Replacement of Hydrazine: Overview and First Results of the H2020 Project Rheform

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Abstract

The goal of the EU Horizon2020 project Rheform is the replacement of hydrazine with liquid propellants based on ammonium dinitramide (ADN) for orbital and launcher propulsion systems. Hydrazine and its derivatives are the standard propellants for spacecraft propulsion system since the 1960s, but they are highly toxic and carcinogenic. New regulations will lead to restriction of their use in the near to mid-term. The first part of this article gives an overview on ADN-based propellants and of the Rheform project. The second part contains the results of thermochemical calculations showing the influence of propellant formulation on performance and combustion chamber temperatures.

1. Introduction

The development of orbital propulsion systems based on green propellants is currently a priority of both European and American space agencies. They aim at a complete replacement of hydrazine based systems. Hydrazine and its derivatives have been the standard for spacecraft propulsion system since the 1960s. These propellants are highly toxic and carcinogenic, increasing the complexity and cost of testing, shipping, handling and launch preparation. In 2011 hydrazine was added to the candidate list of substances of very high concern (SVHC) by the Europe's Registration Evaluation Authorisation and Restriction of Chemicals (REACH) [1]. Both NASA and ESA are considering the risks restriction or prohibition in the use of hydrazine in the near to mid-term.

The substitution of hydrazine with green propellants may lead to significant benefits, if suitable propellants are selected. Very interesting replacements for hydrazine are liquid propellants based on ammonium dinitramide (ADN, $NH_4^+ N(NO_2)_2^-$) [2]. They have the following advantages compared to hydrazine:

- Lower overall life cycle cost due to simplified handling especially at the launch site.
- Higher overall performance (I_{sp}).
- Higher volumetric specific impulse due to higher density leading to smaller tanks, and therefore reduced structural weight.

More details on ADN-based propellants and propulsion systems are given in section 2. In section 3 the Rheform project and its goals are presented. In section 4 the results of thermochemical calculations for different compositions of ADN-based propellants are shown.

2. ADN-based propellants - State of the Art

2.1 Development of ADN-based propellants.

ADN is mainly intended as an oxidizer in solid rocket propellants [3]. In the beginning of the 1990s, the Swedish Defence Research Agency, FOI, supported by the Swedish Armed Forces, started its research on ADN in order to develop minimum smoke solid propellants for tactical missile applications. Due to the mixture of ADN with water and the very low volatility of ADN (Vapour pressure: 1.7×10^{-12} mm Hg at 25 °C [4]) the handling of the liquid propellant is safer than hydrazine.

In the mid-1990s, FOI in cooperation with the Swedish Space Corporation, SSC, started to study ADN based liquid monopropellants [5]. The first ADN-based liquid monopropellants developed, LMP-101 and LMP-103, had poor thermal stability, but this was solved by adding a stabilizer [6] later published to be ammonia. The liquid monopropellant LMP-103S is easy to handle and to transport due to its low toxicity and low sensitivity, and has received a UN/DOT 1.4S transport classification in its transport configuration (a 5 L polyethylene jug, in a wood box with absorbent); thus allowing it to be transported on commercial passenger aircraft. Moreover, there are no Substances of Very High Concern (SVHC) in the propellant LMP-103S provided by ECAPS; hence it is compliant with the European REACH regulation.
The first in-space demonstration of an ADN-based propulsion system was conducted on the PRISMA spacecraft. During the launch campaign of PRISMA, ECAPS was responsible for the propellant loading at the Yasny launch base (including cold gas, LMP-103S and hydrazine). The handling of LMP-103S was evaluated and declared to be a "non-hazardous operation" by the Yasny Range Safety, so SCAPE suits were not required during the PRISMA LMP-103S loading operation [7].

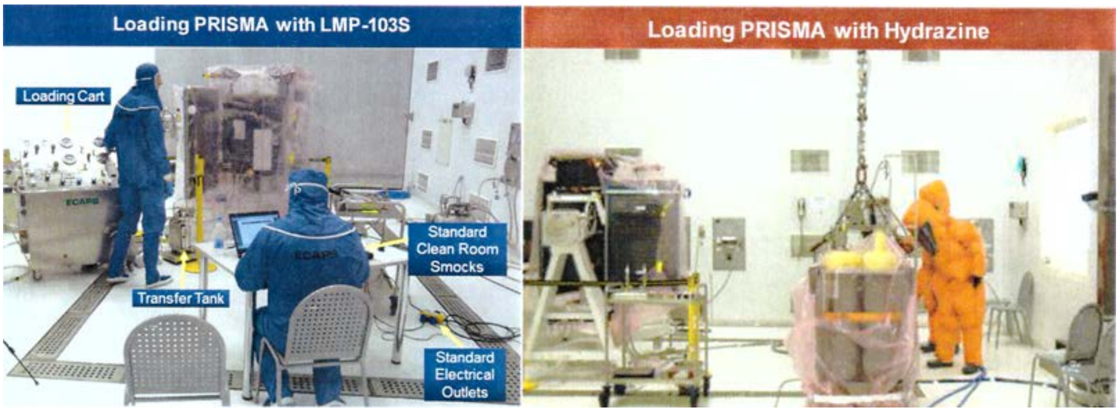

Figure 1: Propellant loading of Satellite PRISMA

Over the last years, FOI has continued to improve the propellants and has developed the high performance propellant FLP-106. One of the main advantages of this propellant is the very low volatility.
In conclusion it can be said that ADN based liquid propellants are one of the most promising technologies to replace hydrazine and to increase the overall performances.

2.2 In-space demonstration of ADN-based propellants - PRISMA

As previously mentioned, the first in-space demonstration of ADN-based propulsion system was conducted with the PRISMA spacecraft. PRISMA is a pair of test bench spacecrafts, focussing on the areas of formation flying and propulsion technologies. Launched in June 2010, they have been in full operation since August 2010 demonstrating autonomous formation flying as well as rendezvous and proximity operations using a suite of formation sensors. The larger of the two spacecraft, Mango, shown in Figure 2, is equipped with two propulsion systems: a baseline hydrazine propulsion system and a High Performance Green Propulsion (HPGP®) system. The HPGP system is a flight demonstrator and additionally provides the ΔV required for the formation flying manoeuvres. The PRISMA HPGP propulsion system employs a conventional monopropellant architecture (same architecture from tank to latch valve for hydrazine) and is built from Commercial Off-The-Shelf (COTS) components with extensive flight heritage [8]. The storable liquid monopropellant LMP-103S has a higher combustion temperature than monopropellant hydrazine. This requires that the reactor bed, thrust chamber and nozzle are made from refractory materials that are able to withstand the increased temperatures. The PRISMA HPGP propulsion system, with its two 1 N thrusters, is designed to provide a total ΔV of more than 60 m/s.

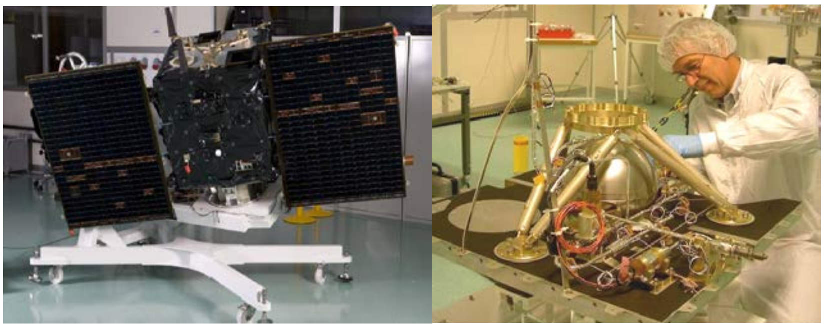

Figure 2: PRISMA main satellite Mango and HPGP integration

The basic mission for the HPGP system has been successfully completed and all objectives corresponding to TRL-7 have been met. Additionally, via the PRISMA extended mission, the HPGP demonstration has evolved into a full qualification; thus reaching TRL-9 (in 2012) for this category of mission [9]. More than 344 sequences comprising over 50,525 pulses have been performed in continuous, pulse or off-modulation mode. Performance mapping has been performed by executing firing sequences with pulse durations from 50 ms up to 100 s and pulse mode firings have been performed with duty cycles ranging from 0.1 % to 99 %. Pulse trains lasting up to 90 min have been executed. Pulse mode and single impulse bit predictability has been demonstrated to be very accurate for the HPGP system. The accumulated burn time is more than 3.5 h to date and 76 % of the propellant being consumed. The remaining propellant will be used to provide ΔV for extended mission objectives before eventual decommissioning.
For a given tank size, the HPGP propulsion system on PRISMA has been demonstrated to provide on an average approximately 32 % higher ΔV capability over hydrazine at a 1 N thrust level, due to the combination of its 24 % higher density and 8 % higher I_{sp} when used in steady state firing, as shown in Table 1. The performance increase over hydrazine in single pulse firing was even larger due to the efficient pulse mode operation of the HPGP thrusters.

Table 1: In-Space demonstrated performance of HPGP, using LMP-103S

Mode	**Calculation Method**	**Value in comparison with hydrazine**
Steady State Firing	I_{sp} for last 10 s to 60 s firings	6 - 12 % higher I_{sp} 30 - 39 % higher ρI_{sp}
Single Pulse Firing	T_{on}: 50 ms – 60 s First half of the mission	10 - 20 % higher I_{sp} 36 - 49 % higher ρI_{sp}
Pulse Mode Firing	T_{on}: 50 ms – 30 s Duty Factor: 0.1-97 %	0 - 12 % higher I_{sp} 24 - 39 % higher ρI_{sp}

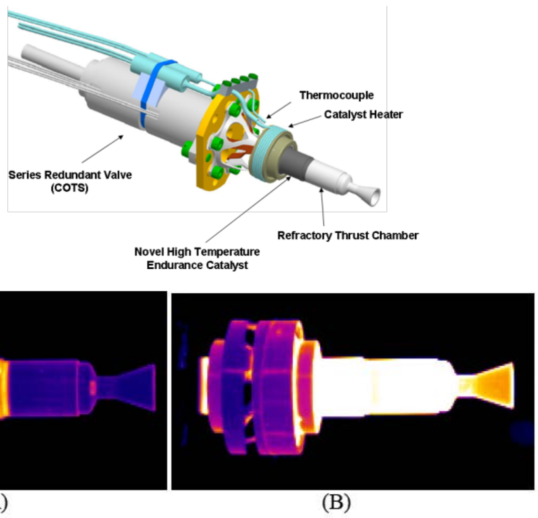

Figure 3: HPGP thruster design (A Preheating, B Operational Mode)

2.3 Commercial application of ADN-based propellants - Skybox

Skybox Imaging recently became the first commercial company to baseline ECAPS HPGP technology, implementing a propulsion system design with four 1 N thrusters on their second generation small satellite platform (< 150 kg). The first propulsion module, delivered in 2014, will serve to qualify the system design for use in an entire constellation of small satellites intended to provide customers easy access to reliable and frequent high-resolution images of the Earth.
A detailed trade study of various propulsion technologies and vendors was conducted by Skybox during the selection process [10]. A key technical requirement for the propulsion system was to provide the maximum possible ΔV (for continued orbit maintenance and mission flexibility) within a limited internal volume typical for small satellites. Additionally, in light of the commercial nature of the project, the overall life-cycle cost was considered to be of utmost importance.
The results of that study showed that the HPGP solution selected provides significantly more on-orbit ΔV compared to traditional monopropellant systems. Moreover such system has the lowest projected life-cycle cost compared to the other liquid propulsion technologies evaluated. ECAPS has been awarded a contract by Skybox to supply 12 complete HPGP propulsion system modules for the SkySat constellation. The HPGP modules are to be delivered in 2015 and 2016. The HPGP systems are manufactured by ECAPS in Sweden, but the propulsion systems use tanks from ATK [11], valves from Moog and thrust chambers from Plasma Processes. Therefore, the flagship of Europe will be based on components that are manufactured in the US and restricted under the export control laws of International Traffic in Arms Regulations (ITAR) or the Export Administration Regulations (EAR).

3. The Rheform Project

Rheform is a project funded from the European‘s Union Horizon 2020 programme. The acronym Rheform stands for: “Replacement of hydrazine for orbital and launcher propulsion systems”. The consortium comprises 7 partners from industries, SMEs, universities and research institutes and represents 4 European countries: Austria, France, Germany and Sweden. The Rheform project runs from 01.01.2015 to 31.12.2017.

Goals

The goal of Rheform is the replacement of hydrazine by ADN-based liquid propellants for orbital and launcher propulsion systems. The two baseline propellants for the project are LMP-103S and FLP-106. As mentioned in section 2, these propellants require a combustion chamber made of special materials that are currently ITAR restricted. In the project, new propellants compositions will be developed and tested aiming at reducing the combustion temperature such that ITAR-free materials can be used for catalysts and combustion chambers. Another focus of the project is to improve the cold start capabilities and thus reduce the need for pre-heating of the system. In order to achieve these goals two different paths will be followed. On one side the catalytic ignition system will be improved. On the other side thermal ignition systems will experimentally be assessed. Here conventional systems, such as torch igniters, as well as advanced systems, such as laser ignition systems, will be considered. If necessary a combination of catalytic and thermal ignition will be considered. The propellant blend and ignition method will be verified with one or two demonstrator(s), equivalent to a TRL of 5. In the project existing numerical models will be adapted to describe the processes in the propulsion system. Possibilities for the optimization of the production process of ADN will be studied as well in order to reduce the cost of producing this fuel.

Partners

A list of the partners and of the key personnel for the Rheform project is given in Table 2.

Table 2: Rheform Partners

Partner	Country	Project Key Personnel
DLR	Germany	Michele Negri Christian Hendrich
FOI	Sweden	Niklas Wingborg Martin Skarstind
CNRS	France	Yann Batonneau Romain Beauchet

FOTEC	Austria	Charles Kappenstein Carsten Scharlemann Sebastian Schuh
ECAPS	Sweden	Robert-Jan Koopmans Mathias Persson Kjell Anflo
Airbus	Germany	Wilhelm Dingertz Ulrich Gotzig Peter Gambach
Lithoz	Austria	Martin Schwentenwein

Most partners have close ties with each other. The DLR and Airbus are located on the same site in Lampoldshausen, Germany. FOI and ECAPS are both located near Stockholm and use the same test facilities. The close proximity allows a direct collaboration and assures short response times.

FOTEC, Lithoz and CNRS work together on different projects. In the project they complement each other for the work on catalyst design, preparation and testing: CNRS and Lithoz will prepare catalysts. CNRS will test small amount of propellants while FOTEC will perform tests with simuli. Additionally, the consortium member ECAPS has already built a catalytic ignition system and is therefore the member with the most experience in building catalytic ignition systems for ADN-based liquid propellants in Europe.
To address also the commercialisation of the project results, the consortium consists also of a prime of space propulsion in Europe, Airbus.
The consortium also includes a space propulsion prime, Airbus, which is helpful in evaluating the commercialisation possibilities of the project results.

Methodology

A schematic representation of how the research in the Rheform project is structured shown by means of a flowchart in Figure 4.

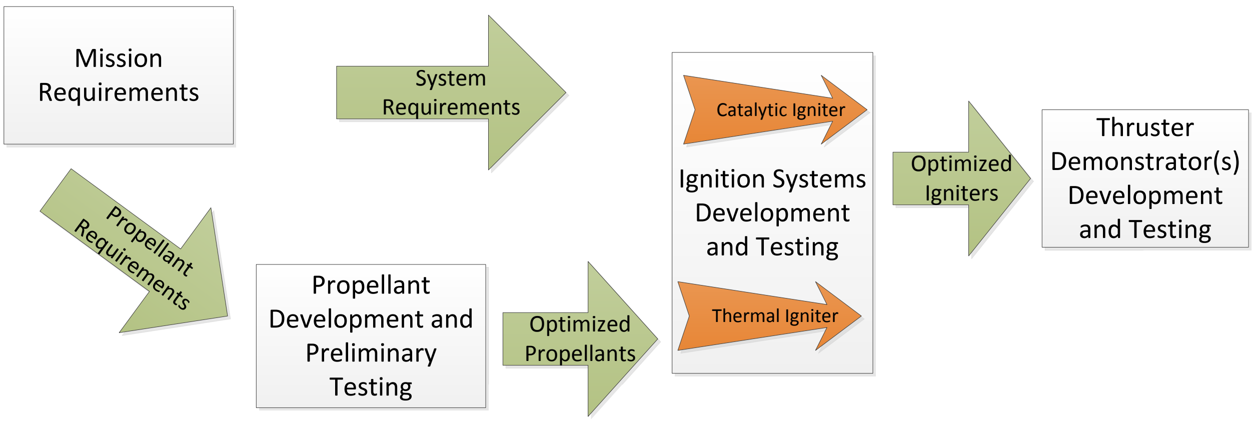

Figure 4: Flowchart of the Rheform Project

In the first phase the requirements on the propellant and on the propulsion system will be defined. The use of COTS components previously developed for hydrazine systems will be considered strived for. In this phase also different ITAR-free combustion chamber materials will be considered. Two thrust levels will be considered: one in the range of 10 to 20 N and one in the range of 200 to 400 N.

Based on these requirements, propellants with different composition will be defined and produced. The physical properties of these propellants will be characterized. A preliminary assessment of the ignitability of these propellants will allow the selection of the most promising candidates. The ignitability will be studied using a standard ignition method, the card gap method, as well as an advanced method, laser ignition.

A central point of the project will be the development of ignition methods. Goal is to reduce the amount of power required from the igniters. Two types will be considered: passive and active igniters. Passive ignition will be achieved by means of a catalyst. An important goal here is to reduce the required power for pre-heating. Active ignition will be achieved with thermal igniters. Here, both traditional methods, such as spark plug and torch igniters, as well as novel methods, such as microwave and laser ignition, will be considered. Based on a literature review the most promising methods will be tested experimentally.

Based on the propellant blends and ignition methods selected in the previous phase, one or two thruster demonstrators will be built for further investigation. Goal of the demonstrator is to reach a development level equivalent to a TRL of 5.

4. First Results: Thermochemical Calculations

One of the first activities of the Rheform project was performing thermochemical calculations of different propellant formulations. Calculations of the combustion temperature (T_c) and of the specific impulse (I_{sp}) assuming thermochemical equilibrium were conducted using NASA's CEA code. The same simulation parameters as in the GRASP project were used (p_c = 10 bar, ε = 40, frozen state and vacuum expansion), allowing a direct comparison of the results. The GRASP project was an EU funded project on the development of green propellants running from 2008 to 2011. More information on this project can be found in ref. [12]
For both baseline propellants (FLP-106 and LMP-103S) the specific impulse and combustion temperature were calculated with increasing the water content. Moreover the influence of variation of the relative amount of reactants on the performance was investigated.
The thermochemical data provided in the CEA database were used for all compounds. The only exceptions were ADN, monomethylformamide (MMF, CH_3NHCHO) and aqueous ammonia. For ADN the heat of formation was taken from Kon'kova et. al. [13], $\Delta H_f^0 = -134.6\ \mathrm{kJ/mol}$. This was added to the heat of solution for ADN, which can be calculated with the following equation as suggested by FOI:

$$\Delta H_s \ \ (\mathrm{kJ/mol}) = +36.6 - 0.194(\%\mathrm{ADN}) \qquad (1)$$

The heat of formation of the solution used for the calculation was recalculated with the following equation for each formulation, based on the effective percentage of ADN after the addition of water.

$$\Delta H_{f,liquid} \ \left(\frac{\mathrm{kJ}}{\mathrm{mol}}\right) = -98.0 - 0.194(\%\mathrm{ADN}) \qquad (2)$$

For MMF the heat of formation of -247.4 kJ/mol was used [14]. The enthalpy of formation for the solution of 25 % ammonia in water was determined by linear interpolation of the values available in literature [15] (measured with solutions of 15.9 % and 32.1 % ammonia in water). From this the value of enthalpy was determined to be -78.37 kJ/mol.

Variation of the Composition of FLP-106

The calculated values of the specific impulse and the combustion temperature for FLP-106 with increasing amounts of added water are shown in Table 3.

Table 3: Overview of the combustion temperature (T_c) and of the specific impulse (I_{sp}) as a function of the water content for FLP-106 variations; p_c=10 bar; ε=40, frozen.

Propellant	H_2O [%]	ADN [%]	MMF [%]	T_c [°C]	I_{sp} [s]
FLP-106	23.90	64.60	11.50	1904	258
FLP-106 + 1 % Water	24.65	63.96	11.39	1877	257
FLP-106 + 2 % Water	25.39	63.33	11.27	1850	255
FLP-106 + 3 % Water	26.12	62.72	11.17	1823	254

FLP-106 + 4 % Water	26.83	62.12	11.06	1796	252
FLP-106 + 5 % Water	27.52	61.52	10.95	1769	251
FLP-106 + 6 % Water	28.21	60.94	10.85	1742	249
FLP-106 + 7 % Water	28.88	60.37	10.75	1716	248
FLP-106 + 8 % Water	29.54	59.81	10.65	1690	246
FLP-106 + 9 % Water	30.18	59.27	10.55	1664	245
FLP-106 + 10 % Water	30.82	58.73	10.45	1639	243
FLP-106 + 11 % Water	31.44	58.20	10.36	1613	241
FLP-106 + 12 % Water	32.05	57.68	10.27	1589	240
FLP-106 + 13 % Water	32.65	57.17	10.18	1564	238
FLP-106 + 14 % Water	33.25	56.67	10.09	1540	237
FLP-106 + 15 % Water	33.83	56.17	10.00	1517	235

Figure 5 shows the influence of variation of the relative amount of reactants on the combustion temperature T_c. The ADN content is plotted on the x-axis, the MMF content is on the y-axis. The gap between ADN and MMF content for 100 % reflects the water content.

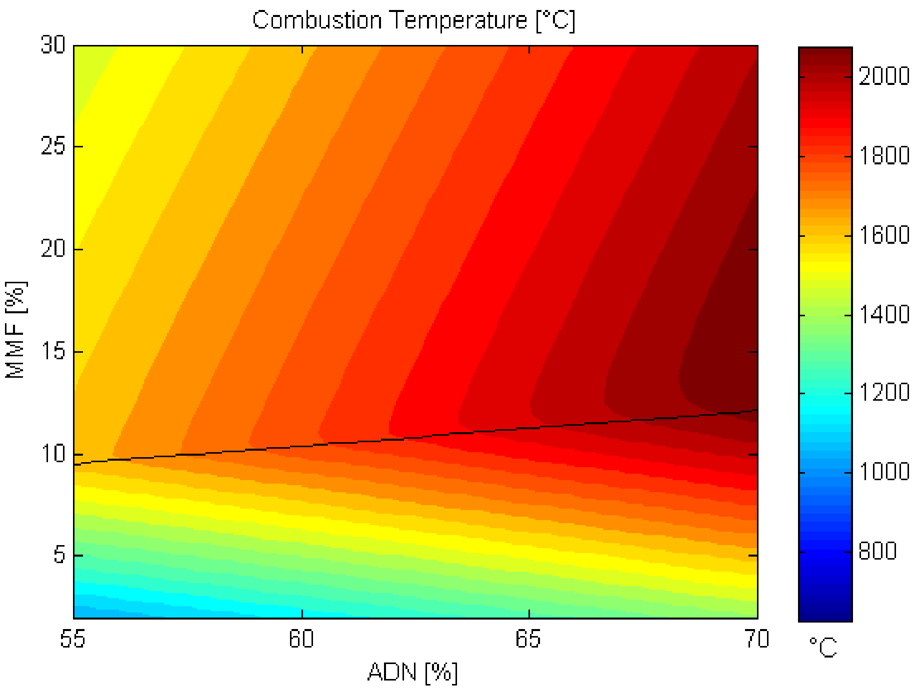

Figure 5: Combustion temperature T_c of ADN-MMF-H_2O monopropellants, as a function the composition. The black line indicates stoichiometric conditions. The gap between ADN and MMF content for 100 % reflects the water content.

For the same compositions, the specific impulse was calculated and shown in Figure 6

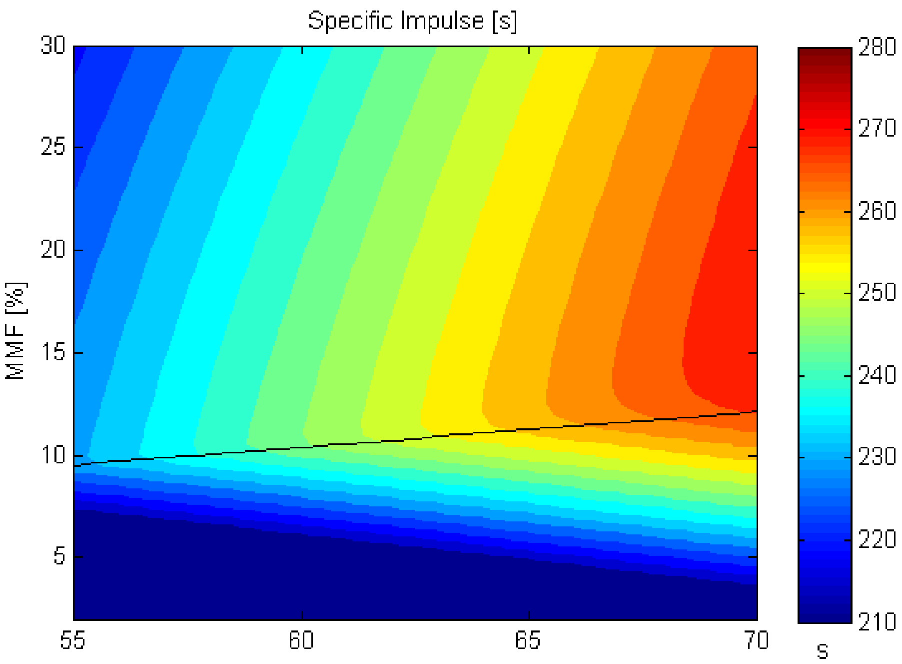

Figure 6: Specific impulse of ADN-MMF-H_2O monopropellants, as a function the composition.The black line indicates stoichiometric conditions. The gap between ADN and MMF content for 100 % reflects the water content.

In Figure 7 the influence of the variation of the relative amount of reactants for ADN-MMF-H_2O monopropellants on the specific impulse and the combustion chamber are shown in the same graph.

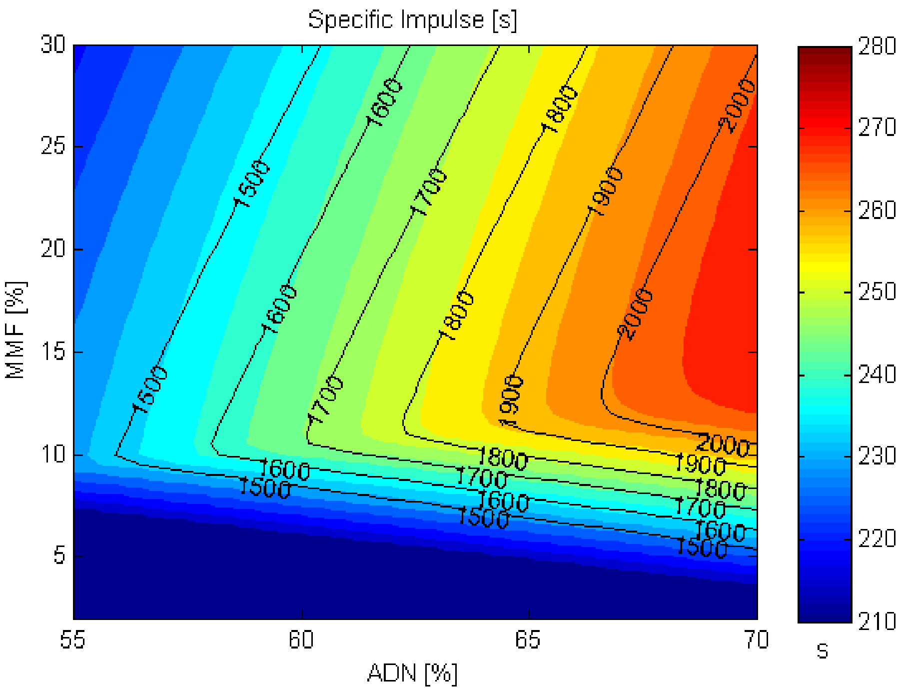

Figure 7: Specific impulse, indicated by colours, and combustion temperature, indicated by isotherms, of ADN-MMF-H_2O monopropellants as a function of composition. The gap between ADN and MMF content for 100 % reflects the water content.

Variation of the Composition of LMP-103S

The same calculations varying the relative amount of the components of LMP-103S (ADN, Methanol, Ammonia, and Water) were conducted. In Table 4 the influence of adding water to LMP-103S on combustion temperature (T_c) and of the specific impulse (I_{sp}) is shown.

Table 4: Overview of the combustion temperature (T_c) and of the specific impulse (I_{sp}) as a function of the water content for LMP-103S variations; p_c=10bar; ε=40, frozen.

Propellant	H_2O [%]	ADN [%]	Methanol [%]	Ammonia [%]	T_c [°C]	I_{sp} [s]
LMP-103S	13.95	63.00	18.40	4.65	1645	254
LMP-103S + 1% Water	14.80	62.38	18.22	4.60	1619	252
LMP-103S + 2% Water	15.64	61.76	18.04	4.56	1593	250
LMP-103S + 3% Water	16.46	61.17	17.86	4.51	1568	249
LMP-103S + 4% Water	17.26	60.58	17.69	4.47	1543	247
LMP-103S + 5% Water	18.05	60.00	17.52	4.43	1519	245
LMP-103S + 6% Water	18.82	59.43	17.36	4.39	1495	244
LMP-103S + 7% Water	19.58	58.88	17.20	4.35	1471	242
LMP-103S + 8% Water	20.32	58.33	17.04	4.31	1448	240
LMP-103S + 9% Water	21.06	57.80	16.88	4.27	1426	239
LMP-103S + 10% Water	21.77	57.27	16.73	4.23	1404	237

Figure 8 shows the influence of variation of the relative amount of reactants on the combustion temperature T_c. The ADN content is plotted on the x-axis, the methanol content is on the y-axis. The difference between ADN and methanol content for 100 % is the varied aqueous ammonia content.

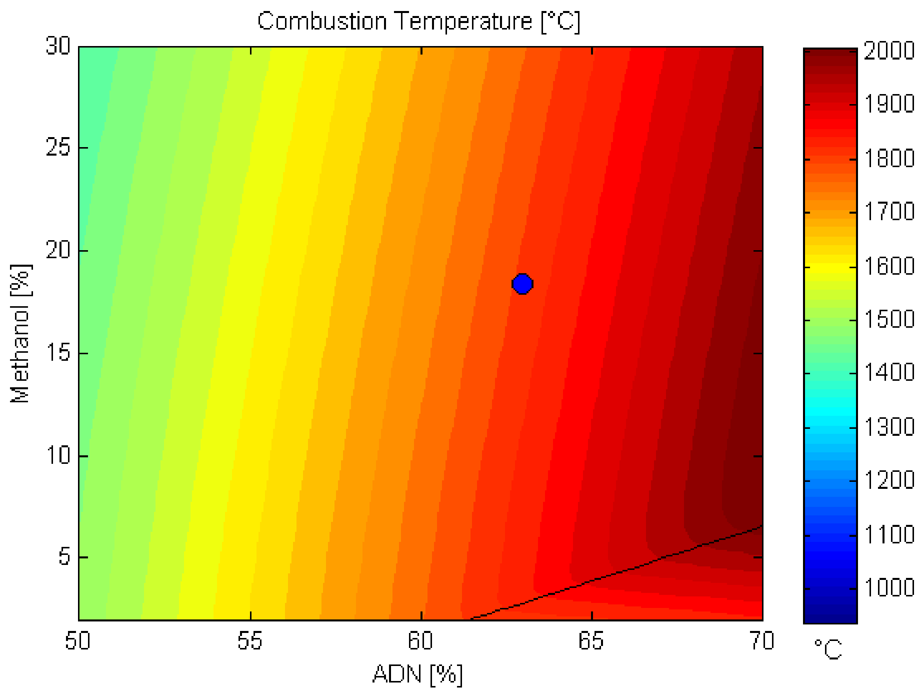

Figure 8: Combustion temperature T_c of ADN-Methanol-NH_3 (aq. 25 %) monopropellants, as a function the composition. The black line indicates stoichiometric conditions. The blue point indicates the composition of LMP-103S. The gap between ADN and MMF content for 100 % reflects the NH_3 (aq. 25 %) content.

The specific impulse in dependency of the composition is plotted in Figure 9.

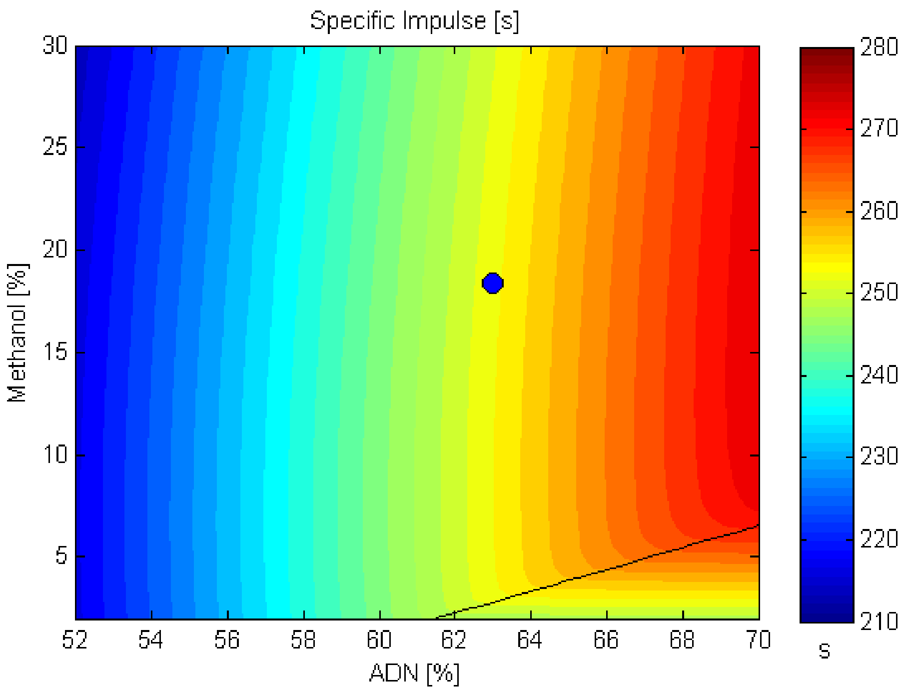

Figure 9: Specific impulse of ADN-Methanol-NH_3 (aq. 25 %) monopropellants, as a function the composition. The black line indicates stoichiometric conditions. The blue point indicates the composition of LMP-103S. The gap between ADN and MMF content for 100 % reflects the NH_3 (aq. 25 %) content.

Figure 10 is a combination of the two previous graphs, showing how the I_{sp} and the combustion temperature are affected by changes in composition.

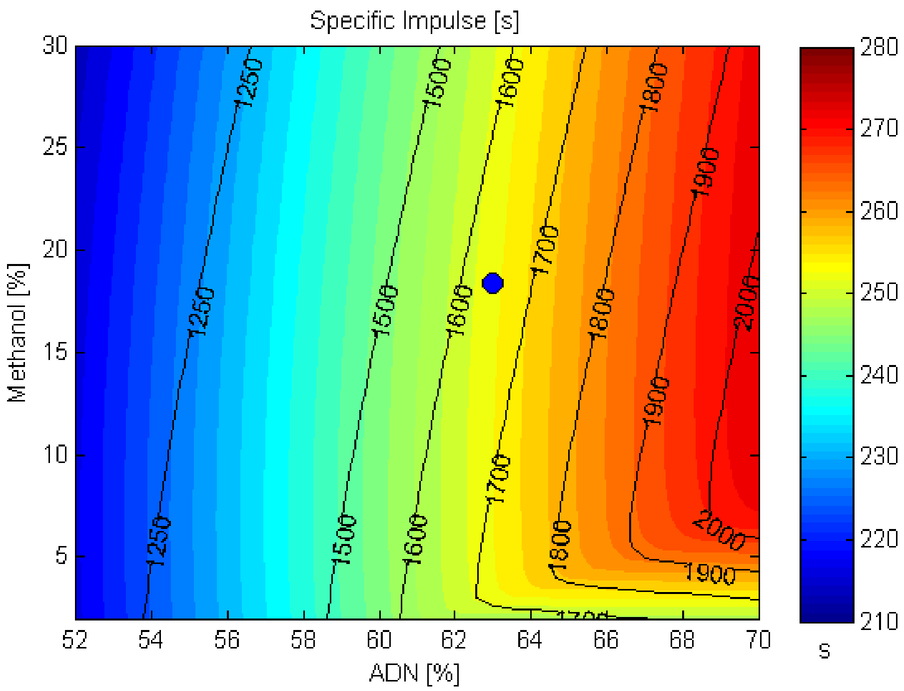

Figure 10: Specific impulse, indicated by colours, and combustion temperature, indicated by isotherms, of ADN-Methanol-NH_3 (aq. 25 %) monopropellants as a function of composition. The blue point indicates the composition of LMP-103S. The gap between ADN and MMF content for 100 % reflects the NH_3 (aq. 25 %) content.

Discussion and Propellants Selection

The results of the thermochemical calculations show that an addition of water to the existing propellants leads to a reduction of the combustion temperature. In rocket thruster without active cooling, the requirements on the combustion chamber temperature are mainly set by the material of the combustion chamber. In Table 5, a list of different possible combustion chamber materials is given. A typical value of the maximum temperature admissible for each material is also listed. Using NASA CEA it was possible to determine the propellant composition suitable to keep the combustion temperature within the limitation posed by the material.
Most of the propellants proposed are the baseline propellants, FLP-106 and LMP-103S, with the addition of water. For combustion chamber temperatures of 1700K and higher, also other variations of the propellants are proposed, shown in grey in the table. It should be noticed that these other compositions proposed were optimise only on the results from NASA CEA calculations. They may not be usable for problems of stability, solubility, or ignitability.

Table 5: Propellant suggested for different combustion chamber materials.

Material	Tmax [°C]	Propellant	Tc NASA CEA [°C]	I_{sp} [s]
Super Alloy	1250	LMP-103S + 17.4 % H2O	1253	226
		FLP-106 + 27.7 % H2O	1249	217
Platinum Rhodium	1500	LMP-103S + 5.8 % H2O	1499	244
		FLP-106 + 15.7 % H2O	1500	234
Platinum Iridium	1600	LMP-103S + 1.8 % H2O	1598	251
		FLP-106 + 11.5 % H2O	1601	241
Ceramic	1700	FLP-106 + 7.6 % H2O	1700	247
		64 % ADN 18 % Methanol 18 % Ammonia (Aq 25%)	1700	257
		65 % ADN 26.5 % Methanol 8.5 % Ammonia (Aq 25%)	1699	259
	1800	FLP-106 + 3.9 % H2O	1798	253
		66 % ADN 19 % Methanol 15 % Ammonia (Aq 25 %)	1800	263
		67 % ADN 27 % Methanol 6 % Ammonia (Aq 25 %)	1801	267
	1900	FLP-106	1904	258
		68 % ADN 20 % Methanol 12 % Ammonia (Aq 25 %)	1900	268
		69 % ADN 28 % Methanol 3 % Ammonia (Aq 25 %)	1901	270
Rhenium Iridium	2000	70 % ADN 22 % Methanol 8 % Ammonia (Aq 25 %)	1993	274

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