Green Propellants: Global Assessment of Suitability and Applicability

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The increasing interest in so-called green propellants is driven by several motivations. The lower toxicity level of green propellants promises reduced operating expenses to protect human operators and the environment, effectively reducing the required costs. This is true not only for handling, transportation and storage of the propellants, but also for hardware development and ground tests in general. Furthermore, the envisioned increased human presence in space requires the use of propellants that are less dangerous to human health than those presently used.

Due to such prospects, individual research efforts were initiated worldwide to investigate various candidates that are considered to be green propellants. However, a comprehensive and unbiased effort to investigate green propellants was missing up to this point. For this reason a European consortium, financed by the European Commission in the 7th Framework Program (FP7) and consisting of 11 entities from 7 European countries, was established to investigate green propellants in a most comprehensive fashion. One of the first goals of this project, called "Green Advanced Space Propulsion" (GRASP), is the establishment of a green propellant survey and assessment of the possible technological and financial impact.

The present paper presents an overview of the GRASP consortium and the structure of the project. It further provides a comprehensive survey of green propellant candidates, together with the assessment strategy to evaluate their suitability to replace presently used propellants. Near term applications and a possible future use of green propellants as well as the technological and financial impact of a replacement of presently utilised propellants with green propellants, are discussed.

I. Introduction

Most of the presently flown propulsion systems are using propellants which are considered toxic and extremely dangerous for the personnel handling them. This includes hydrazine, monomethyl hydrazine (MMH), unsymmetrical dimethylhydrazine (UDMH) as fuels and mixed oxides of nitrogen (e.g.MON-3) and nitrogen tetroxide (NTO) as oxidizers to name only some. In spite of their toxicity they are extensively used due to their good performance, hypergolicity, extensive experience and space heritage existing for such systems and, plainly spoken, because most companies active in the propulsion development sector have focused only on such systems and have no business alternative.

However, especially over the last two decades the situation has changed. Interest in the toxicity of propellants first appeared in the mid 1990s, when more stringent procedures began to be introduced for the ground-handling of hydrazine[1]. The allowable 8-hour threshold level values (TLV) decreased continuously and were by 1995 roughly two magnitudes lower than that in 1965[2]. This does not only increase the efforts and therefore the costs for the development of toxic propellant based systems but also for handling the propellant and the propellant procurement costs. For example, while by 1990, NASA procured hydrazine for 17.00 $ per kg, in 2006 they had to pay already 170.00 $ per kg – a tenfold increase in a relatively short time[3].

Due to such cost increases interest in propellant alternatives surfaced again. The search for such alternatives was however not spearheaded by the relevant industry itself but by academia, research institutes and smaller companies. Even up to this date, for the most part the relevant industry shows only marginal interest in green propellants.

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The reasons for this are complicated but considering the cost and in particular the anticipated return of investment associated with, for example a communication satellite, one can comprehend their reluctance to utilize new technologies. Besides the financial aspects, the technical complexity and in general the missing, or perceived missing, information and background with regard to green propellants might have been another reason for the industry not to embrace the search for propellant alternatives.

As mentioned before, many different groups worldwide are working on the development and investigation of green propellants. In general, each group advertises mainly their particular propellant or system they are working on. What was missing is a comprehensive and unbiased investigation of green propellants in general. The GReen Advanced Space Propulsion project (GRASP), is an activity funded by the European Union in the 7th Framework Project and its main aim is to provide such an assessment. The present paper provides some information about the GRASP efforts, the team which is working on it and also presents some of the first results in regard to an initial green propellant assessment and market analysis.

II. GRASP overview

The GRASP project aims to provide the European industry with a comprehensive review of propellants which are considered to be a viable alternative to the presently used highly toxic and carcinogenic propellants (e.g. hydrazine, MMH, UDMH). These alternative, so-called green propellants will reduce the potential harm to human operators and the environment and thereby significantly reduce the associated costs. At the same time such propellant alternatives should provide similar or better performance and have minimum impact on spacecraft level. Such an ideal propellant is not easily identifiable and the search for it requires a significant effort.

The new concepts of “green propellants” and “green propulsion” is linked to the more general interest in the reduction of environmental impact of chemicals and fuels by developing alternative and sustainable technologies that are non-toxic to living things and the environment and the appearance of “green chemistry”. Therefore, GRASP promotes the “Twelve Principles of Green Chemistry” which are of particular interest in the field of propulsion: prevent waste; design safer chemicals to be fully effective, yet have little or no toxicity to humans and the environment, use renewable feedstocks, analyze in real time to prevent pollution, minimize the potential for accidents including explosions, fires, and minimize release of harmful substances into the environment.

The GRASP project will run for three years. Project kick-off was in December, 2008. As one of the first tasks of GRASP, a market analysis was established. Such a market analysis shall help to understand the market environment, to identify the major players and decision makers, elucidate the space propulsion market volume and in general identify the most promising business opportunities for green propellants. For this purpose a relative large data base of market relevant information was established, compiled and analysed. Some of the main results are given in the present paper.

Along with the market analysis a status quo of the green propellant research was established. In this initial phase of the GRASP project, a number of roughly 100 possible green propellants have been suggested by the GRASP team members. To deal with this large number of candidates a system of filters was established; while each filter is focusing on certain aspects of the propellants, the system of filters is designed to systematically identify the most promising green propellant candidate. Although the system of filters is appropriate for such a task, the GRASP team is aware of the danger to reject a propellant based on certain filter applied early in the assessment process although this particular propellant might have in a global assessment done well or better than others which were not rejected in an early phase. Nevertheless, this risk was accepted by the GRASP team in order to include as many propellant candidates as possible in the early stage of the assessment.

In the very initial phase a choice was made which propulsion systems will be included within GRASP. To a certain extent, this constitutes at the same time the very first propellant filter (e.g. solid propellant booster systems were excluded). The second filter consisted of a toxicity assessment of the propellant candidates to identify those which really can be considered to be “green”. For those propellants which were considered by GRASP to qualify as green propellants, an extensive data and information base was established. This data base was used to further assess the candidates according to certain criteria.

The GRASP effort has three primary fields to be investigated: propellants, catalysis and propulsion systems. In order to achieve the above goals a team has been assembled which is composed of some of the major players in the green propellant field in Europe. Each team members has one or several of the above mentioned fields of expertise shown in general terms in Table 1. Each team member will cover a different aspect/field of the green propellant topic to allow the largest possible range of expertise and results.

Table 1: GRASP team composition and expertise

Organisation	Country	Expertise/Responsibility
Austrian Research Centers GmbH (ARC)	Austria	Project coordinator, propulsion system development
Swedish Defence Research Agency (FOI)	Sweden	Catalyst and propellant development
University of Southampton (SOTON)	England	Catalyst, propellant, and propulsion system development
Centre National de la Recherche Scientifique (CNRS/Univ. of Poitiers/LACCO)	France	Catalyst and propellant development
DELTACAT Limited (DC)	England	Catalyst, propellant, and propulsion system development
DIAS - University of Naples "Federico II" (UN)	Italy	Catalyst, propellant, and propulsion system development
German Aerospace Center (DLR)	Germany	Propulsion system development
Evonik Degussa GmbH (DG)	Germany	Propellant development
SNECMA	France	Propulsion system development
Céramiques Techniques et Industrielles (CTI)	France	Catalyst development
Instytut Lotnictwa (IOA)	Poland	Propulsion system development

III. Market Analysis

The implementation of a new technology in an existing branch is always a challenge since well established products and processes have to be reconsidered and potentially changed. In the case of the space industry, which is inherently conservative in its nature and in general opposed to any changes of the status quo this is even more difficult and in most cases a very slow process. To understand better the needs and limitations of the market, a market analysis was established. It shall assess the market potential of green propellants, possible business opportunities and cost impacts. In the following a summary of this market analysis is provided.

Based on various sources[3,4,5,6,7,8,9,10,11,12,13,14] the total market volume in terms of propulsion systems and its main supplier has been assessed (the Russian market is excluded due to the lack of available data). It should be stressed that all those systems are using toxic propellants (NTO, MON-3, hydrazine, UDMH, MMH etc.). One result of this assessment is shown in Figure 1. The left side of Figure 1 shows the main supplier of bipropellant ACS/station keeping thrusters and the number of systems flown. The right side of Figure 1 depicts the total number of propulsion units flown as a function of the launch year, propulsion type and thrust class. Similar assessments have been conducted for apogee thruster systems and monopropellant ACS/station keeping thruster.

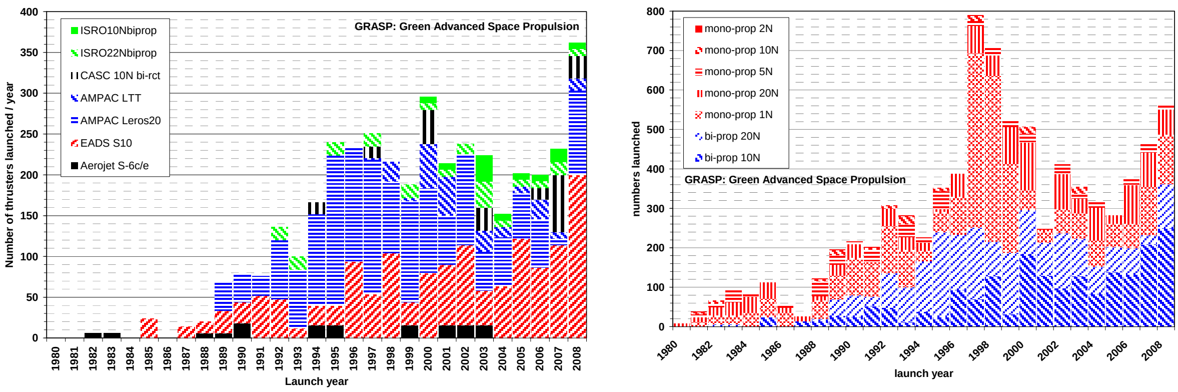

Figure 1: Main supplier for bi-propellant ACS/station keeping thrusters (left) and total number of propulsion systems flown (right)

Based on the available data it can be concluded that attitude control application showed thrust levels typically <20N with mono-propellant and bi-propellant systems. Furthermore, the 1N thruster class (mono-propellant) and bi-propellant thrusters with thrust levels <=10N have currently (and in the near future) the highest market volume. A clear tendency to lower thrust levels for ACS purposes was observed. Total number of units is on average ~400 units. Thrust generation during apogee maneuvers and attitude control for OFASV will be the main application with higher thrust level demand (apogee thrusters: 400 to 500N, OFASV thrusters: 200N and higher). The annual demand is estimated to be 50 to 60 flight units.

Available information suggest that efforts exists at various places to develop propulsion system based on green propellants [15,16,17,18,19,20,21] but at this point in time no commercially available product exists. One of the reasons for this is surely the relative large effort, and associated costs which are still necessary to qualify new propulsion systems based on green propellants and replace existing infrastructure (testing facilities, launch pad fueling facilities etc.). Another reason might be the lack of understanding of the potential of green propellants and in particular the cost reduction potential. It is intuitive that substances with a reduced level of toxicity offer cost reduction in terms of propellant procurement, handling and storage (total ownership cost), ground based test activities, and pre-launch S/C preparation activities to name only some.

Some publications have tried to assess certain aspects of such a cost reduction. One author points out that the reduced hazard level of a propellant has direct and significant impact on the spacecraft/satellite costs with regard to recurring and non-recurring costs[22]. Others point out that by utilizing propellants which do not require the use of SCAPE units significant cost reduction could be achieved in launch preparation and support activities, GSE and personnel protection in general, and assembly and test preparation in particular[23,24].

In spite of the impressive cost reduction potentials shown by the above authors, they deal with only some aspects of the cost issue. A more global assessment is needed to make a clear case for green propellants. However, every company has a different cost environment and different regulations (which impacts again the costs) and facing the same situation chances are high that two different companies will assess the total cost reduction significantly different. It was therefore felt inappropriate to provide real numbers. Instead GRASP focused more on identifying areas impacted by a switch to green propellants rather than to quantify the cost reductions.

As an example of this strategy, Figure 2 shows the detailed steps of pre-launch to post-launch activities[25] including the areas where cost reductions are assumed. For this particular example, cost reductions due to the use of propellants with a reduced hazard level are derived from lower facility rental costs by reducing the time needed for fuelling and pre-launch preparation, lower labor costs due to reducing the number of persons and time needed, lower and cheaper man hours to a normal working environment (compared to SCAPE suits environment).

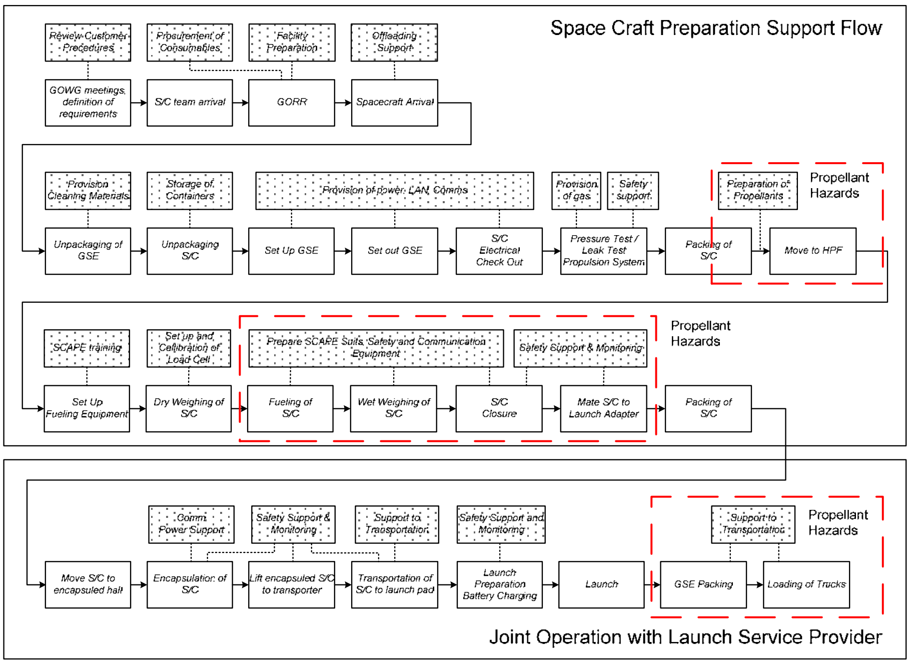

Figure 2: Pre-launch/launch/post-launch activities process flow (as provided by AstroTech Launch Services)[25]

Similar assessments have been conducted for the general propellant handling flow, on-ground test process flow, and spacecraft fuelling process. One has to point out that propellants, independent if they are toxic or green, are in general explosive and extremely flammable. Subsequently, the above processes have many steps which will only marginal or not at all change when using green propellants. On the other hand, all of those processes have a sufficient number of steps which are assumed to be directly influenced by the hazard level of the propellant. Using green propellants is assumed to reduce cost in nearly every field related to the propellant handling.

One particular field of cost reduction is frequently overlooked, namely the propellant procurement costs. Although the propellant costs for a mission are negligible, the propellant procurement costs associated with the development and qualification phase are not. As a rule of thumb for on-ground tests during a development phase an amount of propellant is required roughly 80 to 100 times the amount of propellant which is needed during thruster operational life time. Considering that a large mission requires propellant in the range of tons (with e.g. MMH for 280€/kg) and that some of the green propellants offer a cost reduction of more than a factor 10, the propellant procurement costs indeed constitute a significant cost reduction potential of green propellants.

IV. Green Propellant Assessment

A propulsion system definition was the very first filter GRASP applied. The GRASP effort excludes electric propulsion systems (EP), cold gas system and any solid propellants used presently for booster systems. The propellants used for the major EP system are non-toxic and no necessity exists to replace them based on any toxicity issues. Cold gas system have much lower performance (I_{sp}) than the systems GRASP strives to replace and in most cases also do not have any toxicity issues. Although solid boosters are without doubt the propulsion systems with the most significant detrimental impact on the environment (in operation) of all launch systems, they are excluded at this point of time from the GRASP assessments simply in order to systems under assessment.

Based on the above, the GRASP team established a list of possible green propellant candidates. Those suggestions are based on literature references of past and present research with substances the individual authors have declared as green propellants. In some cases, substances have been included for which no propulsion related literature exists but were considered by a GRASP team member to be a possible candidate as a green propellant.

A: Toxicity assessment

Based on the nature of the GRASP project and its goals, the green propellant candidates were assessed based on their toxicity characteristic. Such an assessment is not as straightforward as it might seem. The general term toxicity is in fact a multifaceted term and depends very much what property or attribute the individual user considers to be the most important. The definitions of toxicity seem to be nearly as numerous as there are propellant candidates. Different studies use different definitions including TWA/TLV[26], TEHF[27], and predicted half life[28]. Other possible parameters with which the toxicity of a substance is frequently defined includes LD50 (oral), LD50 (dermal), LC50 (inhalation), NIOSH REL, OSHA PEL, OSHA STEL, R Phrases, NIOSH IDLH, and IRCH.

Other teams even introduced their own definition of toxicity[29,30] or pick and choose propellants according to a particular characteristic they consider important, e.g. the existence of atoms of chlorine is for one author a property which disqualifies a substance to be considered a green propellant[31]. One study[32] avoids the problem by identifying substances that can in general be considered as a "'Reduced Hazard Propellant" (RHP) while others dismiss such a notion[31].

In addition to the multitude of different toxicity parameters, even for one single parameter (e.g. LC50) there are large inconsistencies in the published data. It is not uncommon to find for one substance sources identifying it as extremely hazardous to human beings and others which define the danger to human beings as benign. Besides the fact that most toxicity studies are conducted with living test species and are therefore subject to large statistical errors, one of the problems is the large variations of test conditions including the general test set-up and used procedures as well as the use of a variety of different test specimens (rats, mice, rabbits etc.).

Facing the above complexity in assessing the toxicity and in general, the hazard level, the GRASP team uses a two step process to identify green propellants. All substances are initially assessed based on the existing R phrases using the data base from Sigma-Aldrich[33] or the Oxford University Physical and Theoretical Chemistry Department[34]. In the second step the substances are assessed according to their acute toxicity data, a term which refers to the dangers associated with propellant handling. The fact that for the more commonly used chemicals the R phrases subsume the acute toxicity ensures the consistency of this assessment and allows inclusion of substances for which no or only insufficient acute toxicity data are available.

The most pertinent R phrases considered in this assessment are shown in Table 1. A substance with a high number of existing R phrases is obviously a more dangerous substance then one with a lower number (without weighting the nature of the R phrase). As a matter of fact, the GRASP reference propellants (hydrazine and its derivates) have each with 3 existing R phrases the highest number. All others substances with such a high number have been immediately discarded as green propellants. For some substances included in the initial list of green propellants no R phrases have been found. All other investigated propellant candidates have one or two existing R phrases.

In the second step, the acute toxicity was assessed for those propellants which have less than 3 existing R phrases. The term acute toxicity refers to danger of a substance associated with a situation in which the handling personnel come in contact with the substance (e.g. leakage). The dominant parameters are here LC50 (inhalation) and LD50 (dermal). The acute toxicity data have been classified according to the EU Hazard Statements[35], shown in Table 2. As a reference, hydrazine has LD50 (oral): 60-200 mg/kg, LD50 (dermal): 91-290 mg/kg, LC50: 330 ppm.

Table 1: R phrases considered in the GRASP assessment

Category	R (Risk) Phrases
toxic or very toxic	23 through to 28
Very serious irreversible effects	39
serious chronic effects	48 and 68
cancer with or without inhalation	45 and 49
genetic damage	46
fertility or embryo damage	60 and 61
harmful or very toxic to aquatic life	50 through to 53
eco-toxic or damage to ozone layer	54 through to 59

Table 2: EU Hazard Categories

Exposure route	Category 1	Category 2	Category 3	Category 4	Category 5
Oral LD50 mg/kg	<5	<50	<300	<2000	>2000
Dermal LD50 mg/kg	<50	<200	<1000	<2000	>2000
Inhalation LC50 mg/m^3	<500	<2000	<10000	<20000	>20000

The above discussed toxicity assessment results in three types of substances:

Type 1: (i) Lack of R phrases and acute toxicity data
(ii) ≥ 3 R phrases
(iii) LC50 values from the category 1
Type 2: Two or less R phrases but no acute toxicity data
Type 3: Two or less R phrases and acute toxicity data of category 2 or higher

Type 1 propellants are not considered “green” and are excluded from further assessment. For type 2 propellants information are available to classify them in a first step as “green” but further investigation/assessment would be required to confirm their “green” status. For propellants of type 3, sufficient information is available to classify them with a certain confidence as “green”. The results are summarized in Annex I.

B: General assessment

In a second step in the present assessment, the propellant candidates from type 2 and 3 were further assessed based on their propellant characteristics and performance data. A full assessment of a propellant includes at least the following parameters:

A: Performance	a. High specific impulse b. High specific impulse density	**B: Combustion properties**	c. Readily ignitable d. Small activation energy e. Small time delay f. Self-sustained combustion g. Hypergolicity
C: Liquid properties	h. Low toxicity i. Low freezing point j. High boiling point k. High density l. High specific heat m. High thermal conductivity n. Low vapour pressure o. High decomposition temperature	**D. Others**	p. Simple handling q. Simple transport r. Good storability s. High safety t. Low cost

Obviously an investigation of compliance to so many requirements is even for only one propellant a major task. In view of the number of propellants assessed within the first stage of GRASP, an investigation covering all the parameters is impractical. For a first assessment the following parameters and requirements have been defined:

Table 3: Assessment categories and definitions

Category	Sub-category	Requirement
Toxicity		Defined as a type 3 propellant*
Performance	Specific impulse	Propellants or propellant combinations with a specific impulse equal or higher than 95% of NTO/MMH and hydrazine (monopropellant) respectively are rated positive
	Specific impulse density	Propellants or propellant combinations with a higher specific impulse density than NTO/MMH and hydrazine (monopropellant) respectively are rated positive
Storability	Phase	Propellants which are storable in liquid phase at standard condition (1 bar, 293 K) are rated positive
	Freezing point	Propellants with a lower freezing point 263 K are rated positive
	Boiling point	Propellant with a higher boiling temperature than 313 K are rated positive
Development status		Propellants for which at least a laboratory verification of propulsion characteristics and validation of performance exists are rated positive.

(*) Although type 2 propellants can be considered "green", final classification requires additional data. Therefore this assessment focuses on those propellants for which the "green" status can be declared with the highest confidence (type 3 propellants)

GRASP furthermore assessed and documented the material compatibility, transport regulations, handling properties, and propellant procurement costs of the various propellants. However, while for some of the investigated propellants no information with regard to these issues are available at all, for others only insufficient information or information which were not regarded as reliable (e.g. only internet sources) have been found. For example, while material compatibility is provided for some propellants by the propellant suppliers, they in general do not include detailed information on how such material compatibilities were tested and have to be taken therefore with caution. Although unfortunate, it was therefore deemed appropriate not to publish such information at this stage of the project in its full extent. GRASP intents to assess them at a later stage of the project although not for all of the initially suggested propellants but for a smaller, better manageable number.

To comply with the public nature and mandate of GRASP (financed by the EU via tax payer money), namely to provide the space propulsion community and, in general, the interested public audience with a database about green propellants, the GRASP team members compiled relevant information about the substance, the research background and a data base of pertinent data for all the investigated propellants. The latter includes, performance data (I_{sp}, $I_{sp,d}$), mixture ratio (stoichiometric, optimal), density, viscosity, thermal conductivity, specific heat, surface tension, critical pressure/temperature and others (all together 25 parameters). If not included already in the assessment criteria (see Table 3) those data will be the used by GRASP for further assessments in the next assessment steps but can also be used by other research groups for their individual purpose. The sheer amount of collected data renders it impossible to include it all in the present paper. However, starting from 1st of August, 2009, GRASP will have a dedicated project web-page, http://grasp-fp7.eu, via which those information and data can be accessed.

Performance:

Propellant performance was evaluated using in most cases the Gordon & McBride code. Literature data have been used only for those substances for which no material data base was available in the code. It has to be stressed that most of the provided performance values are of theoretical nature and experimentally obtained values can differ.

For comparison reasons, all propellants have been assessed for the same conditions, i.e. a combustion chamber pressure of 10 bar, nozzle extension ratio of 40, and frozen flow conditions from throat on. With regard to bipropellant systems, based on the oxidizer choices included in this assessment only hydrogen peroxide (H_2O_2) and nitrous oxide (N_2O) are considered to be viable and storable oxidizers. Comparison of the obtainable specific impulse for some fuels shows that with H_2O_2 (90%, wt) an I_{sp} improvement of roughly 3% compared to liquid nitrous oxide can be obtained as shown in Figure 3 (LOX was included for reference purpose). Comparing the specific impulse densities, the advantage of hydrogen peroxide gets more distinctive with an improvement of 13% compared to liquid nitrous oxide. Operating with liquid nitrous oxide comes along with a significant mass penalty since nitrous oxide has to be stored at a tank pressure of >55 bar to ensure its liquid state (somewhat compensated by simplifying the pressurization system). On the other hand, operating with gaseous nitrous oxide results in a severe decrease in obtainable impulse density. Both hydrogen peroxide and nitrous oxide require much large mixture ratios then LOX but at the same time their performance is a weaker function of the mixture ratio and offer therefore certain system advantages not present when using LOX.

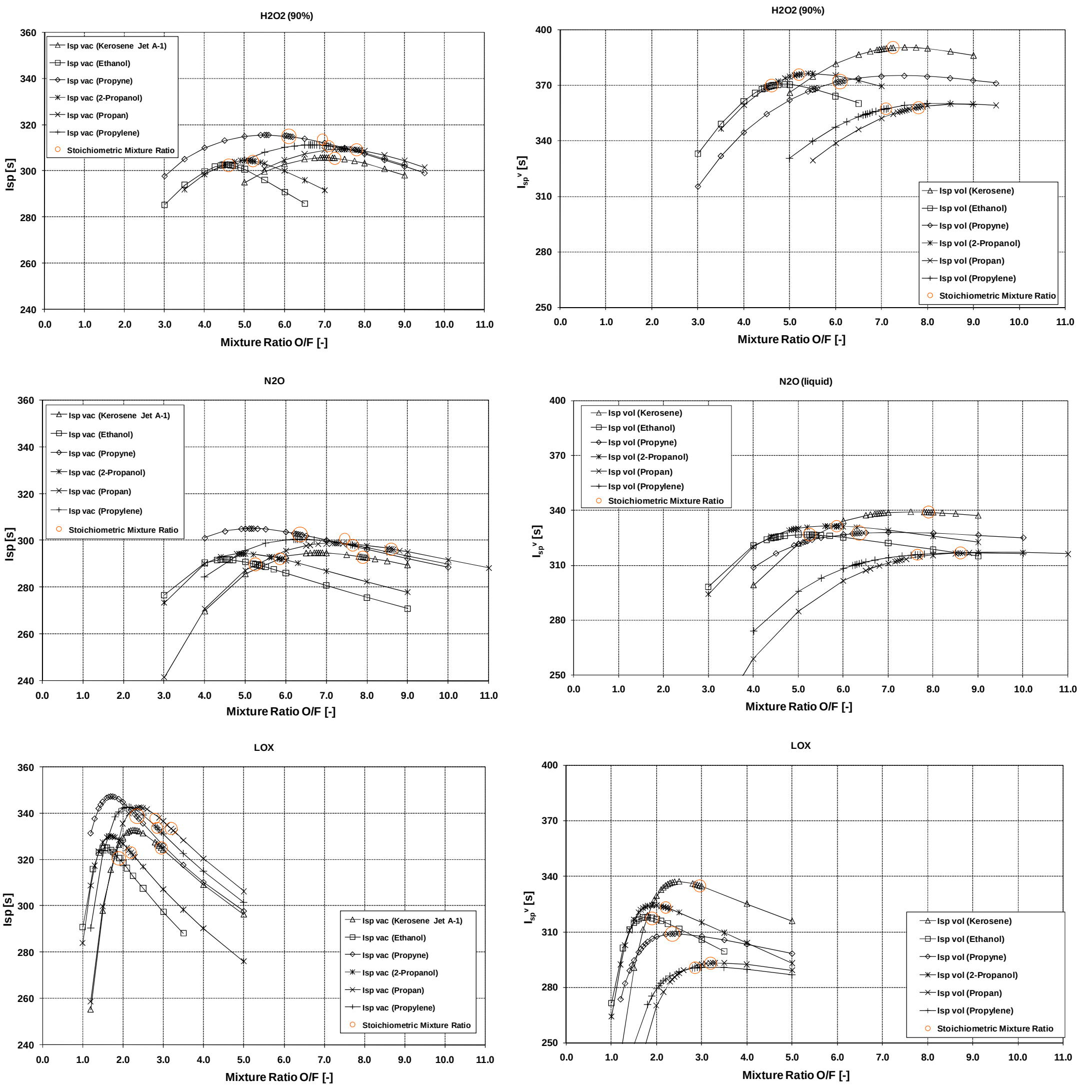

Figure 3: Obtainable specific impulse with H_2O_2, N_2O, and LOX

Considering the above, only the results obtained with hydrogen peroxide (90%, wt) are shown in the following for the bipropellant and hybrid systems.

As shown on the left side of Figure 4, a good number of assessed monopropellants offer increased specific impulse ranging from 10% to nearly 30%. Considering the higher propellant density for many of the propellants, the impulse density gain ranges between 20% up to 80%. Similar results are obtained for hybrid systems when operated with hydrogen peroxide (90%, wt) (Figure 4, right). Various ionic liquids are in particular very promising alternatives to hydrazine (in Figure 4, left, with solid HAN and HNF for comparison) . The obtainable specific impulse for those monopropellants nearly reaches levels usually only seen in bipropellant systems but offer all the systems advantageous of a monopropellant, e.g. only one propellant feeding system.

The results for bipropellant systems indicate that in terms of specific impulse only a marginal number of propellants show the same or better performance than the reference propellant combination NTO/MMH (see Figure 5). Most investigated propellant combinations suffer a specific impulse decrease between 2.5% and 6%. On the other hand, for many of those propellant combinations the impulse density is between 5% and 6% higher than for NTO/MMH. The latter can be considered as an advantage but since most missions are driven by a Δv requirement, the real advantage derived from an increased impulse density has to be investigated based on a case-to-case basis.

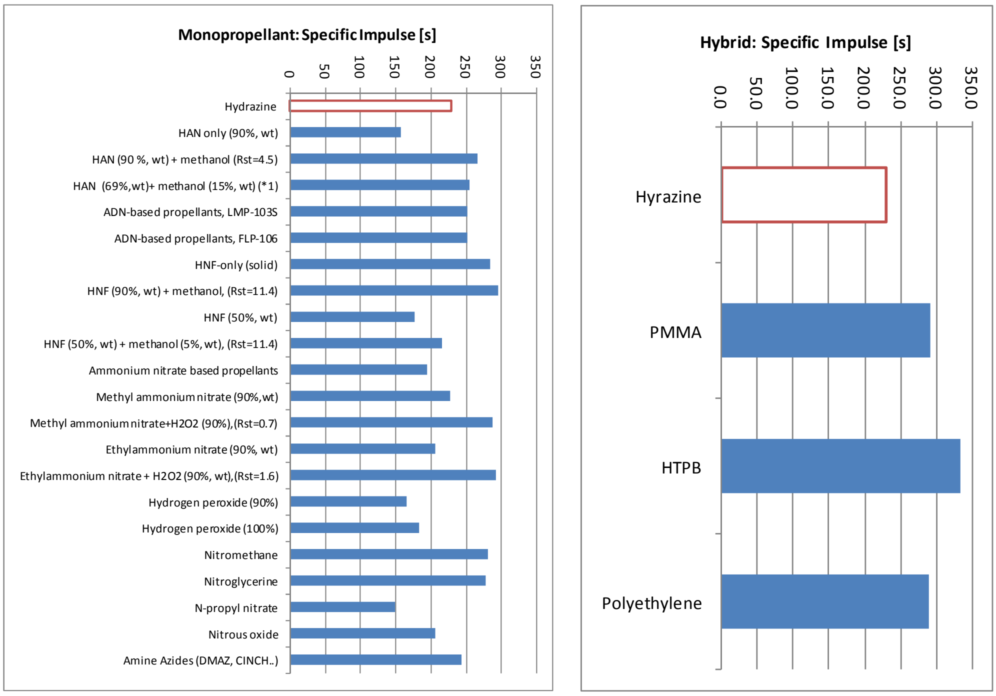

Figure 4: Specific impulse for green monopropellants and hybrids in comparison with toxic reference propellants

<u>Storability</u>:

One of the major concerns for a propellant is its storability. Storability depends on many characteristics including the physical state of the propellant (solid/liquid/gaseous), minimum/maximum storage temperature, general stability, sensitivity to light, heat, temperature variation, contamination, static discharges, polymerization tendencies and many others. As a first step the required temperatures to store the propellants in their liquid phase were assessed. Figure 6 shows the temperature window of the various propellants (fuel and oxidizer) for which reliable data were obtainable. Many of the assessed green propellants offer a much broader temperature window than the toxic reference propellants. Since this has potentially a significant effect on the propellant feeding system (relaxation of insulation or propellant heading requirements) it is considered an important advantage of the assessed green propellants.

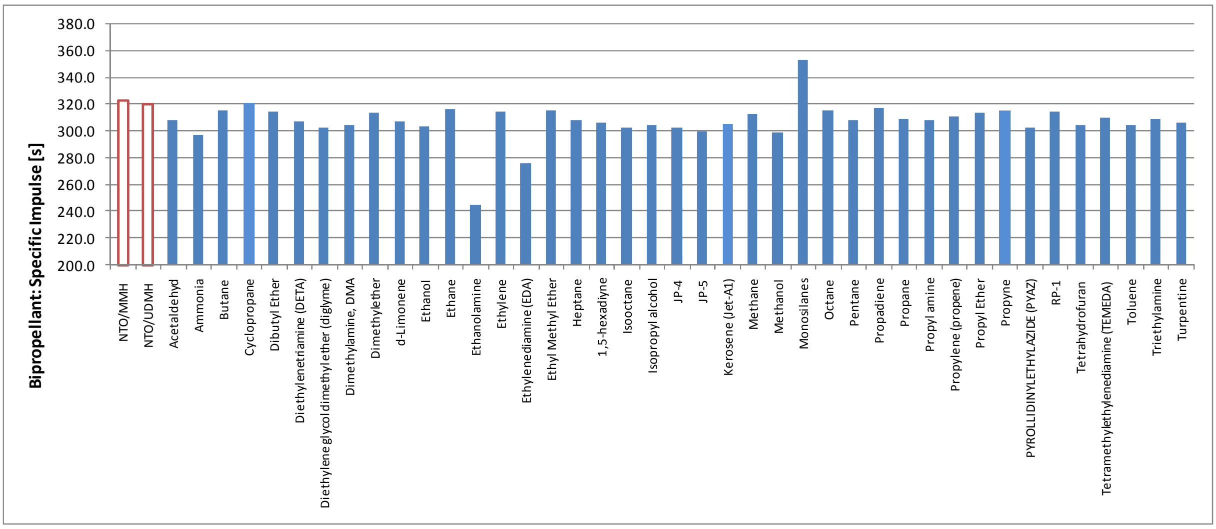

Figure 5: Specific impulse for green bipropellant fuels and 90% hydrogen peroxide in comparison with toxic reference

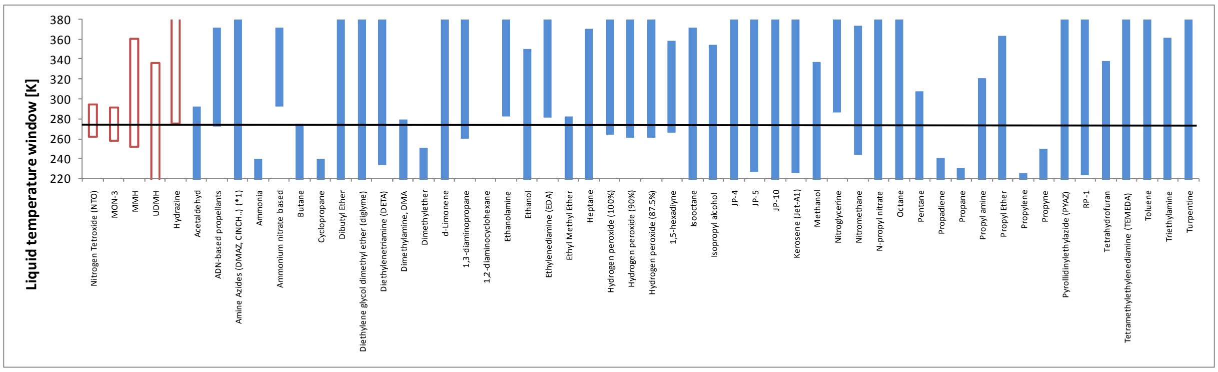

Figure 6: Temperature window for liquid storage

<u>Development Status</u>:

The GRASP team compiled propellant information including background information to the individual propellants in particular its history with regard to its use/investigation as a rocket propellant (those information sheets are not included in the present paper). Based on the compiled information a simple ranking has been established with regard to their development status. The following four categories have been used to rank them:

Table 4: Development status ranking

Criterion	Rating
Sufficient availability of substance/propellant properties	1
Laboratory verification of propulsion characteristics and validation of performance	2
Existing propulsion system and/or significant level of maturity	3
Existing space heritage	4

Based on this ranking one can estimate if a propellant can be considered for a near-term usage (<10 years) or if its possible usage might be further in the future. All propellants which have reached a development status of 2 are considered possible candidates for a near-term application.

V. Assessment results

Based on the requirements shown in Table 3 the final results of this initial assessment are depicted visually in Figure 7 to Figure 9. Green fields indicate compliance with the criteria as defined in Table 3, red fields the opposite. White fields indicate that the information is not available. For comparison purposes the reference propellant, hydrazine for monopropellants and hybrids, and NTO/MMH for bipropellants, is given in each figure.

The results for monopropellants depicted in Figure 7 show clearly that several monopropellants exists which offer equivalent or better performance than hydrazine. Their storability characteristics in terms of temperature window is in many cases superior to hydrazine and their development status suggests a possible application in less than 10 years. Most important, they all offer a significant reduction in toxicity, implying a recognizable increase in handling safety and a reduction in costs (handling, transport, waste treatment etc.). Even more advantageous results are shown in Figure 8 for hybrid systems. Toxicity and storage characteristics are both superior compared to hydrazine. The theoretical calculations also suggested significantly improved performance. However, it has to be stressed again that most of the performance data provided here (mono-, bipropellant, hybrids) are of theoretical nature. For example, in the case of hybrids it is well known that issues with regression rates etc. can have a severe impact on the actual values obtained in experiments resulting in lower values than suggested by theoretical calculations.

As expected the results for bipropellant systems are much more complex than for monopropellants. The choice for oxidizers for bipropellant systems is very limited. With regard to the oxidizers assessed within GRASP, liquid oxygen offers the highest performance but is, due to its cryogenic nature, not considered to be a storable propellant and therefore not suitable for in-space propulsion (which is the main focus of GRASP).

Further oxidizer options assessed within GRASP are blends of ionic liquids with water. However, the expected performance of such blends is either lower than that of nitrous oxide and hydrogen peroxide or the blend has a poor storability in terms of allowable storage temperatures.

Based on the present assessment, this leaves only nitrous oxide and hydrogen peroxide as possible alternatives for a replacement of currently used toxic oxidizers (e.g. NTO). Both propellants have their merits and disadvantageous. Nitrous oxide has lower performance in terms of obtainable specific impulse but in particular in terms of specific impulse density even if stored as a liquid. Its high vapor pressure requires pressurized storage at roughly 55 bar to ensure storage in its liquid phase, therefore detrimentally impacting the propellant feeding system.

Hydrogen peroxide can be stored as liquid for a relative large temperature range (much higher boiling temperature than that of NTO and MON-3). Its performance exceeds that of nitrous oxide in terms of specific impulse and, due to its high density, also in terms of specific impulse density. Storability has been proven for both ground and space conditions.

Material compatibility and long term storability are surely two issues which have to be investigated for both oxidizer alternatives. Other than nitrous oxide, hydrogen peroxide has space heritage. However, with regard to the fact that this heritage is more than 30 years old, renewed efforts have to be initiated to confirm its performance, but in particular its material and COTS compatibility, again.

Based on Figure 9, the fuels which have been assessed as most beneficial (only one negative (red)) include:

- D-limonene
- Ethanol
- Isopropyl alcohol
- JP-4
- JP5
- Jet-A1
- Octane
- Propyl amine
- propyl ether
- PYAZ
- RP1
- Tetramethylethylenediamine
- Turpentine

JP4, JP5, RP-1, and PYAZ are propellants with very limited availability in Europe and are therefore excluded in the following discussion.

All of the above remaining fuels have a confirmed significantly reduced hazard values and for the larger part they can be handled using gloves and goggles only. They have, when combusted with hydrogen peroxide (90%, wt), a specific impulse decrease in comparison with NTO/MMH ranging from 2.5% to 6.2% (based on theoretical calculations), which is to a certain degree counterbalanced by their improved specific impulse density characteristics. All of them can be stored in their liquid phase in a wider and more advantageous temperature range than any toxic reference fuel.

High purity d-Limonene, octane, propyl ether, and tetramethylethylenediamine seem to be excessively expensive. With the exception of octane, the procurement costs for those propellants range between a factor 5 and 16 times the price for MMH or UDMH (281 €/kg[36]). If those procurement costs are verified, a switch to such propellants might be rather improbable. In addition the development status of those propellants is rather low requiring extensive development efforts. The procurement costs for ethanol, isopropyl alcohol, kerosene (Jet-A1), and turpentine are very low. The most expensive one, propyl amine, is still a factor of 4 times cheaper than MMH or UDMH. In addition, the development status for ethanol, Jet-A1, and isopropyl alcohol was assessed to be relatively high.

In summary, based on this initial assessment, ethanol, isopropyl alcohol, kerosene, and turpentine appear to be the most favorable green propellants from the list of assessed bipropellant fuels.

Dibutyl ether, diethylenetriamine, heptanes, and triethylamine were assessed as only "possibly low in toxicity", i.e. for those propellants the toxicity information was not as sufficient as for the others. If more toxicity information would be available and they could be categorized as "probably low in toxicity" those fuels would have also very advantageous properties. They have large temperature windows for storage, performance values similar to the above ones, and the procurement costs are roughly 10% of MMH and UDMH. In conclusion, although all of them have a low development status, if the toxicity of those propellants can be elucidated and found sufficiently low, then they qualify for further investigation.

Butane and Dimethylether are both considered to have a very low toxicity and promise good performance in terms of specific impulse (315 s and 316 s). Other assessment results for those two propellants are less advantageous since they require pressurized tank vessels in order to store them in liquid phase. However, in order to store them in liquid phase, butane and dimethylether require a storage pressure of a minimum of 2 bar and 4.4 bar respectively. This might be considered sufficiently low, in view of their relatively high specific impulse, to be considered acceptable.

Last, but not least, two further fuels shall be especially mentioned: methane and silanes. Methane has found its entry in many projects and propulsion developments, in particular as a replacement for liquid hydrogen. Here it offers improved specific impulse densities and its cryogenic nature is of less concern considering it replaces another cryogenic fuel. Considering the above and its high development status, methane is considered by a majority of the space propulsion community to likely take over some of the missions formerly occupied by liquid hydrogen.

The performance of monosilane is very favorable. As a matter of fact, monosilane exceeds the specific impulse of all toxic fuels and all the assessed green propellants by a large margin. However, the assessment pointed out the disadvantageous in terms of storability due to its gaseous phase. Although this is true, higher forms of silanes, starting with trisilanes, can be stored in their liquid phase at standard conditions. The development status of these higher silanes is rather low and many issues require significant efforts before those higher order silanes can be considered to be valuable fuels for space propulsion applications. It will therefore be further investigated within GRASP.

Monopropellants Comparison with Hydrazine							
	Toxicity	**Performance**		**Storage**			**Development status**
Propellant	Probably low in toxicity	Isp	Ispd	Phase	Freezing	Boiling	>2
Reference: Hydrazine		230	232	l	275 K	387 K	4
HAN-based							
ADN-based, LMP-103S							
ADN-based, FLP-106							
HNF-based propellants							
Ammonium nitrate based							
Methyl ammonium nitrate based							
Hydrogen peroxide (90%)							
Hydrogen peroxide (100%)							
Nitromethane							
Nitroglycerine							
N-propyl nitrate							
Nitrous oxide							
Amine Azides (DMAZ, CINCH..)							

Figure 7: Assessment results for monopropellants

Hybrid Comparison with Hydrazine							
	Toxicity	**Performance**		**Storage**			**Development status**
Propellant	Probably low in toxicity	Isp	Ispd	Phase	Freezing	Boiling	>2
Reference: Hydrazine		230	232	l	275 K	387 K	4
PMMA							
HTPB							
Polyethylene							

Figure 8: Assessment results for hybrids

Bipropellants Comparison NTO/MMH with H2O2 (90%)/fuels							
	Toxicity (fuel)	**Performance (H2O2/fuel)**		**Storage (fuel)**			**Development status**
Propellant	probably low in toxicity	Isp	Ispd	Phase	Freezing	Boiling	≥ 2
Reference: NTO/MMH		323 s	372 s	liquid	252 K	360 K	4
Acetaldehyd							
Ammonia							
Butane							
Cyclopropane							
Dibutyl Ether							
Diethylenetriamine (DETA)							
Diethylene glycol dimethyl ether (diglyme)							
Dimethylamine, DMA							
Dimethylether							
d-Limonene							
1,2-diaminocyclohexane							
1,3-diaminopropane							
Ethanol							
Ethanolamine							
Ethane							
Ethylene							
Ethylenediamine (EDA)							
Ethyl Methyl Ether							
Heptane							
Hydrogen							
1,5-hexadiyne							
Isooctane							
Isopropyl alcohol							
JP-4							
JP-5							
Kerosene (Jet-A1)							
Methane							
Methanol							
Monosilanes							
Octane							
Pentane							
Propadiene							
Propane							
Propyl amine							
Propylene (propene)							
Propyl Ether							
Propyne							
PYROLLIDINYLETHYLAZIDE (PYAZ)							
RP-1							
Tetrahydrofuran							
Tetramethylethylenediamine (TEMEDA)							
Toluene							
Triethylaluminium							
Triethylamine							
Turpentine							

Figure 9: Assessment results for bipropellants

VI. Conclusion

GRASP strives to provide the industry with the necessary information and propellant data background to assess the potential and the impact of a replacement of the commonly used toxic propellants (e.g. NTO, hydrazine and its derivates). For this purpose, a comprehensive information and database for green propellants, their properties and their performance was established.

The market analysis has shown that although a significant large market for propulsion system exists, up to this date no propulsion system relying on green propellants is commercially available. Partially this is due to the existing toxic systems which offer reliable performance and have a large space history. A switch to a new technology is associated with relative large costs and risks. It is assumed that near term application will focus on small satellites and science mission. Only when sufficient space history has been collected a broader application of green propellants is expected. Special interest in green propellants could come for so-called emerging players, i.e. small start-up companies which are not yet burdened with an existing propellant infrastructure but also small research companies and academia (e.g. CubeSat and microsatellite producers).

However, in case the legal environment in the respective countries becomes more restrictive in terms of handling toxic propellants, the major players in the propulsion industry might have to react fast to ensure their share of the market and the above predictions might be extended to a much larger market.

An initial assessment of propellant candidates has identified those propellants which GRASP considers to be "green", i.e. offer a reduced toxicity level. Considering the difficulties with the toxicity assessment in general, GRASP can only provide suggestions. It is the sole responsibility of the individual entities to ensure the safety of their facility and personnel.

Following the toxicity assessment, an extensive database (including 25 performance and material property data sets) has been established for all the candidates which are considered to be green. Based on those data a further down-selection has been conducted providing a final choice of propellants GRASP considers to be viable green propellant candidates. Considering the amount of investigated propellants, this assessment cannot capture certain details and GRASP acknowledges that some of the propellants which were excluded might perform well in a more global assessment. All recommendations of GRASP have to be seen in the context of the applied criteria.

Based on the choice of propellants provided in the present paper, GRASP will focus its effort on those to extend and fine-tune the data base for those propellants and initiate propellant and catalyst tests. The test results will provide the basis for a further down selection. In a final step the propulsion systems for a small choice of green propellants will be developed and tested. GRASP aims to provide the industry and the interested community at the end of the project with a choice of real alternatives for the presently used toxic propellants.

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Starting from 1st of August, 2009, the GRASP project will have a webpage dedicated to the GRASP project: http://grasp-fp7.eu. The webpage will also contain the information and database to which this paper referred several times. Interested parties will be able to download this database. GRASP also lives from input from the community in general. Therefore the GRASP web page will be interactive, i.e. if information/data are missing, or if the viewer considers parts of them inaccurate, they can suggest new data. If those data are provided together with a relevant reference they will be used to update the GRASP database.

VII. Nomenclature

ACS	Attitude Control System	OFASV	Orbital Facility Assembly and Service Vehicles
COTS	Cost effective of the shelf	OSHA PEL	Permissible exposure limit [ppm]
EU	European Union	OSHA STEL	Short-term exposure limit [ppm]
GRASP	Green Advanced Space Propulsion	PMMA	Poly(methyl) methacrylate
GSE	Ground Support Equipment	SCAPE	Self Contained Atmospheric Protective Assembly
HTPB	Hydroxyl-terminated polybutadiene		
IRCH	Total Hazard Score	TLV	Treshold level value
LC50 (inhalation)	The LC_{50} is the concentration in air over a period of four hours associated with a 50% chance of death within two weeks	TEHF	Toxicological and Environmental Hazard Figure
LD50 (oral)	The oral LD_{50} is the single dose associated with a 50% chance of death within two weeks [mg/kg]	TWA	time-weighted average
LD50 (dermal)	The skin (dermal) LD_{50} is the single dose associated with a 50% chance of death within two weeks [mg/kg]	R Phrases	Risk phrases
MMH	monomethyl hydrazine	UDMH	Unsymmetrical dimethylhydrazine
MON-3	Diniotrogen tetroxide + 3% (wt) nitric oxide		
NIOSH IDLH	Immediately dangerous to life or health [ppm]	I_{sp}	Specific impulse [s]
NIOSH REL	Eight-hour recommended exposure level [ppm]	$I_{sp,d}$	Specific impulse density [s]
NTO	Nitrogen tetroxide, N_2O_4		

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Annex 1: Toxicity assessment of green propellant candidates

Propellants	Type	Comments	Oral [LD50] [mg/kg]	Inhalation [LC50] [ppm/4h]	Dermal [LD50] [mg/kg]	Vapour presure (Pa)	LD50 (oral) EU category	LC50 (oral) EU category	LD50 (dermal) EU category
Toxic storable propellants									
Hydrazine	1	rejected - EU Category 1 Inhalation Hazard	60	330	91	1917	3	1	2
MMH	1	rejected - EU Category 1 Inhalation Hazard	32	34	95	4800	2	1	2
MON-3	2					120658			
UDMH	1	rejected - EU Category 1 Inhalation Hazard	122	252	1060	14500	3	1	4
Monopropellants									
ADN-based propellants	3		1290		3000	800	4		5
Amine Azides (DMAZ, CINCH..)	3	no R phrase data	967	2000	NA		4	3	
Ammonium nitrate based propellants	3			2000		<1		3	
Ethylene oxide		rejected, adverse r phrase score of 3	72	800		140000	3	2	
Ethyl nitrate	1	rejected - EU Category 1 Inhalation Hazard				8531			
HAN-based propellants	3					<1			
He		regarded as a non-toxic asphyxiant				--			
HNF-based propellants	3					<1			
Hydrogen peroxide (87.5%)	3		805	2876	2000	375	4	3	5
Methyl ammonium nitrate	3					<1			
N2		regarded as a non-toxic asphyxiant				--			
N4 and other Nx	1	rejected, insufficient data, no R phrase data							
Nitrous oxide	3			1068		5059900		2	
Nitromethane	2		940		2000	3700	4		5
Nitroglycerine	2		105		280	0.03	3		3
N-propyl nitrate	3	no R phrase data		10000		5332		4	
Propane	2					830000			
Tetranitromethane	1	rejected - EU Category 1 Inhalation Hazard	130	18		1066	3	1	
Bipropellants oxidizer									
ADN/water	3								
HAN/water	3								
Hydrogen peroxide	3		805	2876	2000	375	4	3	5
Nitrous oxide	3			1068		5059900		2	
Oxygen (O2)	3								
Tetranitromethane	1	rejected - EU Category 1 Inhalation Hazard	130	18		1066	3	1	
Bipropellants fuels									
Acetaldehyd	3		661	13300	3540	100646	4	4	5
Amine Azides (DMAZ, CINCH..)	3	no R phrase data	967	2000	NA		4	3	
Ammonia	3		350	2000		28790	4	3	
Aniline	1	rejected - EU Category 1 Inhalation Hazard	250	250	820	100	3	1	3
Allyl-dipropinylamine	1	rejected, insufficient data, no R phrase data							
AFRL-4	1	rejected, insufficient data, no R phrase data							
Bicyclopropylidene	1	rejected, insufficient data, no R phrase data							
Butane	3			272000		212793		5	
C-stoff	1	rejected, insufficient data, no R phrase data							
Cyclopropane	2								
Diethylenetriamine (DETA)	2		1080		1090	29	4		4
Diethylene glycol dimethyl ether (diglyme)	3		4760	11000		266	5	4	
Dimethylamine	3		698	4540		170000	4	3	
Dimethylether	3			164000		424000		5	
Dibutyl Ether	2		11000		10000	639	5		5
d-Limonene	3		4400	45430	6100	266	5	5	5
1,3-diaminopropane	2		308		176	700	4		2
3,3'-diaminodipropylamine	1	rejected - EU Category 1 Inhalation Hazard	738	6	103	5	4	1	2
1,2-diaminocyclohexane	2		4556			510	5		
2,5-Dimethyltetrazole	1	rejected, insufficient data, no R phrase data							
Ethylene	3		NA	5640	NA	over critical point		3	
Ethanol	3		7060	46500	20000	6580	5	5	5
Ethane	2					3740000			
Ethyl Methyl Ether	2								
Ethylene oxide	1	rejected, adverse R phrase score of 3	72	800	187	146000	3	2	2
Ethanolamine	2		1720	213	1025	58	3	2	2
Ethylenediamine (EDA)	3		500	10808	560	1426	4	4	3
Furfuryl alcohol	1	rejected - EU Category 1 Inhalation Hazard	160	233	400	53	3	1	
Heptane	2		3200			5300	5		
Hydrogen (H2)		regarded as a non-toxic asphyxiant							
1,5-hexadiyne	3								
Isooctane	3		5000	3078	2000	4235	5	3	5
Isopropyl alcohol	3		5045	17500	12500	4860	5	4	5
JP-1	1	rejected, insufficient data, no R phrase data	25000		5000	200	5		5
JP-3	1	rejected, insufficient data, no R phrase data							
JP-4	3		25000		5000	13790	5		5
JP-5	3		25000		5000		5		5
JP-10	2		18800	1194			5	2	
Kerosene (JetA-1)	3		5000	NA	2000	10000	5		5
Methane (CH4)	3			250000		over critical point		5	
Methanol	3		5628	64000	15800	16900	5	5	5
Monomethylamine	1	rejected - EU Category 1 Inhalation Hazard	100	360		37200	3	1	
Monosilanes	3	no R phrase data		9600	3540	over critical point		3	5
Octane	3		10000	24895	3400	1330	5	5	5
Pentane	3			121000	446	57089		5	3
Propyl Ether	3			38000		7731		5	
Propane	3		NA	5000	NA	830000		3	
Propyne	2		N/A	N/A	N/A	524483			
Propylene	3		N/A	253444	N/A	1068240		5	
Propyl amine	3		370	2310	560	3300	4	3	3
Propadien	2								
3-Prop-2-ynyloxy-propyne	1	rejected, insufficient data, no R phrase data							
RJ-5	1	rejected, insufficient data, no R phrase data							
RP-1	3					2000			
Tetramethylethylenediamine (TEMEDA)	3		268	1318	5390		3	2	5
Tetrahydrofuran	3		3000			17473	5		
Toluene	3		636	8000	14100	2924	4	3	5
Triethylaluminium	3					133			
Triethylamine	2		460		570	6898	4		3
Trimethylaluminium	2								
Tris(azidomethyl)amine	1	rejected, insufficient data, no R phrase data							
Tri-prop-2-ynyl-amine	1	rejected, insufficient data, no R phrase data							
Turpentine	3		5760	29000	5010	76900	5	5	5
Xylidine	1	rejected - EU Category 1 Inhalation Hazard	467	149		1426	4	1	
Hybrid fuels									
HTPB	3		not applicable	not applicable	not available	not applicable			
PMMA	3		not applicable	not applicable	not available	not applicable			
Polyethylene	3		not applicable	not applicable	not available	not applicable			