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Nitrous Oxide Based Green Propellants: Fuel Selection and Miscibility Analysis Method

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Abstract

Over the past 25 years, the search for safe, high-performance spacecraft propellants has highlighted nitrous oxide (N_2O) as one promising alternative to the toxic but efficient and state-of-the-art hydrazine-based propellants. Monopropellant N_2O offers key advantages such as low toxicity, self-pressurization and high specific impulse but requires high temperatures for its decomposition to start and can exhibit low combustion efficiency. This study explores premixed N_2O-fuel blends as potential monopropellants with the aim of lowering ignition temperatures while ensuring efficiency and safety. Therefore, the focus is on blends close to the Lower Explosion Limit (LEL), where small amounts of fuels are added to N_2O. To define and characterize such mixtures, a preliminary fuel selection process is performed and suitable candidates for blending with N_2O are identified on defined criteria. The selection results in compounds from the family of the hydrocarbons, alcohols, hydrides and silanes. In addition, a test-bench has been developed to evaluate the miscibility of these mixtures to ensure homogeneous blending and prevent phase separation. The results can provide insights into assessing the range of mixture ratios below the explosion limit that yields functional blends.

1. Introduction

Since the beginning of space exploration, combinations of propellants used for spacecrafts have been studied with the goal of high performance, low cost, storage capability, commercial availability and mission success. With regard to propulsion systems, since the 1950s, hydrazine and its derivatives (such as monomethylhydrazine, MMH and asymmetric dimethylhydrazine, UDMH) have been one of the most widely used propellant species. In monopropellant systems, hydrazine (N_2H_4) is preferred, while in bipropellant systems MMH is employed in combination with nitrogen tetroxide (NTO). Their extensive utilization over the decades is especially due to their high-performance characteristics. However, these propellants are highly toxic, corrosive and carcinogenic compounds[1]. In addition, it is a family of expensive chemical substances. For this reasons, in recent decades, there has been a focus on studying more environmentally friendly propellants, which pose less hazard to humans and do not undergo significant performance losses, while also being more cost-effective[2]. Fuel alternatives are categorized based on the type of propulsion system they support. Monopropellant systems, primarily used in satellite and orbital propulsion, play a crucial role in space operations by enabling precise maneuvers such as attitude control and small trajectory adjustments. These maneuvers help maintain the spacecraft's position and orientation throughout its mission. Among the most promising alternatives for monopropellant systems are propellants based on ammonium dinitramide (ADN) and hydroxylammonium nitrate (HAN), as well as hydrogen peroxide (H_2O_2), nitromethane (CH_3NO_2) and nitrous oxide (N_2O).

ADN and HAN are ionic compounds used in water-based solutions to create liquid monopropellants. Two notable ADN-based propellants are LMP-103S and FLP-106, both of which exhibit higher density and energy content compared to hydrazine[3,4]. The development and validation of LMP-103S have been reported extensively and was repeatedly successful in test flights[5]. Similarly, the HAN-based propellants SHP-163 and ASCENT (formerly AF-M315E) have been widely adopted due to their energetic performance and have been flight-proven[6]. Hydrogen peroxide (H_2O_2) is used as propellant in highly concentrated solutions (85 to 98%, known as high-test peroxide, HTP). It is a high density liquid and can decompose exothermically in water, producing relatively high performance (vacuum specific impulse of 192 s) at low cost[7]. Nitromethane is used as a solvent in various industrial applications. It has a high density and can serve as base for energetic gel propellants. Extensive studies have been conducted to characterize nitromethane-based propellants and optimize their ignition processes[8,9].

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Nitrous oxide is an odorless, colorless and non-flammable gas under standard conditions. It has been commonly used in the medical field as anesthetic due to its ability to induce euphoria, which is why it often referred to as "laughing gas"[10]. Beyond healthcare, N_2O finds application in other industries. For example, it is utilized in the dairy industry as a food processing propellant and it can also be injected into car engines to enhance performance by increasing the oxygen supply during combustion[11]. In the early 2000s, nitrous oxide gained interest as a potential green propellant for space propulsion, owing to its lower toxicity compared to traditional propellants (Globally Harmonized System of Classification and Labeling of Chemicals - GHS - class 5[12]). It is used as an oxidizing agent in bipropellant systems, as a monopropellant and as a gas in cold gas thrusters, for whose application it is one of the densest propellants[13]. It also offers the possibility of multimode configurations[14], i.e. combinations of all three of the above options, which provide numerous benefits for small satellites[15]. Regardless of its many advantages, N_2O has a lower density and specific impulse if compared to hydrazine. Furthermore, it is important to note that N_2O is a potent greenhouse gas, ranking third in terms of global emissions, after carbon dioxide (CO_2) and methane (CH_4). Agricultural activities, particularly the use of nitrogen-based fertilizers, are the primary sources of emission[16]. Despite its lower concentration, N_2O is about 310 times more effective than CO_2 at trapping heat in the atmosphere[17,18], amplifying its effect. However, when N_2O undergoes complete combustion, as in its use as a propellant for space applications, it decomposes into less harmful components, which reduce its environmental impact. This and its non-toxic properties are the reason why nitrous oxide is referred to as a "green" propellant.

1.1 Advantages and Challenges of N_2O as a Monopropellant

Nitrous oxide is a versatile propellant that can be used as a monopropellant in self-pressurized systems. Monopropellant systems offer several advantages, including reduced mass and complexity, as well as increased reliability. N_2O is notable for its high specific impulse (around 204 s with 10 bar chamber pressure and expansion ratio $\epsilon = 200$, as shown in Fig. 3) and inherent stability, making it easy to store and transport in tanks. With a vapor pressure of approximately 51 bar at 20 °C, N_2O enables a self-pressurized propulsion system[19]. As liquid N_2O is consumed, the remaining liquid partially vaporizes due to the increasing free volume within the tank, which results in the pressurization of the remaining N_2O. At this point, both the liquid and gaseous phases coexist in a saturated state. This self-pressurization feature further simplifies the system and reduces its mass by eliminating the need for an external pressurization source. In addition, when exposed to a heated catalyst, N_2O can rapidly decompose into nitrogen and oxygen, releasing significant thermal energy at high temperatures[20]. This energy can be harnessed to generate thrust within a thrust chamber. However, despite its advantages, monopropellant N_2O using catalytic decomposition faces several challenges due to the high temperatures required for the reaction to occur. N_2O begins to decompose between 520 and 850 °C[19,21]. Catalysts are used to reduce the activation energy but they still require heaters to reach the necessary temperature, leading to long preheating times and high thermal loads during operation. Additionally, these systems may suffer from low combustion efficiencies, which could be due to a decrease in the temperature of the catalyst during tests or a reduction in the catalyst's reactivity caused by consumption and erosion of its surface[22]. A further concern is the potential risk of backfiring into the tank system, particularly when N_2O is stored in a premixed blend with other fuels[23,24].

Over the past few decades, N_2O has been extensively researched and characterized as a monopropellant. In particular, NOx monopropellants have emerged as a promising category of green propellants[25] and nitrous oxide fuel blends (NOFB) have gained significant attention. In these systems, N_2O is premixed with one or more substances in liquid form in a single tank, and it is later used as a monopropellant, either in its gaseous or liquid state. The primary advantage of this approach is its ability to combine the simplicity of a monopropellant system with the performance characteristics of a bipropellant system. Numerous studies and patents have been published on this type of application and various propellant combinations have been tested. For example, Firestar/ISPS has developed an N_2O-based monopropellant that incorporates various organic fuels called NOFBX[23]. Additionally, the Defense Advanced Research Projects Agency (DARPA) has conducted research into mixtures of N_2O with acetylene (C_2H_2), patented as NA-7[26], and propane (C_3H_8)[27]. Since 2014, the German Aerospace Center (DLR) has been conducting research on blends of nitrous oxide with various hydrocarbons, namely ethane (C_2H_6) and ethene (C_2H_4), as part of its "Future Fuels" project[28,29]. This initiative aims to characterize these propellant mixtures, test ignition methods, analyze the thermal loading and evaluate flashback arresters. The results have demonstrated the potential of NOFBs as effective monopropellants for space propulsion. It can therefore be concluded from experience that certain fuels, when blended with N_2O, can enhance its properties and improve its performance as a monopropellant. Of particular interest is the investigation of blends based on N_2O, where a small amount of fuel is added to N_2O, at mixture ratios below the explosion limit. This can ensure safe operation while providing the benefits of a pre-mixed monopropellant.

Therefore, this study aims to further investigate the performance of nitrous oxide fuel blends. Specifically, it explores various propellant combinations with N_2O that may address the challenges associated with monopropellant

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N_2O and serve as viable alternatives for space propulsion. The propellants are chosen based on a set of predefined criteria and are analyzed both as individual chemical species and as a mixture. The development of a test bench designed for the miscibility analysis of these propellants is also described. Since the primary objective is to achieve a homogeneous mixture below the explosion limit (i.e. high mixture ratios) for safe operation, the investigation of the miscibility range of these substances is required.

2. Fundamental Concepts on N_2O Decomposition

When used as a monopropellant, nitrous oxide decomposition can be activated by various systems. The thermal energy is generated through the decomposition of N_2O into nitrogen (N_2) and oxygen (O_2), as described by the following reaction[30]:

$$N_2O \longrightarrow N_2 + \frac{1}{2}O_2, \quad \Delta H = -81.6\frac{kJ}{mol} \tag{1}$$

For the decomposition to start, high temperatures are required[23], which are difficult to achieve and can pose risks to the propulsion system's components. The most common method to accelerate this reaction is to pass N_2O through a catalyst. The decomposition of N_2O is a relatively clean reaction, leaving no residues on the catalyst, unlike many organic reactions[31]. This allows for easy monitoring of catalyst degradation. Several metals and metal oxides serve as active catalysts, often achieving N_2O decomposition at lower temperatures compared to gas-phase homogeneous reactions[31]. Especially noble metals such as ruthenium, rhodium, palladium, platinum, gold and iridium exhibit high catalytic activity and are resistant to impurity gases compared to other catalysts[32]. Rhodium-based catalysts are among the most effective for N_2O decomposition[31], capable of reducing the decomposition temperature to around 250 °C[33]. The support material of the catalyst significantly influences the catalyst's activity. Al_2O_3 is widely used as support material in many industrial applications due to its textural properties and surface chemical composition[34]. A comparative study of different catalysts on this support material[35] revealed that the catalytic activity decreases from rhodium to palladium to platinum. Further data showed that rhodium is more active than iridium, which has a decomposition temperature of 400 - 450 °C[31].

While catalysts can lower the decomposition temperature of nitrous oxide, achieving temperatures below approximately 250 °C has not been recorded. However, when comparing different propellant blends using the same catalyst, some fuels may offer the advantage of reducing this temperature further and facilitating N_2O decomposition.

3. Fuel Selection for N_2O Based Blends

It is important to examine the propellants that can be used in fuel blends with N_2O as a premixed monopropellant. A thorough literature review has been conducted to identify potential candidates, with a particular focus on chemical compounds that have shown promising results when interacting with N_2O. The selected substances:

- must be liquid or gaseous at standard conditions in order to allow miscibility with N_2O;
- must have low toxicity, as must the products of the reaction;
- should start a reaction with N_2O at lower temperatures than those required for the monopropellant N_2O;
- should produce high thermal energy when reacting with N_2O (enthalpy of formation, specific impulse);
- must not react suddenly with N_2O, as the fuels can potentially be stored together for a long time;
- must be relatively easy to produce and available on the market.

3.1 Most Promising Propellants

First and foremost, the hydrocarbon family is considered in the selection due to its extensive analysis background at DLR[36], both as bipropellant systems with N_2O and as premixed blends. Specifically, the compounds ethane (C_2H_6) and ethene (C_2H_4) are selected. Other light hydrocarbons were included in the list of candidates for the mixtures, including methane (CH_4), ethyne (C_2H_2), propane (C_3H_8), propene (C_3H_6), propyne (C_3H_4) and butane (C_4H_{10}). Each of these hydrocarbons offers unique advantages and challenges. Alkanes, alkenes and alkynes are all viable fuel options, but their performance tends to decrease while their density increases with longer chain lengths[37]. Heavier hydrocarbons have longer chains and higher molecular mass, which is not necessarily associated with an increase in performance. They generally have a higher boiling point, which makes them more difficult to vaporize. They have a higher density, lower energy density and higher viscosity, which can lead to poorer combustion efficiency. They are also generally

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more hazardous to humans and the environment[38]. For these reasons, they were discarded in this selection. In addition to hydrocarbons, ammonia (NH_3) is added to the selected propellants, as well as alcohols. Methanol (CH_3OH) and ethanol (C_2H_5OH) were considered from this family. Short-chain alcohols exhibit higher volatility and lower freezing points compared to their long-chain counterparts, making them easier to handle and store in space. Additionally, short-chain alcohols tend to have better combustion properties[39]. Long-chain alcohols, are less suitable for mixtures with N_2O because, due to their higher molecular weight, tend to be very viscous liquids with low vapor pressures, making them difficult to mix effectively with N_2O. At last, the chemical family of the silanes is added to the selected propellants.

Ethane and Ethene are both non-toxic substances that are easily available on the market. They have a similar vapor pressure to N_2O, which makes it easier to create a self-pressurized system. Mixtures of nitrous oxide and ethene can also achieve high specific impulse (I_{sp}) values. Ethane on the other hand offers a higher density between the two (1.356 kg/m^3 at 20 °C, 1 bar), which is advantageous for fuel storage.

Methane, being gaseous at standard conditions, has a much lower boiling point than N_2O. This makes a premixed liquid monopropellant combination of the two unsuitable[31].In fact, the premixed N_2O-based monopropellant would be stored as liquid in the spacecraft's tank, offering density advantages and the possibility of controlled release by allowing the propellant to vaporize directly when injected into the thruster. Nonetheless, combinations of N_2O-CH_4 are feasible and were considered during the selection process. This is especially due to the fact that N_2O is a more potent oxidizer than O_2 that reacts quickly with CH_4, lowering its activation energy required for the decomposition reaction[31].

Ethyne, also known as acetylene, has been extensively investigated in mixtures with N_2O and developed in the patented propellant NA-7. NA-7 was used in the ALASA program[26], developed in 2012 with the goal of providing a new, cost-effective and rapid launch vehicle for small satellites, enhancing the flexibility of space access. However, despite its potential, the ALASA program was eventually canceled in 2017 due to safety concerns surrounding the propellant. NA-7 exploded in two ground tests, highlighting the risks associated with acetylene, which is very reactive and unstable. At high pressures and temperatures, C_2H_2 can decompose spontaneously, releasing large amount of energy, which can lead to violent explosions.

Propane has been extensively researched in combination with nitrous oxide by DARPA[27] and Allied Aerospace Industries. One of the key advantages of propane is its lower freezing point compared to hydrazine, which helps prevent line freezing in space[40]. Furthermore, the N_2O-C_3H_8 propellant combination offers rocket performance comparable to MMH/NTO but with more benign characteristics, although it comes with a slightly lower storage density. However, it has a higher density than the other hydrocarbons.

Propene, an alkene with a carbon-carbon double bond, is more reactive than propane due to this unsaturated bond. This double bond also makes propene more easily ignitable than its corresponding alkane, enhancing its suitability as a fuel. Physically, propene shares similar properties with propane, including density and boiling point. The increased reactivity and ease of ignition make propene an ideal choice as a fuel. The N_2O-C_3H_6 propellant combination has been successfully demonstrated in practice, with companies like Dawn Aerospace launching a bipropellant system into orbit in 2021[41]. Moreover, a mixture of 90 % C_3H_8 and 10 % C_3H_6 as fuel to be premixed with N_2O has shown to reduce the light-off temperature by about 25 %[42] in comparison to C_3H_8-N_2O blends.

Propyne, also known as methylacetylene, is an alkyne with a carobon-carbon triple bond, which makes the molecule more reactive than its alkane and alkene counterpart. It has gained interest as a space propellant, despite its low density and high volatility, since propargylic derivatives (compounds characterized by an alkyl group derived from propyne) with the inclusion of heteroatoms are considered good candidates for rocket propulsion[43].

Butane has been effectively utilized as a propellant in small satellite propulsion systems due to its high storage density and low storage pressure[44]. For instance, it has been successfully employed in Surrey's SNAP-1 spacecraft[45]. Additionally, N_2O serves as a valuable reagent for the oxidative dehydrogenation of butane over a catalyst, offering superior selectivity compared to O_2[46]. This process involves a series of hydrogen abstraction steps, where N_2O acts as a soft oxidant. This minimizes total oxidation reactions, enhances selectivity towards partially oxidized products and increases process safety by reducing the risk of explosions.

Ammonia has promising properties regarding the decomposition of N_2O. This happens by lowering the necessary activation energy needed to start the decomposition reaction[47]. NH_3 has also a high density and a good thermal conductivity. On the downside, its specific impulse is one of the lowest among the selected propellants, especially at high mixture ratios, as shown in Fig. 3. Its burning velocity is much lower than hydrocarbons and its minimum ignition energy is higher. A potential advantage is using propane as additive in NH_3-N_2O blends to improve reactivity and reduce ignition energy[48].

Methanol and Ethanol are light aliphatic alcohols that can act as hydrogen donors, enabling the reduction of N_2O

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into N_2 and H_2O in the presence of an appropriate catalyst, such as a ruthenium-based complex[49]. During this process, the alcohols undergo oxidative dehydrogenation, forming the corresponding carboxylate derivatives. Methanol-N_2O can achieve high decomposition yields with this catalyst system. Under standard conditions, both alcohols are in the liquid state, meaning that the premixed blend needs to be delivered to the catalytic bed in its liquid form. Ethanol is a promising fuel for mixtures with N_2O and it has already been used in bipropellant systems[50,51]. Its density is similar to that of N_2O at room temperature and its temperature-dependent variation is considerably lower than N_2O. Ethanol boasts a low freezing point and high energy density, making it a good candidate for space propulsion applications. In addition, TNO, NAMMO Westcott and Bradford Engineering published a report[52] in 2018 outlining their selection of the most promising propellants for use in NOFB. The report evaluates various substances on the basis of several key criteria, including physical and chemical properties, such as toxicity, autoignition temperature and gravimetric specific impulse compared to hydrazine. Each substance was scored according to the importance of these parameters. The analysis concluded that ethanol was the most promising propellant for NOFB mixtures, particularly excelling in gravimetric specific impulse and saturated liquid density. The study further characterized the mixture in terms of miscibility and hot flows, where the propellants were fed as separate streams. Successful firings demonstrated high combustion efficiency and minimal pressure fluctuations, underscoring the potential of this propellant blend. Interestingly, in this study, the carbohydrates received the lowest score.

Silanes are compounds composed of silicon and hydrogen atoms, generally less stable and more reactive than their corresponding hydrocarbons. This is due to the fact that the Si-Si bond is less strong than the C-C bond, making the atomic chain of silicon atoms more susceptible to breakage. Furthermore, the Si-H bonds are highly reactive and have the potential to facilitate the activation of N_2O. Additionally, the Si-H bond is less strong than the C-H bond[53]. This explains the progressive and rapid decrease in stability of silanes as the chain of silicon atoms lengthens. The mixture of monosilane (SiH_4) with N_2O, for example, is stable at ambient temperature and atmospheric pressure[54] and SiH_4 is characterized by a higher Isp compared to more toxic traditional propellants[55]. Moreover, SiH_4-N_2O-Ar mixtures have been investigated and have shown higher laminar burning rate, explosion peak pressure and pressure rise coefficient than N_2O-H_2 and hydrocarbon based mixtures[56]. The silane family has been studied with the goal of decomposing N_2O accumulated in the atmosphere, since they are excellent at degrading N_2O under mild conditions through processes of hydrogenation and hydrosilylation[57]. Although a catalyst is still needed for this reaction to occur, the promising reaction temperature has spurred ongoing research into various sustainable catalysts[58,59]. Different silanes can be considered for blends with N_2O but it is important to note that some can spontaneously react with air, specifically with oxygen, and therefore require careful handling. While monosilane is gaseous at standard conditions and highly reactive, lighter liquid silanes are more stable and do not spontaneously ignite in air. However, the risk of forming explosive mixtures with air increases as the length of the silicon chain grows. Some disilanes have been analyzed in the framework of N_2O reduction; for example, hexamethyldisilane ($C_6H_{18}Si_2$) results in a 77% decomposition of N_2O at 20 °C under the catalyst CsF[58]. Furthermore, it has been noted that the addition of phenyl groups to silane compounds increases the amount of decomposed N_2O in a quicker time. However, some of these more complex silanes with phenyl groups are more expensive. As general consideration, it is worth noting that the reaction of silicon compounds with N_2O produces SiO_2, a sand-like powder that can act as an insulating thin film[60]. The properties of this film depend on the concentration of the substances, temperature, and pressure[61]. Therefore, there is a risk that a SiO_2 layer can coat the catalyst and lower its ability to accelerate the reaction. In addition, the release of this compound could pose risks to space applications, as it may damage sensitive equipment, including optical instruments and solar panels. However, if the quantity of the silicon compound is reduced, one can assume that the quantity of the products will also be moderate, even though a decrease in the silane concentration is connected to a decrease in the burning velocity of the mixture[55]. The ones chosen as promising propellants for blends with N_2O are monosilane (SiH_4), one silicon compound with methyl groups (hexamethyldisilane, $C_6H_{18}Si_2$), and one with methyl and phenyl groups (dimethylphenylsilane, $C_8H_{12}Si$[62]). The decision to choose these compounds is linked to the possibility of evaluating the impact of methyl and phenyl groups in the mixtures and to cost reasons. Ultimately, the propellants identified as suitable for blending with N_2O are as follows:

Table 1: Selected propellants for N_2O blends

Methane (CH_4)	Ethane (C_2H_6)	Ethene (C_2H_4)	Ethyne (C_2H_2)
Propane (C_3H_8)	Propene (C_3H_6)	Propyne (C_3H_4)	Butane (C_4H_{10})
Ammonia (NH_3)	Methanol (CH_3OH)	Ethanol (C_2H_5OH)	Monosilane (SiH_4)
Hexamethyldisilane ($C_6H_{18}Si_2$)	Dimethylphenylsilane ($C_8H_{12}Si$)		

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3.2 Selection Criteria

To assess the suitability of the selected propellants for mixtures with N_2O, criteria are established to evaluate them based on their physical and chemical properties, as well as those of their blends with N_2O. Key characteristics sought in the selected fuels are:

1. **Phase State**: In order for the propellants to be miscible with N_2O, they need to be either liquid at standard conditions or have a vapor pressure that allows for liquefaction within a practical pressure range (between room pressure and vapor pressure of N_2O). The phase state of most of the selected substances is gaseous at standard conditions (CH_3OH, C_2H_5OH, $C_6H_{18}Si_2$ and $C_8H_{12}Si$ are in the liquid state).

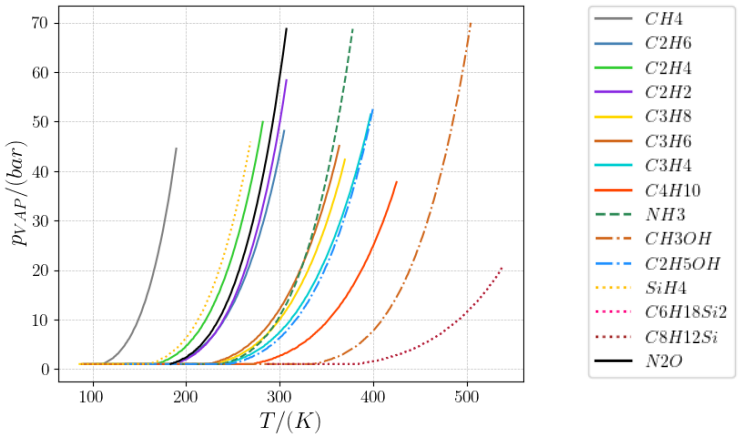

Figure 1: Vapor pressure (p_{VAP}) of the selected substances in relation to temperature obtained using the Thermo library in Python. Left of the curve is the liquid phase, right of the curve is the gas phase

   As shown in Fig 1, the liquid silanes exhibit the largest vapor pressure difference compared to N_2O, as well as methane and methanol, which could potentially complicate their mixing in the blend. In contrast, chemical species with vapor pressure closer to that of N_2O include ethane, ethene, acetylene and monosilane.

2. **Encouragement of N_2O Decomposition**: If the propellant can encourage N_2O decomposition, the required temperature for the reaction to start decreases, addressing one of the main challenges of monopropellant systems. From the literature research, the propellants presenting this advantage are ammonia[47] and the family of the silanes[57].

3. **Vapor Pressure of the Mixture**: The closer the vapor pressure of the fuels is to that of N_2O, the easier it is to create a self-pressurized system. A higher vapor pressure in the mixture results in more effective pressurization. For an ideal mixture adhering to the ideal gas law, Raoult's Law can be applied to compute the vapor pressure of the blend[63]. By using Eq. 2, where P^0 represents the vapor pressure of each chemical species and χ denotes the molar fraction of the components, the mixture's vapor pressure can be calculated for any mixture ratio.

$$P_{mixture} = \chi_{fuel} \cdot P^0_{fuel} + \chi_{ox} \cdot P^0_{ox} \tag{2}$$

   As the mixture approaches the LEL, the quantity of N_2O mixed with the fuel increases and the vapor pressure asymptotically converges to that of N_2O. Fuels with higher vapor pressure are monosilane and ethene. From this perspective, the least favorable is ammonia, as shown in Fig. 2. The figure excludes methane due to its significantly higher vapor pressure compared to the other fuels.

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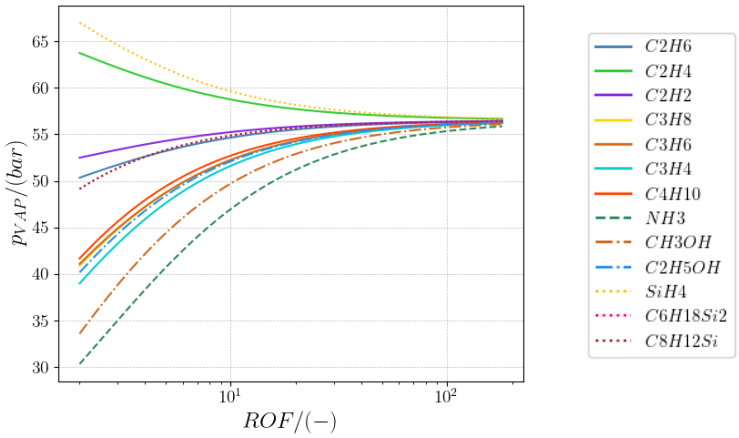

Figure 2: Vapor pressure of N_2O blends at 298 K and p_c = 10 bar, ϵ = 200

4. **Specific Impulse**: In space propulsion, specific impulse is a crucial parameter for determining the performance of a propellant mixture. Using the Chemical Equilibrium with Applications (CEA) program[64], it is possible to calculate the specific impulse and combustion temperature of the mixtures. As shown in Fig. 3 and Fig. 4, the highest I_{SP} and combustion temperature near the LEL is achieved for the hydrocarbons. Alcohols and ammonia have lower values, which translates in reduced performance outputs. In Fig. 3, Fig. 4 and Fig. 5, SiH_4 does not appear in the results because the CEA simulation is performed with a frozen equilibrium at throat. The reason for its missing is that the temperature gets 50 K lower than the temperature range of a condensed species. Because of that, the simulation is interrupted for expansion ratios greater than ϵ = 3. However, the results with ϵ = 3 show that the performance of SiH_4 is in the same range as the other silanes.

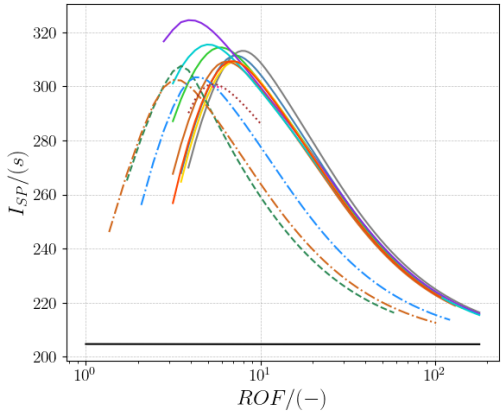

Figure 3: Specific impulse of N_2O blends at p_c = 10 bar, ϵ = 200 compared to monopropellant N_2O for various mixture ratios

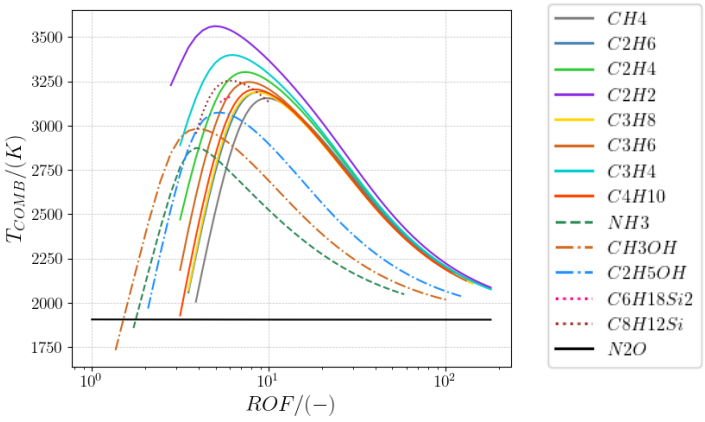

Figure 4: Combustion temperature (T_{COMB}) of N_2O blends at p_c = 10 bar, ϵ = 200 compared to monopropellant N_2O for various mixture ratios

5. **Mixture Density**: The density of the mixtures is important for storage purposes. A higher density allows for more propellant to be stored in the same volume. Based on the calculation of the mixture density in relation to the mixture ratio, as shown in Fig. 5, alcohols and ammonia blends exhibits the highest densities near the LEL.
6. **Thermal Conductivity**: The thermal conductivity of energetic materials is important for both safety and performance assessments. It influences how quickly heat is absorbed, which directly affects the reaction rate. High thermal conductivity is also beneficial for regenerative cooling processes. As shown in Fig. 6, methane has the highest thermal conductivity, whereas butane the lowest.

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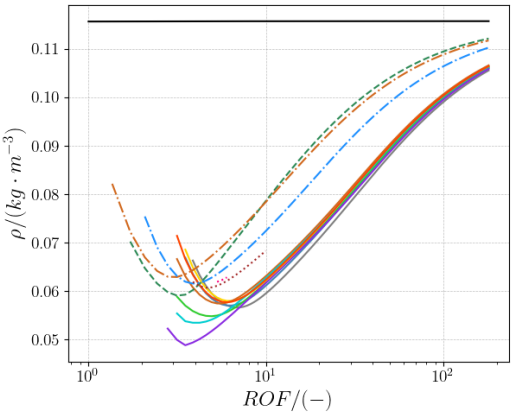

Figure 5: Density (ρ) of N_2O blends at p_c = 10 bar, ϵ = 200 for various mixture ratios

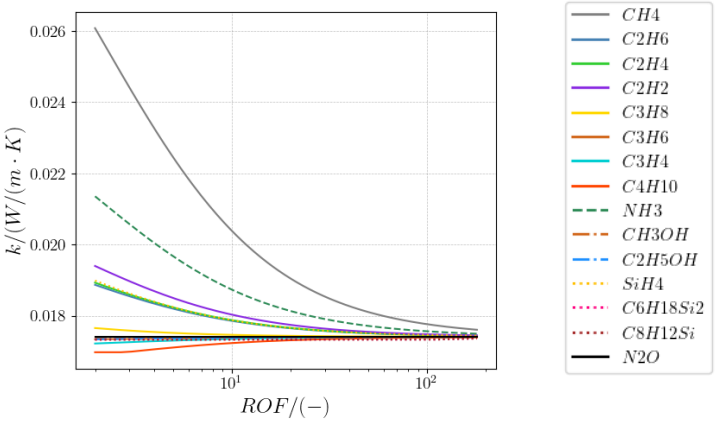

Figure 6: Thermal conductivity (k) of N_2O blends at 298 K and 10 bar for various mixture ratios

7. **Ignition Energy**: The ignition energy must be balanced; it should not be too low or too high. If the ignition energy is too low, the mixture may be easily ignitable, posing a safety risk. Conversely, if it is too high, the fuel may not readily ignite the blends, affecting performance. Acetylene has a very low ignition energy (0.017 mJ in air[65]), which contributes to its high reactivity. Through a calculation of the enthalpy of reaction, C_2H_2 proves also to lead to the most exothermic reaction among the mixtures. In contrast, ammonia requires a relatively high energy; however, its ignition characteristics can be altered by adding propane to the blend[66] (for example in a ratio 90% NH_3-10% C_3H_8). Silane, on the other hand, is expected to have a relatively low ignition energy[67].
8. **Burning Velocity**: A moderately high burning velocity is desirable to achieve fast and efficient energy release, enhancing the overall performance of the propellant mixture. Hydrocarbons typically exhibit high burning velocities (can reach 0.5 m/s[68]) due to the energy released from the C - H bonds and similarly do the silanes[56]. In contrast, ammonia has lower burning velocity (around 5 cm/s[69]).
9. **Stability**: Stable propellants are less prone to degradation over time and are less likely to react unpredictably with other substances, ensuring safer storage, handling and operation. Hydrocarbons are generally stable, although some, like methane, are highly flammable due to their low flash points and others, like acetylene, can form explosive mixtures with air. Information about the stability and incompatibilities of the various compounds can be found in online libraries[70,71]. In addition to hydrocarbons, methanol and ethanol are also stable, as well as monosilane, due to its low boiling point. However, it is highly reactive with oxygen in air, which makes its liquid derivatives (hexamethyldisilane and dimethylphenylsilane) more stable alternatives.
10. **Incompatibilities**: Propellants should not exhibit instability when in contact with components of the feeding system prior to their usage. This ensures that no premature or unintended reactions occur, maintaining the integrity and reliability of the propellant system. While most of the selected compounds are incompatible with strong oxidizing agents, ethyn is particularly challenging as it is incompatible with a wide range of metals, including copper, zinc, mercury, silver and brasses. Further incompatibilities with metals comprise propyne with copper alloys, ammonia with zinc, while also being corrosive to copper and ethanol with platinum. Assessing this is important because metals are often used as catalysts or as structural materials in propulsion systems.
11. **Market Availability**: Propellants that are readily accessible on the market and reasonably priced facilitate easier investigation and promote their adoption for space propulsion. Through an investigation of the market volume of the substances in the European Economic Area[72], it is found that methanol is the most widely traded, with an annual market volume between 10 and 100 million tonnes. Most of the selected hydrocarbons have good market availability, with the exception of acetylene. The market for silanes is also relatively niche.
12. **Other Criteria**: The results of the evaluation performed by TNO, NAMMO Westcott and Bradford Engineering[52] have been factored into the selection, since they identified their own most suitable fuel for NOFB based on diverse criteria. According to the findings, ethanol is presented as promising candidate for N_2O mixtures. Methanol and ammonia also score highly in the evaluation. In contrast, hydrocarbons perform the least favorably.

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3.3 Selection Results

All the criteria and their evaluation for each fuel is represented in the table below. The total score is calculated using the assigned weighting factors and the assumption of each plus or minus sign, with points added or subtracted accordingly.

Table 2: Evaluation of the selected propellants for N_2O blends

Criteria n.	1	2	3	4	5	6	7	8	9	10	11	12	Tot.
Weight	3	4	2	2	2	1	1	1	2	1	1	4	
CH_4	- -		- -	+ +	- -	+ +		+	-		+		-8
C_2H_6	+ +			+	-			+	+			-	5
C_2H_4	+ +		+	+				+	+		+	- -	6
C_2H_2	+ +			+	-	+	-	+	- -	- -		- -	-7
C_3H_8				+				+	+				5
C_3H_6				+				+	+		+	-	2
C_3H_4				+				+		-			2
C_4H_{10}	-			+		- -		+					-2
NH_3		+ +	-	- -	+ +	+	- -	- -		-	+	+	7
CH_3OH	-		-	-	+						+ +	+	1
C_2H_5OH					+				-	-	+	+ +	8
SiH_4	+	+ +	+ +				+	+	-		-		14
$C_6H_{18}Si_2$	- -	+ +						+	+		-		4
$C_8H_{12}Si$	- -	+ +						+	+		-		4

In conclusion, of the total selected propellants, the most suitable for monopropellant blends with N_2O are:

Table 3: Final results of the propellants selection for N_2O blends

Ethane	Ethene	Propane	Propane (90%) and propene (10%)
Ammonia	Ammonia (90%) and propane (10%)	Hexamethyldisilane	Dimethylphenylsilane

Monosilane, despite its promising properties for mixtures with N_2O, was not invesigated further due to availability issues at the time this study is conducted.

4. Miscibility Properties of the Blends

4.1 The Lower Explosion Limit

Since this study aims to evaluate N_2O-based mixtures below the explosion limit and at high mixture ratios, it is essential to determine the mixture ratio at which the Lower Explosion Limit (LEL) is reached for each blend. The lower explosion limit (often referred to the lower flammability limit (LFL) for practical purposes) is defined as the lowest concentration of a gas or vapor in air that, if ignited, will propagate a flame [73] and possibly result in an explosion, causing a rapid release of energy. More generally, this limit can be determined for any gas or vapor in an oxidizing atmosphere (in this case, N_2O). While the definition specifically refers to gases and vapors, this concept is in this study also extended to all selected fuels, including liquids. From public chemical libraries [74] and documents [75] it is possible to find the concentration of the fuel in an atmosphere of N_2O in order to reach the lower and upper explosion limit (UEL). They are expressed in [% mol]. The mixture ratio corresponding to the given concentrations for the LEL can be determined by using the molar masses of the substances.

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Table 4: Lower and upper explosion limits of the selected propellants for N_2O blends at 20 °C and 101.3 kPa obtained from public chemical libraries[74] and documents[75]

Propellant	LEL [% mol]	UEL [% mol]	ROF at LEL [-]
Methane (CH_4)	1.5	45.9	182.9
Ethane (C_2H_6)	1.3	33.0	112.6
Ethene (C_2H_4)	1.4	40.5	112.1
Ethyne (C_2H_2)	–	–	–
Propane (C_3H_8)	0.7	27.0	142.6
Propene (C_3H_6)	0.8	29.5	130.7
Propyne (C_3H_4)	–	–	–
Butane (C_4H_{10})	0.7	27.0	108.2
Ammonia (NH_3)	4.4	65.0	58.7
Methanol (CH_3OH)	1.4	–	98.1
Ethanol (C_2H_5OH)	0.8	–	119.4
Monosilane (SiH_4)	1.9	87.1	72.1
Hexamethyldisilane ($C_6H_{18}Si_2$)	–	–	–
Dimethylphenylsilane ($C_8H_{12}Si$)	–	–	–

The miscibility of the N_2O-based mixtures is to be analyzed up to their lower explosion limit by determining the maximum mixture ratio up to which the components of the blends are no longer miscible but they are still ignitable.

4.2 Theoretical Background of Miscibility

The phenomenon of miscibility refers to the ability of two liquid substances to mix and form a homogeneous solution. When liquids are combined, various types of mixtures can result[76]. Miscible liquids are those that can completely dissolve in each other, resulting in a uniform solution at any concentration. On the other hand, when two liquids are combined but do not fully dissolve at some concentrations, they form a partial solution or a non-ideal mixture, indicating incomplete interaction at the molecular level. This process is influenced by factors such as polarity, molecular interaction strength and temperature. As the concentration of one liquid in the mixture increases beyond a specific threshold, the system reaches saturation, where the solute can no longer dissolve in the solvent. At this point, the mixture becomes unstable, leading to stratification, where separate layers form. Immiscible liquids, on the other hand, fail to mix at any concentrations and immediately separate into distinct layers. If two immiscible liquids are mixed and one is dispersed into the other in small droplets, often with the aid of an emulsifying agent, this is called an emulsion. If the droplets remain suspended but do not dissolve and eventually separate due to gravitational forces, this is known as a suspension. For the blends of N_2O and the selected fuels, however, behavior similar to the emulsions or suspensions is not expected.

Since the blends will be realized in the liquid phase under high pressures, it is relevant to consider their behavior during expansion and phase changes in the thruster, as this can lead to selective evaporation. This process is influenced by the volatility of the components, their interactions and the physical conditions during the evaporation[77]. Differences in the evaporation rates, primarily due to distinct vapor pressures, can result in concentration gradients within the solution. Research has shown that if the non-volatile liquid is denser, the mixture remains uniform and the evaporation rate can be calculated as for an ideal mixture[78]. Conversely, if the non-volatile component is the less dense, convective mixing becomes negligible and mass transfer in the liquid occurs through unsteady molecular diffusion. According to this statement, a uniform mixture is achieved for all mixtures except the ones based on ethene, ethanol and the silanes. This information can be relevant in scenarios where gravity is a factor and can be important for tests conducted under standard conditions. On the other hand, in space environments outside Earth's atmosphere, gravity's influence diminishes and diffusion becomes the dominant mechanism for mass transfer. Furthermore, in the space environment, the absence of buoyancy can prevent clear phase separation between the gaseous and liquid part of the mixture. As a result, extracting only the gaseous phase for use in the thruster may prove challenging and particular hardware might be required. These factors must be carefully considered in the further characterization of the blends.

Due to the limited data available on the specific propellant blends with the selected fuels, experimental testing of the substances' miscibility is required. This will determine the mixture ratio range that ensures homogeneous mixtures and make the blend suitable for functional use in space propulsion scenarios.

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4.3 Experimental Set-up for Miscibility Tests

To evaluate the miscibility of the compounds, an experimental set-up has been designed. The Piping and Instrumentation Diagram (P&ID) and its practical realization are shown in Fig. 7 and Fig. 8.

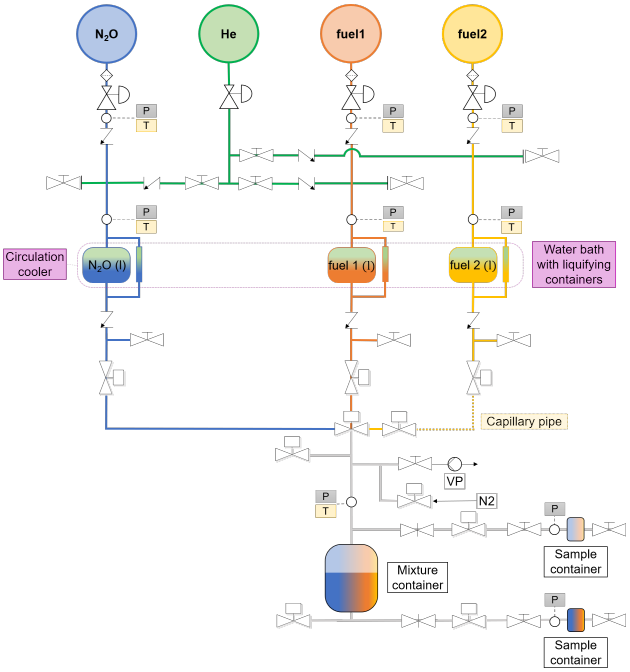

Figure 7: P&ID of the miscibility test-bench

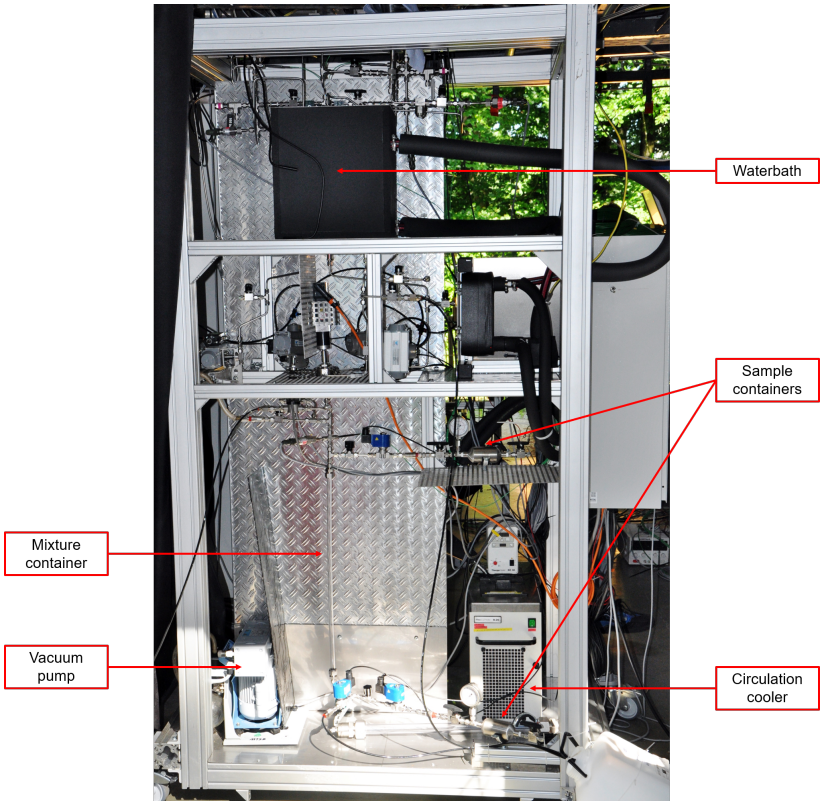

Figure 8: Practical realization of the miscibility test-bench

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The set-up allows for testing the miscibility of one or two fuels in combination with N_2O in a vertical configuration. The substances are directed into containers submerged in a water bath, where their temperature is lowered and the gases are liquefied. The substances are then pressurized with helium, which allows them to move into the mixing containers. Before doing so, volumes of each propellant are measured and dosed inside a pipeline, according to their physical characteristics. The dosing volume allows an increase in the mixture ratio by 1 each time a 2 ml volume of N_2O is added. Since the quantity of the second fuel are very small (representing 10% of the whole fuel mass), the use of a smaller 1.5 mm diameter pipeline is required for its dosage. Afterwards, through a four-way ball valve, controlled by a servomotor, the different liquids are separately introduced into a polycarbonate tube (the mixture container). Due to its transparency, this tube allows the visual observation of the mixture. The polycarbonate tubes have been tested to withstand pressures of up to 100 bar. Samples of the mixtures can be extracted and collected in designated containers for both the liquid and the gaseous phase. These samples can be analyzed using a gas chromatograph to determine the precise quantities of the substances in the mixture. The set-up operates by progressively increasing the mixture ratio through the repeated addition of equal amounts of N_2O to a fixed quantity of fuel. As a consequence, the homogeneity of the mixture can be visually determined through the polycarbonate tube up to the formation of a stratification layer, indicating that the substances can no longer mix. Polycarbonate tubes with different dimensions are selected to allow the Lower Explosion Limits specified in section 4.1 to be reached. By monitoring the composition of the sample after a certain time interval, it is possible to assess whether the mixture remains stable or undergoes separation. Given that the propellant mixtures will be potentially stored in space for an extended period of time, evaluating the long-term stability of the mixture and ensuring that its composition does not change is crucial.

4.4 Expected Results

The literature review identifies the miscible range of a N_2O and ethanol blend up to a mixture ratio of 3.15 [52]. However, this value is significantly lower than the LEL for the blend reported in Table 4. Therefore, a reevaluation of this range will be conducted using the presented set-up to ensure consistency in the testing procedure across all blends. Similarly to ethanol, it is anticipated that the liquid fuels (hexamethyldisilane and dimethylphenylsilane) will have a limited miscibility range, primarily due to the significant difference in vapor pressure. Similarly, the mixture ratio range for N_2O and the hydrocarbons is expected to be broader for the same reason.

5. Conclusions and Outlook

This paper presented a fuel selection for N_2O blends for use in space propulsion applications as a valid green alternative to traditional propellants. Chemical compounds from the families of the hydrocarbons, alcohols, hydrides and silanes have been identified as the most promising fuels and have been evaluated based on specific criteria. The results of this evaluation indicate that ethane, ethene, propane, a mixture of propane (90%) and propene (10%), ammonia, a mixture of ammonia (90%) and propane (10%), ethanol, monosilane, hexamethyldisilane and dimethyphenylsilane are the most promising propellants. Further investigation of these compounds will be essential to finalize the selection of the most optimal fuel. A set-up has been developed to conduct miscibility tests and the selected fuels will be tested in this system to determine the mixtures' miscibility ranges. These results will serve as foundation for subsequent characterization of the fuel mixtures below the explosion limit in order to ensure efficient and safe operation. Once the mixing range is evaluated, the blends can be further described in terms of reaction temperature and ignitability, as well as their interaction with a catalyst. Operational evaluation of the most promising blends can be undertaken in a thruster configuration to assess the monopropellant capabilities for realistic space propulsion scenarios.

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