Development of European Grease Lubricants Stabilized for Long-Term Storage

Executive Summary Report

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Figures

Tables

1 Introduction

1.1 Purpose

This document is the summary report of the project "Development of European Grease Lubricants Stabilized for Long-Term Storage" (filed under ESA contract No. 4000137211/21/NL/KML/rk). In this document, the main project findings are summarised concisely.

1.2 Applicable documents

The following documents are applicable:

[AD 1]	ESA contract: 4000137211/21/NL/KML/rk
[AD 2]	ESA contract No. 4000137211-21-NL-KML-rk_1.0_TN1 Literature Market overview definition of requirements and formulation strategy
[AD 3]	ESTL document DW-004209-01B: "SOT Sample Disk Drawing"
[AD 4]	ESTL document DW-004210-01B: "SOT Guide Plate Drawing"
[AD 5]	ESTL document PRA-ESTL-PR-7508 02: "Grease Lubrication of Components"
[AD 6]	ESA contract: 4000137211/21/NL/KML/rk ; TN1 report
[AD 7]	ESA contract: 4000137211/21/NL/KML/rk ; TN2 report
[AD 8]	ESA contract: 4000137211/21/NL/KML/rk ; TN3 report
[AD 9]	ESA contract: 4000137211/21/NL/KML/rk ; TP1 report

1.3 References

The following documents are referenced:

[RD1]	Senese S, Strube B, Lattner K, Fusco G, Deuschle T, Forster U, Thiel M, Svedevall A, Toledo A, A, Sghedoni M et al.; "Meteosat Third Generation (MTG) Mechanisms: Impact on Instrument Performances"; 18th European Space Mechanisms and Tribology Symposium (Munich, Germany) 2019
[RD2]	A. Wolfberger et al.; "Assessment of the Chemical Degradation of PFPE Lubricants and Greases for Space Applications: Implications for Long-Term On-Ground Storage"; CEAS Space Journal 2021
[RD3]	M. Buttery et al.; "Long-Term Storage Considerations for Spacecraft Lubricants"; Lubricants 2020
[RD4]	NASA; "Braycote Grease Retains Tribological Properties for 17 Years in Controlled Storage"; NASA Engineering and Safety Center Technical Bulletin No 07-01 2007
[RD5]	Buttery, M et al; "Fomblin Z25: A New Method for its Degradation Assessment & Proposal for Safe Operation in Space"; 15th European Space Mechanisms & Tribology Symposium – ESMATS 2013
[RD6]	ESA-ESTL-TM-0194 01: "The Performance of Oils and Grease after Bakeout"; ESTL 2017
[RD7]	ESA-ESTL-TM-0264 01: "SOT Assessment of Pre-Existing Fluid Samples for LTS"; ESTL 2021
[RD8]	ESA-ESTL-TM-0181 01: "Long Term Storage and Cage Impregnation"; ESTL 2018
[RD9]	ESA-ESTL-TM-0131 01: "Fluid Lubricant Behaviour at Cold Temperatures"; ESTL 2014
[RD10]	ECSS-Q-ST-70-02C: "Space Product Assurance – Thermal Vacuum Outgassing Test for the Screening of Space Materials"; ESA-ESTEC 2008
[RD11]	A. Kent et al., "Guidelines for Qualification of New Fluid Lubricants", Conference paper ESMATS 2023.
[RD12]	ESA-ESTL-TM-0375 01: Guidelines for Qualification of New Fluid Lubricants , ESTL 2023
[RD13]	Lu, W. et al; "Catalytic Decomposition of Perfluoropolyether Lubricants" ; IEEE Transactions on Magnetics 2003
[RD14]	ESA-ESTL-TM-0102 01: 'Characterisation of Degradation of PFPE Lubricants', ESTL
[RD15]	ESA-ESTL-TM-0330 01: 'New Mass Spectrometric Techniques for Lubricant Analysis", ESTL
[RD16]	ESA-ESTL-TM-0089: "Further Spiral Orbit Tribometer Assessments of Space Greases", ESTL
[RD17]	ESA-ESTL-TM-0190: "Life of Space Greases in Air", ESTL

1.4 Abbreviations and definitions

ACE	Auto Catalytic Effect
ASTM	American Society for Testing and Materials
CDR	Critical Design Review
CVCM	Collected Volatile Condensable Material
DIN	Deutsche Industrienorm
DLR	Deutsches Zentrum für Luft- und Raumfahrt
ECSS	European Cooperation for Space Standardization
ESTL	European Space Tribology Laboratory
EU	European
FSR	Formulation Screening Review
GEO	Geostationary Earth Orbit
HSE	Health, Safety and Environmental Constraints
LEO	Low Earth Orbit
LLI	Long Lead Item
LTS	Long Term Storage
LVEB	(Regime of) Linear Viscoelastic Behavior
MAC	Multiply Alkylated Cyclopentanes (oil)
MAT	Functionality of Materials
MIS	Mission Constraints
MOS	MetOp Second Generation
MTG	Meteosat Third Generation
NLGI	National Lubricating Grease Institute (US)
OTH	Other Requirements
PDR	Preliminary Design Review
PFPE	Perfluoroalkylpolyether
PoD	Pin on Disc (Tribometer)
PTFE	Polytetrafluoroethylene
r.h.	Relative Humidity
REACH	Registration, Evaluation, Authorisation and Restriction of Chemicals
RML	Recovered Mass Loss
RPM	Revolutions per minute
SADM	Solar Array Drive Mechanism
SoW	Statement of Work
SRR	System Requirements Review
TGA	Thermal Gravimetric Analysis
TML	Total Mass Loss
TRI	Tribological Requirements
TRL	Technology Readiness Level
VOC	Volatile Organic Compounds
WP	Work Package

Space Lubricants for Long-term Storage Application

Controlled vacuum environments as in space applications represent a challenge for the lubrication of tribological components. Besides common space lubricant requirements like e.g. low evaporation, a broad operational temperature range and a high stability during operation, long-term storage properties (LTS) have gained increasing attention recently. The term addresses the time-dependent stability of a lubricant under static conditions, which can mean chemical degradation processes such as oxidation on the one hand, but also the physical separation of oil and thickener in heterogeneous lubricants like greases. Due to extended storage periods of lubricated components on-ground but also during a space mission for several years, it has to be ensured that a lubricant is still functional after LTS. For instance, for the MTG (Meteosat Third Generation) mission, 5-year assembly, integration and testing (AIT) time on ground, up to 17.5-year storage for recurring models plus at least 8.5 years in orbit operation are planned [RD1]. Therefore, the understanding of the impact of LTS on tribological performance of lubricants [RD3], but also the actual LTS behavior of lubricants have gained attention recently. In a current study (funded under ESA-ESTEC Contract No 4000120585/17/NL/MP) the real-time LTS behavior of numerous spacecraft mechanism lubricant elements is investigated, and will be concluded in 2037. In a recent NASA study, the retainment of tribological properties of Braycote 601 EF (Castrol) after 17 years of storage was presented [RD4]. In a publication by Henry et al. artificially aged and 17 year-old Braycote 601 EF were analyzed with different methods, despite of small changes the high stability of the grease as proposed in the NASA paper was confirmed.

In this activity (ESA contract No 4000137211/21/NL/KML/rk), a European-based space lubricant grease with LTS properties was developed. Therefore, requirements that apply to a space lubricant on the one, and to LTS on the other hand were analyzed and specified. For space application, the performance of a lubricant is of highest importance for reliability of mechanisms. Many parameters need to be understood and specified, the most important ones being

- Viscosity
- Outgassing
- Vapor pressure
- Tribological performance
- Compatibility with construction materials

LTS was defined as a stability under cleanroom storage conditions for at least 15 years. It is well-known that lubricants being in operation can change in their properties over time, which can severely impact the functionality of the tribological system. However, fluid lubricants can be susceptible to effects that impact their properties also during storage:

- Oxidation
- Evaporation
- Chemical degradation
- Thermal degradation

- Lubricant creep
- Phenolic cage fluid loss
- Separation of greases

On the basis of this list of parameters, the development of a space grease formulation with LTS properties and its comprehensive characterization was planned and conducted.

Formulation development

Fluids used for space application typically consist of either of the following base oils types – perfluoropolyether (PFPE) or multiple alkylated cyclopentanes (MAC). The most common filler systems for such greases are polytetrafluoroethylene (PTFE) or soap-based thickeners.

The selection of base oil was mainly influenced by LTS requirements. It is known that MAC-based fluids can be susceptible to oxidation. Although little information about the oxidation stability of such oils in long-term storage at room temperature exists, the limited shelf lives provided by manufacturers is an indicator that an inhibition of oxidation over multiple years of storage cannot be ensured. Otherwise, PFPE lubricants exhibit a very high inertness due to their chemical structure [RD2]. Tests with PFPE fluids stored in uncontrolled office environments for more than 40 years confirm this [RD7]. The loss of fluid lubricant through evaporation is another area of concern rather for MAC than for PFPE, due to the oil itself but also due to stabilizing additives being used. As LTS typically takes place in laboratory air or DN_2 environments, the subsequent loss of additives under vacuum may not be a significant concern, however the overall evaporation risk of MAC oils that possess higher vapor pressures is higher than for PFPE oils.

While PFPE are in general more inert and stable than MAC, the auto-catalytic degradation in the presence of Lewis acids (such as FeF_3) makes them highly susceptible to chemical degradation during operation on ferrous materials. This degradation is usually understood to occur during operation (i.e., shearing), however to our knowledge it has not been proven that the species that propagate degradation become totally inactive when the motion is stopped. This can potentially be critical with periods of in-flight inactivity during missions with long cruise phases. To date there is limited experimental data to confirm this risk [RD5], but it however exists as a theoretical possibility.

The analysis of the different parameters potentially affecting LTS came to the conclusion that physical grease separation is expected to be most likely aspect, which can be critical to grease functionality.

Based on the above considerations it was decided to develop a formulation with a PFPE fluid. While it is difficult to assess the discussed parameters in the context of LTS, it is known that PFPE are extremely inert structures with very low vapor pressures, and therefore the risk of a chemically induced LTS failure is expected to be very low.

For the choice of PFPE base oil, several commercially available fluids were reviewed and assessed regarding their suitability for space application. It was decided to develop a blend from two base oils, the Z-type PFPE Fomblin Z25 and the Y-type PFPE Fomblin YR1800. Both display excellent outgassing properties. While Fomblin Z25 possesses a very advantageous viscosity and viscosity index, Y-type PFPE are known to be more resistant against chemical degradation by lewis acids. Different compositions were investigated, it was the target to achieve a high Y-type oil content not only due to chemical stability but also with the goal of a high viscosity, since the separation of dispersions from oils with higher viscosity proceeds more slowly. A blend from Z25/YR1800 67:33 was selected, in order to remain below a viscosity of 400 CSt (at 20 °C). This was defined as an upper specification limit in order to prevent the risk of high torques in application.

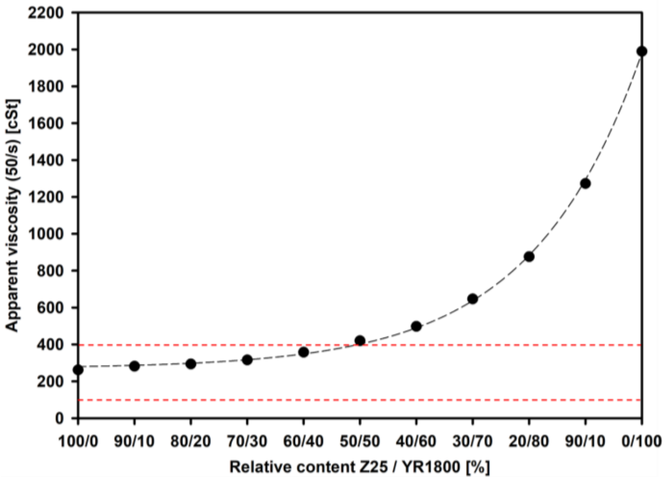

Figure 1: Oil viscosity of blends from Fomblin Z25 and Fomblin YR1800 at various compositions. The red lines indicate the upper and lower specification limits

PTFE was favored as thickener over soap-based systems due to the high chemical inertness and known beneficial outgassing properties. It is common to use micropowders with average particle sizes in the range of 1 µm or below for the production of grease. It is known from the theory of dispersions that small particles display a better stabilization in fluid, as stabilization mechanisms are more effective and Van der Waals attraction energy decreases with particle size. Several aspects such as particle shape and roughness or surface energy further impact the dispersion stability of powder. As only limited information is available for commercial powders, several PTFE types were tested and compared.

First, simple tests were performed with dilute dispersions. A small amount of PTFE was added into oil and ultrasonicated, then the settling behavior was observed over time. Significant differences were obtained. Secondly, greases with 25% PTFE content were rolled with an ointment mill and analyzed. Also here, obvious differences could be found, some of the powders were not even capable to build a stable grease. Oil separation tests were performed with selected candidates.

Overall, results from tests with oil dispersions and greases were very consistent and conclusive, so that a PTFE powder for the formulation development could be selected.

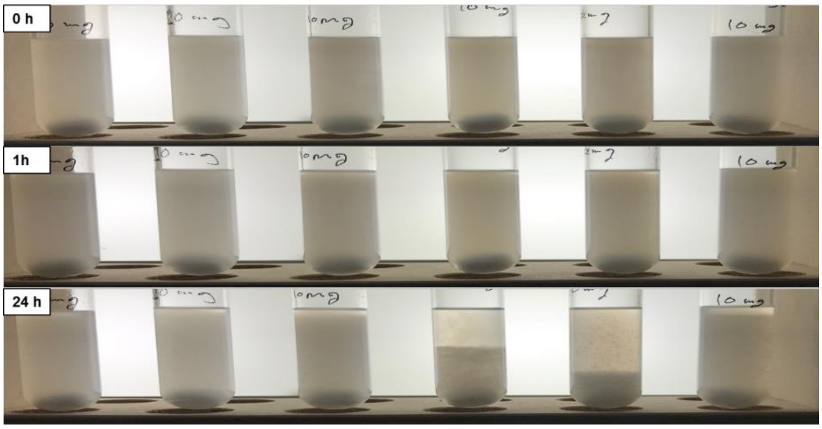

Figure 2: Dispersions from PFPE and different PTFE powders directly after ultrasonication, after 1 h and 24 hours of settling. From left to right: PTFE-9, PTFE-8, PTFE-4, PTFE6. PTFE-7, PTFE-3

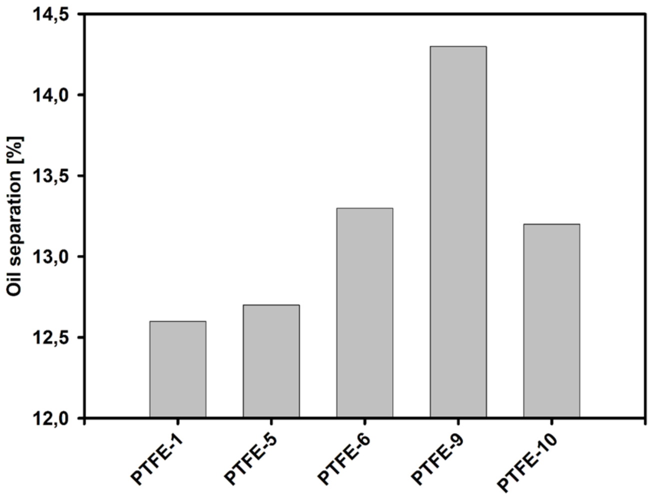

Figure 3: Results from oil separation testing (30h, 204 °C) for selected PTFE candidate powders.

In a next step the optimum PTFE content was investigated. Similar as for oil viscosity, also the viscosity of a grease improves dispersion stability. It is also known that viscosity of a dispersion increases with solid content. Equivalently to the base oil, the grease was specified to remain within certain viscosity limits. Greases with a PTFE content between 21-33% were produced. At PTFE concentrations of >30%, the greases achieved equivalent or higher viscosities than the well-known space grease Braycote 601 EF. As it can be seen in Figure 4, dispersion stability increased with viscosity, indicated by a decrease in oil separation. According to these findings, a PTFE content between 27-31% is most favorable for LTS properties.

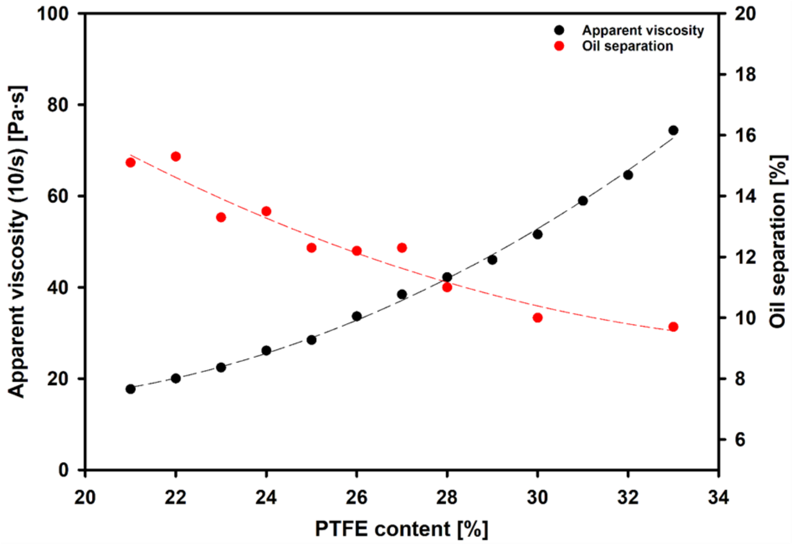

Figure 4: Dependence of viscosity (20 °C, shear rate 10s^{-1}) and oil separation (204 °C / 30h) from grease PTFE content.

Finally, the use of additives for an improvement of chemical stability was investigated. As mentioned, PFPE oils are susceptible to chemical degradation, when they are extensively sheared on ferrous surfaces. Under these conditions Lewis acids can form, which initiate an autocatalyzed degradation. This phenomenon is known for a long time, and approaches to mitigate this behavior by the use of additives have been made. In a publication by Lu [RD13] the stabilization of PFPE was assessed via thermogravimetry. According to this logic, a PFPE oil that is stabilized starts to decompose at higher temperatures or the decomposition proceeds slower. Tests were performed with various additives that were all immiscible with PFPE and added as powder. It was found that the combination of an antioxidant and a metal desactivator showed a synergistic effect and the best performance in TGA tests (see Figure 5). Based on these results it was decided to consider the use of chemical stabilizers for the development of LTS grease formulations.

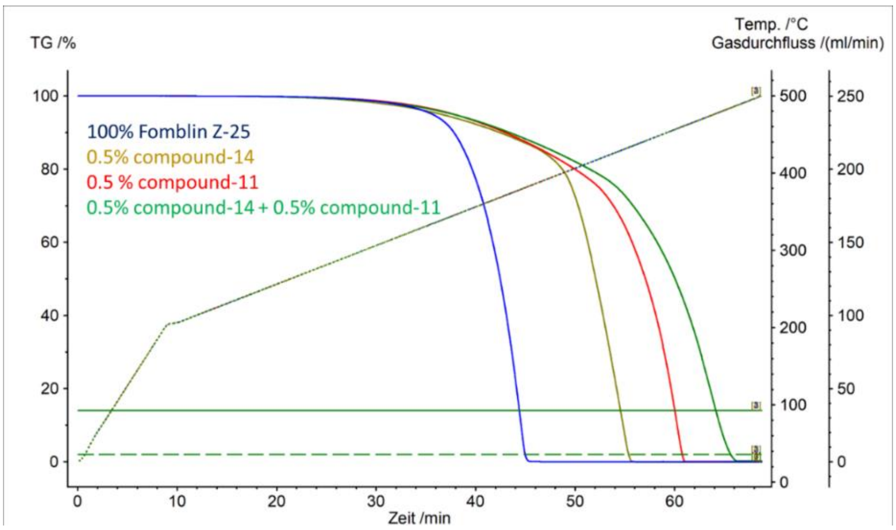

Figure 5: TGA curves for Fomblin Z25 and its mixtures with compound-11, compound-14 and their combination.

Preliminary formulations were manufactured, aged and analyzed as a following step. An accelerated aging at elevated temperatures was performed with the greases to assess their LTS behavior. According to Arrhenius law it was approximated that aging at 100 °C for 22 days would be equivalent to 15 years of storage at room temperature. Knowing that this approach is not accurate and cannot replace real-time testing, it represents a worst-case scenario for the lubricants and is therefore expected to be adequate as an indicator of LTS stability. The overall procedure consisted of a sequence of initial grease characterization, oven aging, manual rehomogenization and repeated characterization. This procedure was also applied to Braycote 601 EF and Braycote 601 Micronic.

For characterization of properties, the following methods were used:

- Oil separation testing at 100 °C for 22 days for investigation of physical grease stability
- Rheological testing for investigation of physical grease stability and viscosity
- Thermogravimetry for analysis of water uptake and chemical stability
- Vacuum tribology for assessment of grease functionality
- In selected cases outgassing measurements according to ECSS-Q-ST-70-2C

Different conclusions could be made from the analysis of preliminary formulations:

- Higher PTFE content is beneficial for LTS properties and for tribological lifetime
- Use of organomodified bentonite is beneficial for tribological lifetime but not for LTS
- Use of additives for chemical stabilization is beneficial for tribological lifetime
- The use of organomodified bentonite and additives was uncritical for outgassing limits
- Artificial aging led to an increase in tribological lifetime (testing of formulation ‘G’). This might be due to an impregnation effect of the oil on the particles
- Oil separation properties after accelerated aging were slightly worse and viscosities slightly different. This was observed for the preliminary greases, but also for the Braycote greases. It is expected that this is not due to an actual property loss but rather due to a non-ideal rehomogenization. Therefore, a more advanced method of rehomogenization should be worked out

On the basis of these results, a final formulation was selected and fully tested.

Testing of final formulation

The final formulation, in the following described as “LTS grease”, was extensively characterized to address the diverse LTS concerns and to demonstrate essential space grease properties.

- Tribological testing

Tribological testing was performed in the SOT test at ESTL Technology. The spiral orbit tribometer (SOT) is a test facility which reproduces the kinematics of an angular contact bearing, and allows for the evaluation of friction and degradation rates (i.e. consumption/wear) of lubricants in detail. The arrangement of the SOT allows the ball to experience rolling, sliding and pivoting – all motions experienced by a ball in an angular contact bearing.

Table 1: Test parameters of SOT tests and summary of results

Test ID	Peak Hertzian Contact Stress [GPa]	Environment/ pressure [mbar]	Temperature [°C]	Average Lifetime [orbits/µg]	Average Steady State CoF
1	2.25	Vacuum / <1.3x10^{-6}	22 ± 3	268.6	0.084
2	1.88	Vacuum / <1.3x10^{-6}	22 ± 3	657.8	0.119
3	1.50	Vacuum / <1.3x10^{-6}	22 ± 3	1540.5	0.119
4	2.25	Vacuum / <1.3x10^{-6}	50 ± 3	207.3	0.108
5	2.25	Vacuum / <1.3x10^{-6}	80 ± 3	156.5	0.092
6	2.25	Cleanroom air moist (55 ± 10% rh) / 101	22 ± 3	5356.9	0.113

Different parameters as the effect of contact stress, environment and temperature were investigated. Plates and ball are made from 440C stainless steel. An extension in lifetime occurred with a reduction in contact stress and a reduction in lifetime with an increase in test temperature. Also, an increase in the lifetime when testing in air was observed, which is also in line with previous testing of PFPE lubricants. Alongside the tribological data captured, mass spectrometric scans were performed and residual gas analysis (RGA) performed to determine degradation species. RGA has been used to characterise PFPE degradation for multiple activities, it is known that the partial pressure of specific species increases as a result of lubricant failure and this behavior was found also for LTS grease.

The tribological data was compared to the results of Braycote 601 EF from earlier test campaigns. It can be seen that the relationship of lifetime against peak contact stress is very similar for the two lubricants, with a similar trend observed for both data sets. Across all stresses the lifetime measured for LTS grease is greater than that of Braycote 601EF, with a similar improvement in lifetime measured for the middle and lower stress values. The LTS grease also results in a longer lifetime than the Braycote 601EF grease when tested in air, this is shown in Figure 6. This increase in lifetime is significantly more pronounced than the lifetime observed in vacuum.

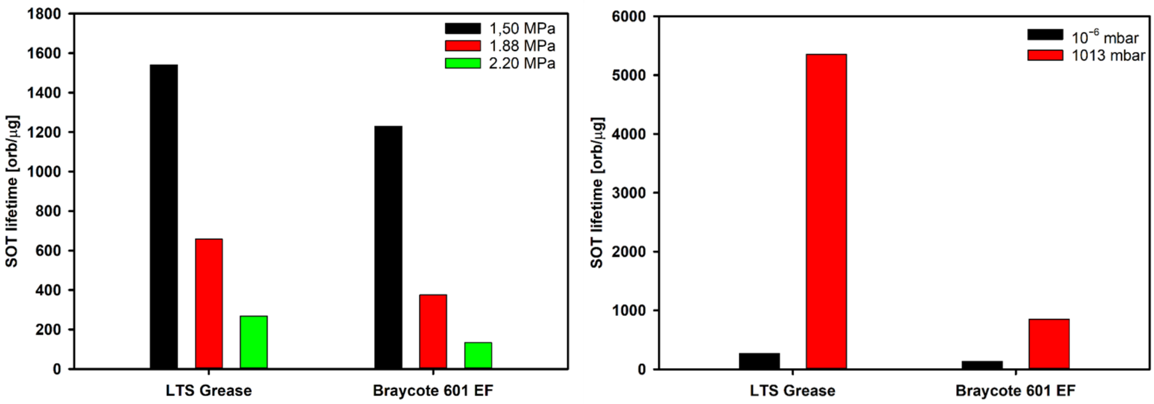

Figure 6: Left: Effect of contact stress on the lifetimes of LTS grease and Braycote 601EF [RD14]. Right: Comparison of SOT lifetimes for LTS grease and Braycote 601EF measured in different environments (Braycote 601EF data taken from [RD14 & RD15]

- Oil separation

Concern exists regarding the separation of the oil from the grease, allowing migration to other surfaces. This can cause changes in the consistency of the grease and in so doing may alter its tribological properties. In the extreme it may create lubrication-starved conditions during initial operations. Oil separation usually occurs on static components and is thus likely to occur during prolonged storage. Oil separation was investigated at room temperature with LTS grease, Ultratherm 2000 and Braycote 601 EF. These tests have been conducted for at least 24 weeks, and are still ongoing for LTS grease (now for more than 6 months).

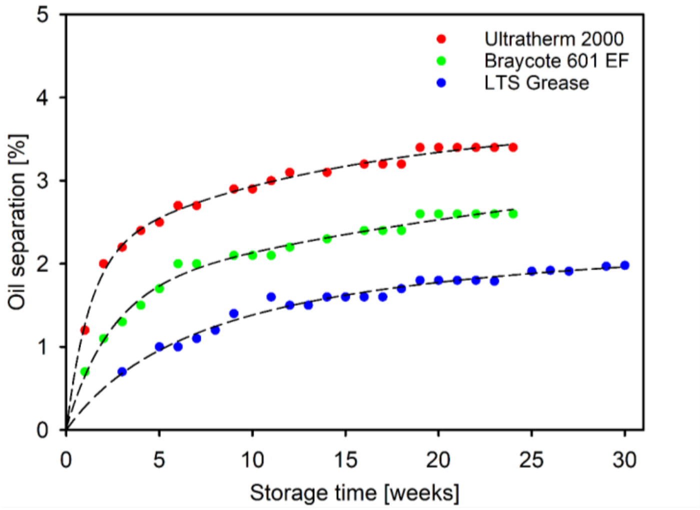

Figure 7: Results from room temperature long term oil separation testing of greases

LTS grease shows less oil separation than the commercial benchmark greases. It appears that the separation converges against a limit over time, this observation is in line with results from a real-time LTS study conducted by ESTL (funded under ESA-ESTEC Contract No 4000120585/17/NL/MP after more than two years. After 30 weeks the oil separation is at 1.95% for LTS grease.

- Phenolic cage fluid loss

The compatibility with the porous phenolic resin Krütex 100P was tested to investigate oil saturation of phenolic bearing cage materials. Such phenolic cages are typically vacuum-impregnated with an appropriate oil prior to assembly, and it is a known difficulty to achieve a full saturation of cages with oil. It is an LTS concern that these cages have a capacity to take up more free oil from the bearing during months or years in storage, reducing the free oil quantity in a lubricant and even leading to starvation (an insufficient supply of lubricant present on balls and raceways to permit proper lubrication). In order to address this concern, a multistep impregnation procedure with the base oil of LTS grease was performed. Weight change was observed for detection of a saturation limit and visual inspection was performed to detect potential incompatibilities. After 3 impregnation cycles, a saturation could be achieved. Furthermore, visual observation showed neither discolorations nor signs of swelling, which indicates that no interaction between the materials occurred.

- Lubricant creep

When for larger volumes of lubricants are applied due to e.g. high temperature operation and/or long lifetime, migration is a likely phenomenon because the mass of the fluid body is high compared to the retaining viscous forces and substrate oil-film surface energetics. Such so-called 1-g migration might impact the subsequent flight lifetime of the mechanism if the oil is able to exit the bearing envelope. During on-ground storage creep is driven by surface energy differentials and also by gravity and can be minimized partly by control of surface features and by the appropriate application of anti-creep coatings which have a lower surface energy than the lubricating fluid. Oil migration tests were performed with the creep barriers Acota EGC 2708 (equivalent to 3M Novec 2708) and Dr. Tillwich Antispread E2 Concentrate. The materials were applied on a steel plate, and one drop of base oil mixture was applied inside the formed shape of creep barrier. The steel plate was placed in an oven at 100 °C and tilted. After 24 hours of testing the oil spreaded over the substrate due to the tilting, but it was not possible to pass the creep barriers. This could confirm the compatibility and usability of the base oil with the tested creep barriers to mitigate lubricant creep risks.

- Material compatibility

Corrosion and material compatibility tests were performed. Therefore, different metals and Polyimide (Tecasint 4011) were coated with grease and stored in a climate chamber for 2 weeks at 80 °C and 60% relative humidity. Samples were analyzed for weight changes and for their visual appearance. The metal samples did not change in weight after all grease was removed. Only the Polyimide increased between 0,13-0,17% in weight. This also happened to the blind reference sample. Hence, this effect was attributed to water uptake.

	1.3505	1.4125	1.4542	3.7075 (Al)	3.1765 (Ti Gr. 5)
weight change [mg]	0,1	-0,1	0,1	0	0,3
before					
after					

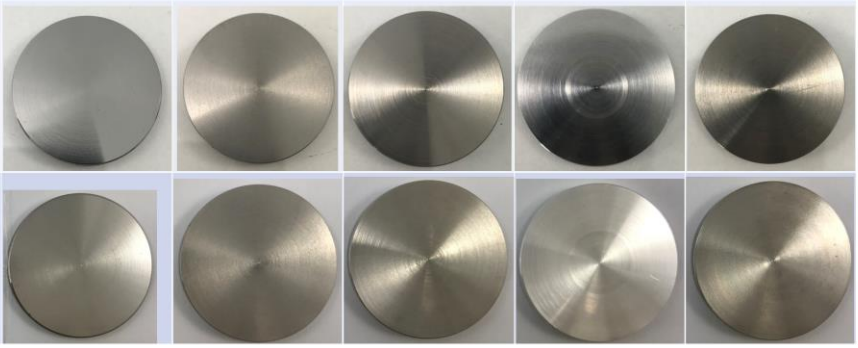

Figure 8: Pictures from corrosion testing before and after storage at 80°C and 60% relative humidity for 2 weeks.

In a separate test, the porous polyamide Nylasint was treated with the LTS grease under identical conditions. The Nylasint material showed a mass increase of 14%, and an increase of 18% in a reference experiment with Braycote 601 EF. It is expected that oil or grease migrate into the porous structure of the material. Visual inspection of the samples did not reveal indications of swelling or incompatibility.

- Other testing

Further tests have been performed with the LTS grease and also Braycote 601 EF. Results are summarized in Table 2.

Table 2: Test results for LTS grease and Braycote 601 EF

Testing	Standard/ Reference	Braycote 601EF ('*' = data from TDS)	LTS grease
Density	DIN EN ISO 12185	1.85 g/ml*	1.97 g/ml (calculated)
Thermal stability (TGA)		Mass loss 2% (N_2): 369°C Mass loss 2% (O_2): 368°C	Mass loss 2% (N_2): 333°C Mass loss 2% (O_2): 312°C
Outgassing properties	ECSS-Q-ST-70-02C	TML* <1% max ; CVCM* <0.1% max	TML: 0.16%; RML: 0.12%; CVCM: 0.03%
Evaporation	ASTM D972	2% max*	0.68%
Oil separation	ASTM D6184	204°C/30 h: 12.4% 100°C/22 d: 6.0%	204°C/30 h: 8.0 % 100°C/22 d: 7.1 %
Viscosity	DIN 51810-1	Viscosity of base oil*: 110-170 cSt (38°C)/ 40-50 cSt (99°C) Viscosity index of base oil*: 340 min Viscosity of grease: 10/s: 60.1 Pa·s ; 100/s: 8.7 Pa·s	Viscosity of base oil: 167 cSt (40°C) / 36 cSt (100°C) Viscosity index of base oil: 262 Viscosity of grease: 10/s: 53.6 Pa·s ; 100/s: 9.0 Pa·s
Dropping Point	ASTM D2265	182°C min*	200°C (ASTM D566)
Pour Point	ASTM D5985	-73°C max	-71°C (ASTM D5950)
Upper use temperature	FAG FE 9 / ECSS-Q-ST-70-02C		125°C (lifetime FAG FE9: 131 h)

Conclusions

The project goal to develop a grease formulation that is based on European resources, that fullfills essential criteria to be applied in space industry and that shows LTS properties was achieved. All relevant properties could be demonstrated, in direct comparison with the commonly used space grease Braycote 601 EF the LTS grease formulation showed equivalent or better performance. The target TRL of 4 for this development was achieved. Next steps require a further standardization and scale-up of manufacturing, and additional testing to build confidence in the LTS performance, but also in the general tribological performance in space applications.