)

Parameters	Propellant currently in use	HAN-based propellant		ADN-based propellant	H_2O_2 propellant	N_2O propellant (liquid)
	Hydrazine	SHP163	AF-M315E	LMP-103S		
Freezing point (°C)	2	←30	-22	-6	-7	n/a
Density (g cm^{-3})	1.0	1.4	1.5	1.4	1.3	0.7
Theoretical specific impulse (s)	239	276	266	182	255	206
Density specific impulse (g cm^{-3} s)	241	390	390	256	332	153
Adiabatic flame temperature (K)	1183	2166	2166	1154	2054	1640

impulses and the density specific impulses compared to hydrazine. This review paper articulated the propellant solutions that we can apply for reel mission of satellite controlling.

3.2. Substitution of ammonium perchlorate

Rocket propulsion applications, including missiles and rocket launchers, often utilize ammonium perchlorate (AP) based propellants as their oxidizing agent. These propellants offer numerous benefits, such as good specific impulses, high reliability, and a straightforward manufacturing process [78]. Consequently, AP based propellants are frequently utilized in booster applications such as space exploration rockets, aircraft ejection systems, and international ballistic missiles. The manufacturing process for solid propellants involves various considerations, including ingredient selection and production steps. According to Holmes et al. [79], at least ten different ingredients are necessary for creating a solid propellant. Additionally, Ramohalli et al. [80] reported that the production of propellants requires at least 30 distinct steps. However, the toxicity and the cost issues of AP are not negligible, hence its substitution with different eco-friendly solid propellants is necessary. In this review, we consider the development of two so-called green solid propellants: ADN and AlH_3 with good performances and excellent combustion characteristics.

3.2.1. ADN system

Ammonium dinitramide (ADN) is a white solid salt consisting of the ammonia cation (NH_4^+) and the dinitramide anion [$N(NO_2)_2^-$]. This energetic compound has a high oxygen balance of 25.8%, a melting point of 93 °C, and an onset decomposition temperature of 150 °C [81]. ADN is highly hygroscopic and readily soluble in water and other polar solvents but is barely soluble in non-polar solvents. At 25.0 °C, the critical relative humidity for ADN is 55.2%. The solid-state density of ADN is 1.81 g cm^{-3} [81–86]. ADN exhibits an endothermic melting peak at 91–93 °C, followed by an exothermic decomposition in the range of 150–210 °C, and one endothermic decomposition of ammonium nitrate (AN) in-situ formed during the decomposition of ADN. The TGA curve of ADN indicates a single-stage decomposition with 100% mass loss at 220 °C. ADN has an oxygen balance of 25.8%, which is lower than that of AP at 34%. However, its high heat of formation (-125.3 kJ mol^{-1}) compensates for this disadvantage when compared to AP (-283.1 kJ mol^{-1}). The density of ADN crystals is 1.885 g cm^{-3}, which is lower than that of AP at 1.950 g cm^{-3}. Nevertheless, the hygroscopicity and impact sensitivity of ADN are higher than those of AP [87]. As a water-soluble ionic salt, UV-Visible spectrometry is an effective method for analyzing ADN crystals. Two peaks at 214 and 284 nm characterize the UV spectrum of ADN [88]. Although ADN is a newly discovered oxidizer, it exhibits incompatibility issues with isocyanates (a cross-linking agent used for HTPB-based propellants) [89]. ADN/HTPB-based propellants can be produced; however, their energetics are still low, and their mechanical properties are poor. Therefore, consistent efforts are being made within the rocket science community to select suitable fuel binders and plasticizers to be used with ADN to achieve maximum energy. Some of the fuel binders used with ADN are HTPB [90–94], Polycaprolactone (PCL) [95, 96], PEG-PPG [91], GAP [97–101], Desmophen [100], poly-BAMO [102], poly-NIMMO [80], PGN [102, 103], and aluminum hydride (AlH_3) [104].

3.2.2. AlH_3 system

It is important to note that aluminum hydride (AlH_3) is very sensitive to the surrounding oxygen and moisture, which makes it unstable and leads to its spontaneous decomposition into aluminum and dihydrogen at standard conditions of thermodynamics (Eq. 1). The decomposition process of AlH_3 exhibits a sigmoidal-shaped TG curve, which can be divided into three parts: an induction period (I), an acceleratory period (II), and a decay period (III) [105]. The induction period is the onset point of the decomposition, corresponding to the breakup of the surface layer and the nucleation of aluminum. The acceleratory period is caused by a quick hydrogen-releasing process, which contributes to the growth of the aluminum phase in two and three dimensions [106]. The decay period is the end of the decomposition when the concentration of the hydride phase is limited, and the growing aluminum particles begin to overlap.

To modify the decomposition rate of AlH_3, the nucleation of aluminum is the key step that can be accelerated or decelerated. Therefore, controlling the time of the induction period can control the decomposition rate [107]. The decomposition pathways for various polymorphs of AlH_3 are slightly different. The less stable β- and γ-AlH_3

may undergo an exothermic transition process to α phase during the decomposition reaction. For instance, at elevated temperatures (> 100 °C), β- and γ-phase are generally transformed to the more stable α-phase (Eq. 2), while there is a direct decomposition into Al and H_2 at lower temperatures (≤ 100 °C) [108] (Eq. 3). Additionally, the decomposition mechanisms of γ-AlH_3 particles at the outer layer and the inner part have been developed [109, 110].

$$\alpha\text{-}AlH_3 \rightarrow Al + 3/2\ H_2 \quad \text{(Eq. 1)}$$

$$\beta/\gamma\text{-}AlH_3 \rightarrow \alpha\text{-}AlH_3 \rightarrow Al + 3/2\ H_2 \quad \text{(Eq. 2)}$$

$$\beta/\gamma\text{-}AlH_3 \rightarrow Al + 3/2\ H_2 \quad \text{(Eq. 3)}$$

When AlH_3 is exposed to an oxidative atmosphere, the thermal reaction becomes more complicated than under inert atmospheric conditions. The thermal reaction can be divided into three stages: the dehydrogenation and passivation stage, the primary oxidation stage, and the secondary oxidation stage [111].

Finally, we can conclude that green propellants are a class of rocket propellants that are environmentally friendly and have a lower impact on the environment compared to traditional propellants. They have several applications in various fields, including space exploration, military, and commercial aviation. Some of the uses and applications of green propellants are:

• Space exploration: green propellants can be used in space exploration missions, including satellite launches and interplanetary missions. They offer higher performance and lower toxicity compared to traditional propellants, reducing the risk of contamination in space.

• Military applications: green propellants can be used in missile systems, providing safer handling and transportation, and reducing the environmental impact of missile launches.

• Commercial aviation: green propellants can be used in commercial aviation, reducing the carbon emissions and environmental impact of air travel. They can also improve the fuel efficiency and range of aircraft, leading to significant cost savings for airlines.

• Small-scale applications: green propellants can be used in small-scale applications such as model rockets and amateur rocketry. They offer a safer and more environmentally friendly alternative to traditional propellants, making rocketry more accessible to hobbyists.

Overall, the use and application of green propellants offer a significant reduction in the environmental impact of rocket launches, space exploration, military, and commercial aviation. As technology continues to advance, we can expect to see wider adoption of green propellants in various industries.

4. Conclusion

In this review, we summarized the current state of research and development in green rocket propellants. It is indeed true that traditional rocket propellants, such as hydrazine, are highly toxic and pose risks to human health and the environment. As a result, researchers have been exploring alternative propellants that are more environmentally friendly, safer to handle and offer comparable or better performance.

Solid rocket propellants, such as ADN and AlH_3, have shown promise as green alternatives. ADN, for example, is a low-sensitivity, high-performance solid propellant that has been extensively studied in recent years. Similarly, AlH_3 is a highly energetic propellant that offers high performance and reduced environmental impact compared to traditional solid propellants.

For the reaction control system of satellites, several green propellants have also been developed, including HAN, H_2O_2, N_2O and water electrolysis. These propellants offer several advantages over traditional systems, including improved safety, reduced environmental impact, and better performance.

Overall, the development of green rocket propellants is an exciting area of research, and it is likely that we will see more innovations in this field in the years to come. These developments will not only improve the safety and environmental impact of space missions but also will also enable new missions and exploration opportunities that were previously impossible.

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