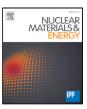


Contents lists available at ScienceDirect

Nuclear Materials and Energy



journal homepage: www.elsevier.com/locate/nme

# Selection of radiation tolerant commercial greases for high-radiation areas at CERN: Methodology and applications

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# ARTICLE INFO

Keywords: Radiation effects Radiation to materials Lubricants Oil Grease Radiation tolerance Radiation damage

## ABSTRACT

At the European Laboratory for Particle Physics (CERN), commercial greases and oils are extensively used to lubricate moving devices operating in high-radiation areas, absorbing doses up to the MGy range. Due to their sensitivity to radiation, lubricants can become fluid or solid in operation, potentially causing component failures and compromising the operation of the accelerator complex. CERN is therefore developing methodologies to select radiation tolerant commercial lubricants for critical Beam Intercepting Devices (BIDs) such as high-power targets, dumps and collimators. A careful selection of greases is fundamental to reduce the risk of failure, to optimize equipment lifetime and to minimize maintenance and unwanted radiation exposure of personnel in high radiation areas.

Three different parameters are considered to select commercial lubricants: the expected total dose absorbed in operation, the maintenance feasibility and the failure impact in terms of accelerator downtime and of personnel involvement in replacement procedures. Based on these criteria, an example of relevant application is presented: the lubrication of the new internal beam dump of the Super Proton Synchrotron (SPS). Experimental radiation damage data allowed the identification of two radiation tolerant commercial greases to lubricate the dump support jack assembly.

#### 1. Introduction

Commercial non-metallic materials are exposed to high levels of radiation during operation in accelerators and high power facilities. Despite their sensitivity to radiation [1], polymeric materials are increasingly used in new-generation facilities. At the European Laboratory for Particle Physics (CERN), various types of both commercial and custom-made components are used in high radiation areas as part of Beam Intercepting Devices (BIDs) or in their proximity. Polymeric components including lubricants, elastomeric O-rings, insulators, resins and glues are often needed as part of these devices.

Lubricants are of particular concern because of their extensive use at CERN. In fact, most mechanism components used in accelerator environments and in particular as part of BIDs, such as bearings, gears, screw jacks and ball screws, require lubrication.

At the radiation dose levels expected in operation, lubricants can experience extreme degradation becoming completely fluid or thickening/ solidifying, and also evolving gas and acid products [1,2]. This can lead to equipment failures and possibly compromise accelerator operation. Few studies have been performed specifically on the radiation resistance of lubricants in recent times. In the last decades irradiation tests of lubricants have been driven by the development of space technology, accelerator facilities, fusion research and nuclear power plants [3–8]. At CERN, extensive irradiation testing of materials was performed from the late 1960's until the early 2000's. The obtained radiation damage test data were compiled in a series of reports part of the Yellow Reports [9] monographs. They are still used as one of the main references for material selection for high-radiation areas. Results were collected using mostly gamma radiation, assuming that comparable effects would be induced by equivalent doses of other radiation fields [9]. Recently, this general assumption has been questioned and the need for further research on this topic is considered necessary for the development of high-radiation facilities [10,11].

More recently, data has been collected on commercial lubricants for use in spallation neutron sources and facilities for the production of radioactive ion beams [12,13]. Grease samples were irradiated in a nuclear reactor mixed neutron and gamma field. The samples exhibited a very wide range of sensitivity to radiation at doses comparable to

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https://doi.org/10.1016/j.nme.2021.101088

Received 2 March 2021; Received in revised form 6 October 2021; Accepted 3 November 2021 Available online 27 November 2021

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the ones absorbed by the most critical equipment at CERN during their lifetime. BIDs such as high-power targets, dumps and collimators are among the devices exposed to highest levels of radiation at CERN [14–17]. In fact, BIDs directly intercept high energy proton beams, producing intense secondary radiation that can damage materials. Because of increasingly demanding particle beam parameters, the risk of premature failure of commercial components used in BIDs is becoming more and more serious and the need for demonstrated radiation tolerant materials is growing.

The collected results highlight the need to carefully select the most tolerant products available on the market and to test them to increase the know-how on this topic. CERN is therefore carrying out irradiation tests to experimentally assess radiation degradation of specific commercial products when exposed to different levels and types of radiation.

The aim of this work is to present a methodology developed to identify the most suitable commercial lubricants for high-radiation areas. Application to a relevant case is discussed: the selection of commercial lubricants for the Super Proton Synchrotron Internal Beam Dump.

Research activities conducted at CERN to investigate radiation effects in lubricants are presented as well. They are currently mostly focused on polyphenyl ether (PPE)-based lubricants, identified as promising highly radiation-resistant candidates available on the market. Investigations on lubricants are ongoing in collaboration with LUBRILOG [18] (France), a company specialized in the development and manufacture of special lubricants since 1987 and with MORESCO [19, 20] (Japan), an R&D-focused Corporation, manufacturing special lubricants for extreme applications since 1958.

#### 2. Radiation effects in lubricants: a selection of results

Greases originate from the dispersion of a thickener in a fluid oil [1]. They are complex multi-phase and non-newtonian systems, whose properties arise from the interaction between the oil, the thickener and other possible additives [21]. Radiation affects all grease ingredients on a scale that depends on the material and irradiation conditions. Most predominantly, oil chain length and thickener structure are influenced, often in competing ways, resulting in overall grease softening or hardening [1,22,23]. Additional radiation effects include gas production, evolution of acid products, and colour change [3].

Nine commercial greases having different compositions were irradiated in an irradiation facility of a nuclear reactor in a previous study [12,13]. In this irradiation facility, mixed neutron and gamma fields are present [24]. Grease samples were simultaneously exposed to an average neutron flux of  $1.72 \ 10^{13} \ cm^{-2} \ s^{-1}$  and an average photon flux of  $1.65 \ 10^{13} \ cm^{-2} \ s^{-1}$ . Duration of the irradiations ranged between 10 min and 15 h. Dose rates ranged from about to 0.75 MGy/h to 0.95 MGy/h, and the neutron component of the total dose represented 65%–70% of the total dose, the remaining being gamma dose. Slight differences in dose rate and in neutron to gamma dose ratio are due to different compositions of the various greases [12,13]. Temperature during irradiation ranged between 50 °C and 70 °C. Irradiations were realized in air, in a ventilated environment and at atmospheric pressure.

Total doses ranging from about 0.1 MGy to more than 10 MGy were delivered to the samples. Three different levels of radiation damage (stable, moderate, severe) were defined in the study to assess the deterioration of the grease at the macroscopic level. Stable and moderate degradation correspond to a variation of worked penetration [25], the selected property, within 10% or higher than 10%, respectively. Severe degradation indicates a structural failure of the grease, becoming fluid or thickening/solidifying.

Fig. 1 shows the radiation-induced degradation of the irradiated greases. Details on the selected commercial products and on this irradiation campaign are reported in [12]. The reported sensitivity range

for the selected greases is very wide, covering about two orders of magnitude. Severe degradation occurs at about 0.3 MGy for the most sensitive greases, while the most resistant one remains stable up to about 12 MGy. Accordingly, the selection of a radiation sensitive lubricant could reduce the lifetime of the lubricated components in high radiation areas by a factor of 100.

For most of the tested greases a progressive softening with dose was observed in the tested dose range. Severe degradation was indicated by a fluidization of the material. By contrast, for one of the greases softening was not observed. The material became hard at high doses. Referring to the thresholds reported in Fig. 1, Klüberlub PETAMO GHY 133 N and MORESCO RG-42R-1 are the two most radiation resistant products among the tested ones. The evolution of their worked penetration, as well as other parameters assessed qualitatively such as colour and texture variation, is briefly discussed.

For Klüberlub PETAMO GHY 133 N, severe degradation is associated with material hardening, as shown in Fig. 2. An evident darkening of the grease with dose can be observed as well. The grease became very hard and sticky at the maximum dose level of 13.3 MGy. Consistency variations remained lower than 10% up to about 9.0 MGy, so the grease is considered stable up to this dose level, as summarized by Fig. 1.

The most radiation-resistant grease irradiated in the study, MORESCO RG-42R-1, remained stable up to almost 12 MGy. Its evolution with dose is shown in Fig. 3. A slight darkening of the grease can be observed. A very mild softening of the material is evident at the maximum tested dose value of 11.4 MGy. However, consistency variations remained lower than 10% up to the maximum dose level, so the grease is considered very stable and radiation tolerant in the whole tested dose range.

RG-42R-1 is a commercial PPE-based product realized with commercial oil RP-42R and an inorganic thickener. The product is patented and it was specifically developed to meet radiation resistance requirements [26]. The superior resistance of PPE products is already demonstrated in literature [1,22]. To further investigate the radiation effects in RG-42R-1 grease, an irradiation study was specifically dedicated to its main component, RP-42R oil [20]. The study evidenced the increase of oil viscosity with dose due to radiation-induced polymerization and a remarkable chemical stability of the oil up to 7.8 MGy. The radiation resistance of RG-42R-1 seems to result from the mutual interaction between a radiation tolerant oil with a sufficiently stable gelling agent. Unlike many other greases which liquefied at dose values ranging between 0.3 MGy and about 2.5 MGy, the thickener of RG-42R-1 remained overall effective at all the tested dose levels [1]. For these reasons, PPE-based products including RG-42R-1 have been selected for further radiation damage studies at CERN, currently ongoing and briefly discussed in Section 5.1.

The results also show that opposite effects can dominate in different greases irradiated in the same irradiation conditions. In addition to the chemistry of the ingredients, the severity and type of radiation damage mechanisms depend on a number of parameters such as total dose, type and energy of radiation, dose rate, oxygen diffusion, temperature, humidity and mechanical stresses, acting in synergy. The role of these parameters in determining the dominant radiation effect mechanism is in many cases still unknown. Gamma irradiation has been so far the most commonly used method to perform such tests, under the assumption that equal effects would be induced by an equal dose delivered by a different radiation field [9]. At CERN, new irradiation test areas where mixed radiation fields will be used for materials irradiation are being designed and built. This will allow this assumption to be verified.

## 3. Selection of lubricants at CERN

#### 3.1. Lubricants used at CERN: challenges and approaches

Commercial lubricants are extensively used at CERN, in both high and low radiation areas. Many moving mechanisms such as rolling

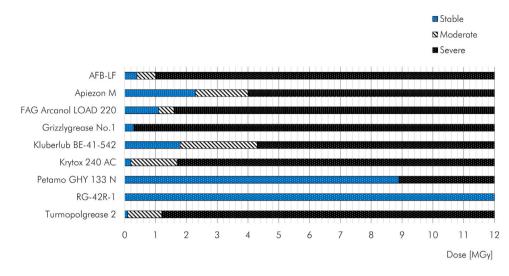


Fig. 1. Damage thresholds for nine commercial greases irradiated in mixed neutron and gamma radiation. Data extracted from [12,13].

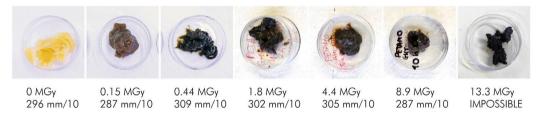


Fig. 2. Klüberlub PETAMO GHY 133 N grease samples; texture and colour variations can be observed as a function of the total dose (in MGy). Worked penetration [25] values are reported as well (in mm/10). At the maximum tested dose value of 13.3 MGy the grease became so hard that penetration test was impossible [12,13,35].

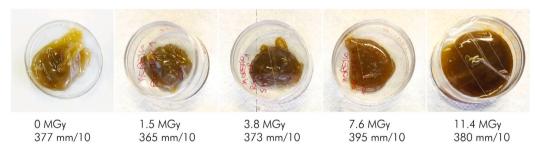


Fig. 3. MORESCO RG-42R-1 grease samples: texture and colour variations can be observed as a function of the total dose (in MGy). Worked penetration [25] values are reported as well (in mm/10) [12,13,35].

bearings, ballscrews, rollerscrews, screw jacks and linear guides equipping different devices need lubrication. In many cases, lubricated components are used in radiation environments characterized by the presence of high energy hadrons. Due to the high levels of residual radioactivity after operation, access to the areas where the lubricants are used is subjected to restrictions, following Radiation Protection regulations. Maintenance is sometimes limited and radioactive waste considerations need to be taken into account.

The approach proposed in this paper combines both theoretical considerations, driving the selection of promising candidate products, and the realization of dedicated experimental tests. The selection of the most suitable commercial lubricant for a specific application needs to be optimized considering the huge price difference between a generic and a special radiation tolerant product which in some cases can reach a factor of 100 or higher. In view of their synthetic formulation, PPE-based products are particularly expensive, and for this reason it is not possible to use them for all CERN applications. An optimized selection of ordinary products is needed as well, to cover many of CERN's requirements. The use of the most expensive radiation tolerant products should be restricted to applications in high radiation areas.

Some key parameters to correctly select the grease for a specific application are analysed: the total absorbed dose, the feasibility of maintenance procedures and the impact of lubrication failure. Based on the analysis of these parameters, different approaches to the selection of a suitable lubricant are proposed. Apart from these parameters, it is always necessary to take into account the specific role of the lubricant in the analysed equipment.

## 3.2. Total absorbed dose

The total dose absorbed by a lubricated component during its lifetime is traditionally used as the main predictor of radiation damage. The values shown in Table 1 are tentatively used as thresholds for lubricant degradation, based on the information currently available in literature (e.g. the CERN's Yellow Reports [9]) and on previous experience at CERN.

The proposed classification is intended as a rough reference only. The mentioned thresholds might vary depending on irradiation conditions and specific applications. The validity of total dose as the main reference is currently being questioned and will be verified by parallel Table 1

Lubricant damage based on total absorbed dose: a classification.

Dose MGy	Expected damage	Recommendations
<0.1	Negligible	Ordinary products
0.1-1.0	Moderate/severe	Radiation resistant tested products
1–10	Moderate/severe	Special products with demonstrated resistance in this range
>10	Severe, failure	Avoid polymeric lubricants if possible

radiation damage studies performed in different radiation fields (see Section 6).

#### 3.3. Maintenance feasibility

Re-lubrication and replacement of a lubricated element or of a whole assembly represent possible strategies to extend the lifetime of lubricated equipment in high radiation areas. Unfortunately, this is rarely feasible or easy at CERN, mainly as a result of access limitations to these areas due to residual activation and the need to keep radiation exposure of personnel as low as reasonably possible, in agreement with the ALARA principle.

Even when technically feasible, maintenance is restricted to specific time slots during the year, for example corresponding to planned accelerator shutdowns and is further limited by Radiation Protection regulations in case of short cool down times.

#### 3.4. Failure impact

Lubrication failure can have a dramatic impact on machine operation, potentially leading to equipment damage and unforeseen shutdowns of the whole accelerator complex. This is for example the case of major failures of dumps or collimators. The higher the impact of a failure, the more reliable the lubrication needs to be under irradiation.

In the following Section, the methodology proposed above is applied to a recent application at CERN.

#### 4. An application example

#### 4.1. SPS beam dump and its support jack assemblies

At CERN, particles are accelerated to high energies to be used in high energy physics research experiments. Proton beam energies reach 450 GeV/c in the Super Proton Synchrotron (SPS) [15]. As part of the accelerators' routine operations, the accelerated beams are regularly stopped and absorbed by beam dumps when not required anymore.

The new generation internal beam dump of the SPS, referred to as TIDVG5 (Target Internal Dump Vertical Graphite), was installed during the CERN's Long Shutdown 2 (LS2), 2019–2021 (see Fig. 4). The core of the dump is a 5 m long array of absorbing blocks of four materials, consisting of 4.4 m of isostatic graphite, 0.2 m of TZM, 0.4 m of pure tungsten, enclosed within 2.5 m long CuCrlZr [14,15]. High average beam power of approximately 270 kW/h is deposited in the SPS beam dump by the primary proton beam.

Due to the increased beam intensities and energies resulting from recent upgrades, the total energy to be dissipated by TIDVG5 is more than four times the one of the previous dump model [14]. Accordingly, the requirements on the radiation tolerance of the dump components, including its lubrication, became more critical than before, and required further analysis.

For radiation safety reasons related to the expected high residual activation after nominal operation, the dump is housed inside a massive shielding structure with around 2 meters thick multi-layered walls and roof [15].

The TIDVG5 is expected to survive for a minimum of 20 years of operation, corresponding to approximately  $3.4 \ 10^{19}$  protons over

the target lifetime [27]. This expected lifetime is compatible with the general activities and schedule of the accelerator complex and is comparable with typical lifetimes of similar components at CERN [17].

As described in Sections 4.4 and 4.5, maintenance and replacements on BIDs are extremely complex and highly impacting both the accelerator schedule and the personnel safety. The functionality of TIDVG5 will be monitored over time and its lifetime might be further extended in case of satisfactory performance. Prolongation of the lifetime of BIDs and other equipment operating in high-radiation areas is of utmost importance at CERN. As described in the following Sections, BIDs lifetime depends as well on to the used polymeric components and could benefit from a careful selection of commercial products with a demonstrated radiation resistance.

## 4.2. Lubricated components of the TIDVG5

The TIDVG5 is expected to need a yearly re-alignment with the beam line to ensure optimum performance. It is therefore mounted on a set of three support jack assemblies that permit alignment in six degrees of freedom, as shown in Fig. 5.

Three assemblies support the TIDVG5; they vary only in their direction and length of several elements in order to reflect their respective load. The support assemblies are composed of a combination of custommade and adapted commercial components. For example, lifting jacks with a custom gear ratio are commercial products enabling low torque operation with a high positioning precision. These lifting jacks, illustrated in red in Fig. 6, ensure the vertical motion of the dump by transforming gear rotation into screw translation (3, Fig. 6). The lubrication of the contact point can be replenished from the grease reservoir at the bottom of the jack (4, Fig. 6). The horizontal motion of each assembly can be controlled in one direction thanks to a lead screw (5, Fig. 6) combined with a couple of parallel linear roller bearings, one flat and one V-shaped (6 and 7 in Fig. 6). The jack assembly in the other horizontal direction is free to move thanks to V-shaped roller bearings (1, Fig. 6). Finally, potential angles are compensated by a round-shaped interface (2, Fig. 6) between the top of the lifting jacks and the bottom of the upper V roller bearings.

All of these moving components need lubrication to ensure an easy and smooth motion compatible with man-powered operation and a limited intervention time.

## 4.3. Total absorbed dose

At CERN, radiation doses are calculated using Monte Carlo simulations performed in FLUKA [28,29], a code for transport, interaction and energy deposition of energetic particles in materials. The reliability of FLUKA simulations has been demonstrated by satisfactory agreement with subsequent measurements. As an example, beam losses in the Large Hadron Collider (LHC) were studied in [30].

FLUKA simulations of the TIDVG5 area, mentioned in [15], are illustrated in Fig. 7. The following beam properties have been considered: beam momentum of 450 GeV/c, intensity per pulse of 288 bunches times 2.43  $10^{11}$  protons/bunch, assuming an annual number of Protons On Target (POT) of 1.7  $10^{18}$  dumped protons [27].

Simulations show that lubricated elements within jack assemblies are expected to absorb doses ranging between 0.5 MGy and 10 MGy in 20 years of operation. With respective doses of 10 MGy and 4 MGy, the upper V roller bearings and spherical bearings (components 1 and 2 in Fig. 6) fall into the most critical category of lubricant application defined in Section 3.1.

The remaining lubricated components at the base of the jacks would be exposed to doses between 0.5 MGy and 1 MGy; lubricants with a demonstrated radiation tolerance are therefore required.

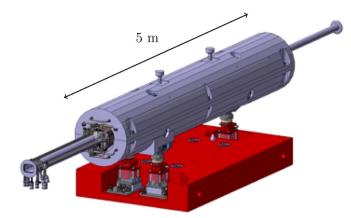


Fig. 4. Isometric view of the TIDVG5 on its three support jack assemblies installed in a cast iron base.

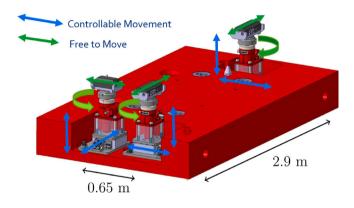


Fig. 5. Directions of motion of support jack assemblies allowing dump positioning with 6 degrees of freedom.

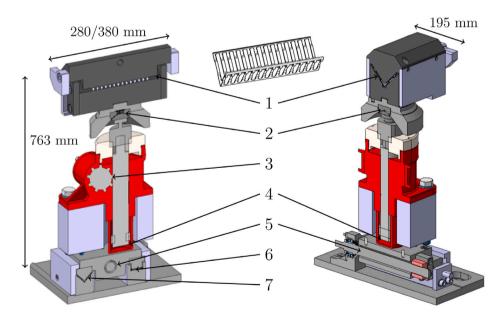
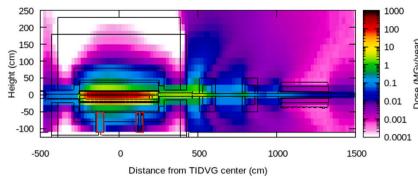


Fig. 6. Components of TIDVG5 support jack assemblies and location of lubricants: upper V roller bearing (1), plain spherical bearing (2), interaction area between gear and threaded screw (3), grease reservoir (4), lead screw (5), lower flat roller bearing (6), lower V roller bearing (7). The internal components of the lifting jack are represented only schematically, as its design is proprietary. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

## 4.4. Maintenance feasibility

FLUKA simulations are used to determine residual dose rates in the TIDVG5; these are of the order of hundreds of mSv/h at the location

of the support jack assemblies after one week of cooling time. Given these high foreseen residual dose rates, frequency and duration of interventions should be minimized. Re-lubrication is not possible, due to difficult access and contamination issues. Support replacement is



Annual Dose from standard dumping of HL-LHC high energy beams (450 GeV/c)

**Fig. 7.** Annual dose in the SPS internal beam dump area, assumed annual POT of 1.7 10<sup>18</sup> protons [27]. The position of support jacks assemblies is highlighted in red. Per year, the upper V roller bearings are expected to absorb up to 0.5 MGy, the spherical bearings at the top of the lifting jacks should be exposed to 0.2 MGy, and the remaining lubricated components to about 0.025–0.05 MGy.

Source: Courtesy of J.A. Briz, A. Lechner and V. Vlachoudis.

possible, but necessarily remotely handled, complex, time consuming and expensive. For this reason, lubrication should ideally last for the whole TIDVG5 lifetime.

#### 4.5. Failure impact

A lubrication failure preventing the TIDVG5 from being properly realigned would necessitate the following time-consuming and difficult operations: removal of the shielding roof and removal of the 25 tonne dump using a remotely operated crane, disconnection and replacement of the upper jack bearings from the dump using robots. This would be followed by replacement of the dump and shielding then a full realignment campaign. These operations would need several weeks to complete. The TIDVG5 support jack assemblies therefore represent an example of a challenging application for lubricants according to the definition proposed in Section 3.

#### 4.6. Selection of commercial lubricants

More than 8 kg of grease are needed to lubricate the three support jack assemblies (Fig. 6). Given the required amount, a careful choice of commercial greases is needed, to optimize performance and price.

As shown in Fig. 1, two commercial greases retain a stable consistency after irradiation in a reactor mixed field up to a dose of about 9 MGy: Klüber PETAMO GHY 133 N and MORESCO RG-42R-1 [12,13]. Fig. 2 and Fig. 3 show samples of the two greases after absorbing the maximum investigated dose. At approximately 10 MGy, PETAMO GHY 133 N became hard and glue-like. On the other hand, RG-42R-1 showed a stable consistency up to about 12 MGy, although the grease starts to show a moderate softening over 10 MGy. Additional analyses performed on the base oil of RG-42R-1 confirmed its overall chemical stability in this dose range [13,20]. Moreover, a slight softening is still compatible with operation unlike hardening.

Based on these considerations, RG-42R-1 was selected to lubricate the upper V roller bearings (component 1 in Fig. 6), where the expected dose during the component lifetime is the highest. The lubricated upper V roller bearings are shown in Fig. 8. The dose expected for all the other components seems compatible with radiation tolerance of PETAMO GHY 133 N as well, so both the products can be used to lubricate the other components. Other factors such as relative price and the amount of required lubricant motivated the final selection of PETAMO GHY 133 N for the components 2, 3 and 4, and of RG-42R-1 for components 5, 6 and 7. The proposed choice of lubricants aims to ensure lubrication over the lifetime of the dumps while optimizing the total costs and satisfying other technical requirements.

Given the expected dependence of radiation effects in greases on irradiation conditions, lubricant useful lifetime in operation will likely differ from the values reported in Fig. 1. The proposed selection of lubricants relies on the assumption that the dose thresholds in operation would remain of the same order of magnitude than the values obtained in testing conditions. Future studies mentioned in Section 6 aim at verifying the validity of this assumption.

#### 5. Experimental gamma irradiation studies

Extensive material studies have been performed at CERN in the last decades on commercial non-metallic materials. Results on lubricants are reported in the Yellow Reports [9]. Considering the evolution of the market, this knowledge needs to be updated by the addition of new data.

Experimental studies are underway to improve the understanding of radiation effects in lubricants, allowing a better selection of products to be used at CERN in high radiation areas.

#### 5.1. Selected lubricants for upcoming studies

Polyphenyls are widely regarded as the most radiation-resistant organic fluids [1,22] and they are successfully used as neutronmoderating coolants in nuclear reactors [31,32]. However, their poor viscosity-temperature properties make them less suitable for use as lubricants than the corresponding polyphenyl ethers [33]. Previous studies on PPE-based products highlight their superior resistance to radiation-induced oxidation [1,33,34], and report an overall radiation tolerance of the order of 10 MGy [1,3,12,20,26,35]. The upcoming irradiation studies are therefore focused on commercial PPE-based products.

PPE oils are compounded with different thickeners by manufacturers to formulate greases with different properties. The amount of thickener used can be varied to obtain final products with the desired consistency.

Scientific research collaborations with international lubricant producers and grease manufacturers have been established, to characterize the irradiated products and to deepen the understanding of radiation-induced effects.

### 5.2. Irradiation plan

During a first irradiation campaign, samples of all the selected materials will be irradiated at doses ranging between 1 MGy and 10 MGy, to assess the general evolution of their main properties with dose. Gamma irradiation will be performed in commercial facilities using  $^{60}Co$  sources, at a dose rate in the order of magnitude of 10 kGy/h.

Mixed field irradiations at comparable doses are underway, using CERN irradiation facilities that have been recently designed and constructed. These activities are briefly mentioned in Section 6.



Fig. 8. Top view of the upper V roller bearing lubricated with MORESCO RG-42R-1. About 200 g of grease were employed in order to guarantee roller lubrication even in the catastrophic case of grease liquefying under irradiation.

## 5.3. Post-irradiation characterization

Radiation damage in materials is studied using a multi-scale approach, comparing material evolution at the mechanical/rheological scale and at the chemical/structural scale. These analyses will be integrated with functional tests aiming to verify the lubricant performance in operation.

Sample characterization includes but is not limited to the following analyses, to be completed in external and in-house laboratories and in collaboration with lubricant manufacturers: rheological analysis; viscosity measurement (for oils); cone penetration test (for greases); drop point test; flash point test; thermal ageing; Total Acid Number (TAN); Nuclear Magnetic Resonance (NMR); Fourier-Transformed Infrared Spectroscopy (FT-IR); Gas chromatography (GC); Gel Permeation Chromatography (GPC); four-ball test.

Each test will be carried out on all samples of oil and grease. The results collected on the irradiated samples will be compared with the unirradiated samples, to assess the evolution of the main properties with dose.

## 6. Plans for future mixed field irradiation studies at CERN

The available data on radiation resistance of greases refer to specific irradiation conditions, which differ from the ones present in operation. Radiation damage to polymers depends on the specific irradiation conditions and this dependence is largely unknown for greases. For this reason, usability thresholds in operation might differ from the ones reported in Fig. 1. This is for example the case of the TIDVG5 described in Section 4.

Intense mixed radiation fields are produced at CERN by proton interaction with targets and with beam intercepting devices [16,36]. During CERN's Long Shutdown 2 (2019–2021), test areas have been set up close to the n\_TOF neutron spallation target and downstream the ISOLDE targets. These radiation fields can be used for material irradiation in parallel with accelerator operation. This will allow radiation effects to be investigated in unique and previously unexplored conditions, better representing the operating conditions in hadron accelerators than gamma radiation. Pilot irradiations are currently ongoing.

Further investigations on the selected products will help to better assess the dependence of radiation damage on different parameters.

## 7. Conclusions

Experimental radiation damage studies are needed at CERN to improve the understanding of radiation-induced degradation mechanisms occurring in lubricating oils and greases. In the present work, methodologies are proposed to guide the challenging selection of lubricants for use in critical mechanical devices in high-radiation areas. As an example, the choice of commercial greases for the internal beam dump of the SPS accelerator is discussed.

Commercial lubricants selected on the basis of their promising chemical composition are being irradiated and will be tested in collaboration with the producers, to experimentally assess their radiation resistance in different conditions. In addition, new in-house mixed-field irradiation test areas have been installed at CERN to investigate radiation effects after exposure to mixed radiation fields, to be compared with gamma irradiations.

The use of commercial lubricants with a demonstrated radiation tolerance is nowadays a prerequisite for the upgrade of existing facilities and for the design of new ones. It is therefore important to continually update and increase the knowledge on radiation damage effects by means of radiation damage studies.

#### CRediT authorship contribution statement

**Matteo Ferrari:** Conceptualization, Methodology, Investigation, Writing – original draft, Visualization, Project administration, Supervision. **Dominika Senajova:** Conceptualization, Methodology, Writing – original draft, Visualization. **Keith Kershaw:** Conceptualization, Writing – review & editing, Supervision. **Antonio Perillo Marcone:** Writing – review & editing, Supervision. **Marco Calviani:** Writing – review & editing, Funding acquisition, Supervision, Resources.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Acknowledgements

The authors acknowledge the Radiation to Electronics (R2E) Project for the support to the activities, the FLUKA development and application team, in particular J.A. Briz, A. Lechner and V. Vlachoudis, the CERN's LHC Injectors Upgrade (LIU) Project and the CERN's High Luminosity LHC (HL-LHC) Project.

#### References

- R.O. Bolt, J.C. Carrol, Radiation Effects on Organic Materials, Academic Press, 1963.
- [2] I.V. Shul'zhenko, R.I. Kobzova, M.B. Kepurova, Y. Izotov, Effects of low-power ionizing irradiation on properties of lubricating greases, Chem. Technol. Fuel Oils 17 (4) (1981) 217–219.
- [3] K. Arakawa, et al., Data on Radiation Resistance of Lubricating Oil JAERI-M 87-141, Japan Atomic Energy Research Institute, September 1987.

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- [4] M. Saito, et al., Development of radiation hard components for ITER blanket remote handling system, Fusion Engineering and Design 109-111 (2016) 1502–1506.
- [5] K. Obara, et al., High Gamma-Rays Irradiation Tests of Critical Components for ITER (International Thermonuclear Experimental Reactor) In-vessel Remote Handling System, JAERI-Tech 99-003, Japan Atomic Energy Research Institute, February 1999.
- [6] S. Srivastava, V. Thomas, Lubricants in Nuclear Technology: A Brief Review, Government of India, Atomic Energy Commission, Bhabha Atomic research Centre, Bombay, India, 1974.
- [7] M. Bruce, M. Davis, Radiation Effects on Organic Materials in Nuclear Plants, Research Project 1707-3, Final Report, Georgia Institute of Technology, Nuclear Engineering Department EPRI NP-2129, 1981.
- [8] W. Rice, W. Cox, The Effects of Nuclear Radiation in Solid Film Lubricants, Technical Report 58-499 ASTIA Document No. 207795, Materials Laboratories of the Wright Air Development Center, Air Research and Development Command, United State Air Force, Ohio, WADC, Jan 1959.
- [9] P. Beynel, P. Maier, H. Schoenbacher, Compilation of Radiation Damage Test Data: Materials Used Around High-Energy Accelerators, Yellow Report CERN-82-10 Part 3, European Organisation for Nuclear Research (CERN), 1982.
- [10] B. Briskman, Specificity of proton irradiation effects on polymers, Nucl. Instrum. Methods Phys. Res. B 265 (2007) 72–75.
- [11] A. Rivaton, J. Arnold, Structural modifications of polymers under the impact of fast neutrons, Polym. Degrad. Stab. 93 (2008) 1864–1868.
- [12] M. Ferrari, et al., Experimental study of consistency degradation of different greases in mixed neutron and gamma radiation, Heliyon 5 (9) (2019) e02489.
- [13] M. Ferrari, Experimental Study of Radiation Resistance in Intense Neutron Fields of Critical Materials and Components for the Construction of the ESS (European Spallation Source) Target System, (Ph.D. thesis), Università degli Studi di Brescia, 2020, Cycle XXXII.
- [14] S. Pianese, et al., Hot isostatic pressing assisted diffusion bonding for application to the super proton synchrotron internal beam dump at CERN, Phys. Rev. Accel. Beams 24 (2021) 043001.
- [15] S. Pianese, et al., Design of the Future High Energy Beam Dump for the CERN SPS, Proceedings of the 9th International Particle Accelerator Conference IPAC 2018, Vancouver, BC, Canada, 2018, http://dx.doi.org/10.18429/JACoW-IPAC2018-WEPMG004.
- [16] R. Esposito, et al., Design of the third-generation lead-based neutron spallation target for the neutron time-of-flight facility at CERN, Phys. Rev. Accel. Beams 24 (2021) 093001.
- [17] E. Lopez Sola, et al., Design of a high power production target for the beam dump facility at CERN, Phys. Rev. Accel. Beams 22 (2019) 113001.

- [18] LUBRILOG and website, https://www.lubrilog.com/ (Accessed 1 December 2020).
- [19] MORESCO and website, https://www.moresco.co.jp/en/ (Accessed 1 December 2020).
- [20] M. Ferrari, et al., Characterization of a polyphenyl ether oil irradiated at high doses in a TRIGA mark II nuclear reactor, Nucl. Instrum. Methods Phys. Res. B 497 (2021) 1–9.
- [21] R. Mortier, et al., Chemistry and Technology of Lubricants, Vol. 14, third ed., Springer, 2010, pp. 411–432.
- [22] J.G. Wills, Nuclear Power Plant Technology, Wiley, 1967.
- [23] J. Davenas, Stability of polymers under ionising radiation: the many faces of radiation interactions with polymers, Nucl. Instr. Mthods Phys. Res. B 191 (2002) 653–661.
- [24] M. Di Luzio, et al., Vertical variations of flux parameters in irradiation channels at the triga mark ii reactor of pavia, Prog. Nucl. Energy 113 (2019) 247–254.
- [25] ASTM d217-21, standard test methods for cone penetration of lubricating grease, 2021, ASTM International, West Conshohocken, PA, 2021, www.astm.org.
- [35] M. Ferrari, et al., Neutron radiation effects on lubricants and O-Rings for target and accelerator applications, in: Spallation Materials Technology, Materials Science Forum Vol. 1024, (ISSN: 1662-9752) 2021, pp. 127–133.
- [26] K. Arakawa, et al., Super highly radiation-resistant grease, United States Patent, Patent Number: 4, 753, 741, Jun. 28, 1988.
- [27] E. Carlier, et al., Sps beam parameters and total dumped intensity for validation of new LSS5 SPS beam dump design, 2018, CERN, EDMS 1760169, SPS-TIDV-ES-0009.
- [28] T. Böhlen, F. Cerutti, M. Chin, A. Fassò, A. Ferrari, P. Ortega, A. Mairani, P. Sala, G. Smirnov, V. Vlachoudis, The FLUKA code: Developments and challenges for high energy and medical applications, Nucl. Data Sheets 120 (2014) 211–214.
- [29] A. Ferrari, P. Sala, A. Fassò, J. Ranft, FLUKA: A multi-particle transport code, 2005, CERN-2005-10, INFN/TC\_05/11, SLAC-R-773.
- [30] A. Lechner, et al., Validation of energy deposition simulations for proton and heavy ion losses in the CERN large hadron collider, Phys. Rev. Accel. Beams 22 (2019) 071003.
- [31] J. Smee, et al., Organic Coolant Summary Report, Canada, AECL-4922, 1975.
- [32] P. Gierszewski, R. Hollies, Organic coolants and their applications to fusion reactors, Nucl. Eng. Des./Fusion 4 (1987) 223–236.
- [33] C. Mahoney, et al., Polyphenyl ethers as high-temperature radiation-resistant lubricants, J. Chem. Eng. Data 5 (2) (1960) 172–180.
- [34] R. Bolt, J. Carroll, Radiolysis and radiolytic oxidation of lubricants, Ind. Eng. Chem. 50 (2) (1958) 221–228.
- [36] R. Catherall, et al., The ISOLDE facility, J. Phys. Nucl. Part. Phys. 44 (2017) 094002.