

Assessment of the radiation damage to the coolant in SciFi tracker

Abstract: The “back-of-an-envelope” estimates show that the effect of the absorbed radiation on the proposed SiPM coolant (C6K) in SciFi tracker is negligible. As a consequence, a study of chemical properties of C6K, primarily of its reactivity with water, has a higher priority compared to the radiation resistance study.

1. Introduction, radiation damage in liquids

Liquids lack fixed internal structure, so the effect of radiation for them is limited to radiolysis, the dissociation of molecules by ionizing radiation or neutrons - unlike in solids for which the impact on mechanical or crystalline properties is also relevant. Radiolysis alters the chemical composition of the liquids, primarily through formation of free radicals. This can lead to increased viscosity, polymerization, evolution of gaseous compounds, loss of dielectric strength, increased acidity. In fully fluorinated fluids, like perfluorohexane C₆F₁₄ or perfluoroketone C₆F₁₂O (Novec 649 [1], or C6K), the formation of acids from radicals can occur in presence of moisture or hydrogen-containing impurities¹. All these effects, at elevated radiation doses (a few kGy to hundreds kGy), might cause visible damages to the cooling systems and changes in their performance, especially in 2-phase applications. A comprehensive study [2] of perfluorocarbons (PFC) C₃F₈ and C₆F₁₄ at CERN demonstrated a high radiation resistance of purified PFC fluids, characterized by a relatively low radiolysis yield and negligibly small production of hazardous compounds. This can be qualitatively understood as a consequence of the tight molecular structure of these compounds, with the structural C-C bonds reinforced by the presence of multiple very strong C-F bonds [3]². On the contrary, the radiolytic properties of perfluoroketones have not been studied at all. It is established that C6K molecules are photolyzed by the UV (with the peak absorption at 307 nm or 4.04 eV) [9]. This property, explaining the very low GWP of C6K, might also result in a lower radiation resistance [4], compared to perfluorocarbons.

2. Estimates for SciFi Tracker application

In the SciFi tracker application [5], C6K is proposed as a coolant in the mono-phase liquid cooling system for silicon photodetectors (SiPMs). The expected radiation dose at the SiPM location is fairly low, <80 Gy, <10¹² neq/cm². Order-of-magnitude estimates, even with exaggerated pessimistic

¹ An organic molecule can be cleaved at its weakest bond in the process of recombination and de-excitation following ionization. In C6K the weakest are the C-C bonds next to the carbonyl C=O group. The fate of the resulting radicals very much depend of the fluid purity. One likely scenario for pure fluids is a recombination. The impurities will cause irreversible chemical transformations. For example, in presence of water, the resulting (CF₃CF₂CO)⁻ radical can form pentafluoropropionic acid (PFPA) CF₃CF₂COOH. In presence of O₂ and water, assuming for the C6K radiolysis the same scenario, as for the photolysis in air [10], the principal products will be CF₃C(O)F and COF₂ (carbonyl fluoride). CF₃C(O)F will further be undergo hydrolysis to give trifluoroacetic acid, while COF₂ will hydrolyze CO₂ and HF.

² Another consequences are their chemical inertness of perfluorocarbons and low viscosity of the liquid phase.

assumptions, show that the expected radiation effect on the coolant will be negligible. First, because of the coolant circulation, only a minor fraction of it is exposed to radiation at the detector. Second, the total amount of the coolant that can be transformed by this radiation turns out to be at a ppm level. It is also appropriate to note that the inline rectification of circulating coolant will be constantly removing the radiolysis products.

2.1 The dilution factor

The peak total absorbed ionization dose and neutron fluence quoted in the TDR [5] are calculated for the SiPMs at the periphery of the 5x6 m² tracking planes. The fluid in the cooling pipes near the middle of the tracker plane will be exposed to the same amount of radiation³. However, the fluid in the large connecting line(s) located farther away from the SiPM (say, in 40-50 cm) will get 30-50% less dose. The dose also decreases towards the corners of the tracker planes. We shall conservatively estimate the total dose reduction by factor **1.5**.

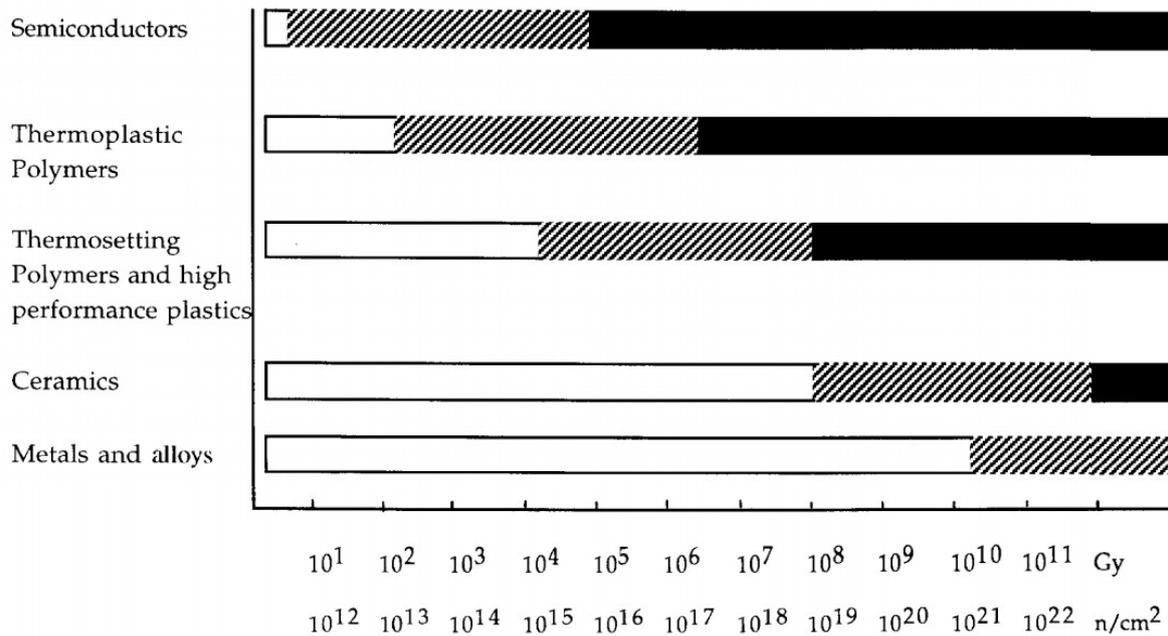
The volume V_d of the coolant exposed to radiation in all pipes around the tracker (including the connecting pipes) is only a fraction of the total coolant volume V_t circulating in the system. Therefore, the dose absorbed by the coolant will be further “diluted” by the factor $f=V_d/V_t$.

Both V_t and V_d depend on the manifolding scheme (serial or parallel). The TDR [5] refers to the serial connection scheme, with $m_c=330$ kg of Novec 649 or C_6F_{14} . The parallel connection will require a bigger volume of all connecting pipes, because a) they will transport (~6 times) higher fluid flow and b) in addition to one feeding pipe per branch, there will be a return line of the same cross-section.

- *Serial connection.* Branch length is $L_b=6*53=318$ cm. Cooling pipe radius r_c is 2 mm, its cross-section is $S_c=0.126$ cm². The cooling pipe volume, including U-bends (10%) is $6*S_c*L_b\approx 44$ ml per branch of 6 modules. A ~2 times thicker horizontal feed pipe has the cross-section $S_f=0.5$ cm² and $V_f=159$ ml. Total per branch: $V_b=44+159=203$ ml. Total per detector = $203*48 \approx 10$ l. The vertical connection/manifolding lines will have a comparable volume, so the total volume V_d of the coolant at the detector is ≈ 20 l. At -40°C, the coolant density is ~ 1.8 kg/l, so the total coolant volume will be $330 \text{ kg}/1.8 \text{ kg/l} = 183$ l and the dilution factor will be $f_{serial}=20/183\approx 0.11$
- *Parallel connection.* Cooling pipe volume, assuming the same S_c as for the serial case and including two 40 cm-long in/out connections per module, is roughly 110 ml. Two identical feed and return lines with the 6 times larger cross-section $S_f=6*0.126=0.76$ cm² have the volume $V_f=S_f*318*2=480$ ml. Total per branch is $V_b=110+480=590$ ml, the total per detector is $0.59*48=28$ l. Including similar vertical side connections we obtain $V_d \approx 60$ l. The total volume of the infrastructure pipework (transfer lines, chiller, pump) will have to be significantly larger than for the serial case, to support the ~6 times higher flow rate. My guesstimate is minimum ~ 240 l. We arrive to the dilution factor $f_{parallel}=60/(240+60)=0.2$, for the total coolant mass $m_c=300*1.8=540$ kg.

For further estimates I assume the second, more pessimistic, scenario, that is $f=0.2$ (and $m_c=540$ kg). Including the above mentioned factor 1.5, we arrive to the overall downscale factor $0.2/1.5=0.13$ for

³ Note that ionization losses in fluorocarbons are close to that of Si (1.664 MeV cm²/g). For example PTFE has $dE/dX = 1.671$ MeV cm²/g [PDG].



the dose and the integrated neutron fluence for the coolant. This gives **11 Gy** and $\sim 0.13 \cdot 10^{12}$

Figure 1: General appreciation of radiation damage to materials (from [6]). No damage (blank), mild to severe (shaded) or destruction (black). The horizontal axis shows the ionizing dose in Gy and the approximate equivalent neutron fluence (note that the effect of 1 Gy roughly corresponds to 10^{11} neq/cm²).

neq/cm², respectively. By all measures, these are very small values for most of materials (see Fig. 1).

2.2 The upper limit for the radiolysis yield

The efficiency of the chemical effect of radiation (further referred to as a radiolysis yield) for chemical compounds is usually characterized by the G-value, the number of molecules, radicals or ions produced or transformed per 100 eV of absorbed ionizing radiation, at the final (chemical) stage of the particle track⁴. In particular, G(-M)-values represent the number of starting molecules permanently changed [7]. The G_{tot} value for all products is, typically, about 2-3 for aliphatic ketones and for the perfluorocarbon C6F14 [7,8]. For further estimates, I assume a grossly exaggerated value $G(-M)=25$ for C6K which corresponds to the physical upper limit given by the energy 4 eV of the α -cleavage of the C6K molecule⁵.

Under this assumption, the dose of 1 Gy can dissociate $G \cdot 6.24 \cdot 10^{18}$ (eV/kg)/100 eV $\approx 1.6 \cdot 10^{18}$ molecules per kilogram of C6K. 1 kg of this compound contains $1000 \text{ g} \cdot N_A / M_w \approx 19 \cdot 10^{23}$ molecules. The estimated total radiolysis yield is, therefore, $1.6 \cdot 10^{18} / 19 \cdot 10^{23} \approx 0.82 \text{ ppm/Gy}$ or, for the expected dose of 11 Gy, **9 ppm**. In other words, not more than $\sim 540 \text{ kg} \cdot 10^3 \cdot 9 \cdot 10^{-6} \approx 4.9 \text{ g}$ (or about 2.7-3 ml, less than a teaspoon) of the coolant will be transformed during the entire lifetime of the project.

⁴ The "chemical" stage starts after $\sim 10^{-6}$ s, after recombinations, de-excitations and all other fast secondary effects have occurred.

⁵ The unknown real value can be only less than that – by up to factor ~ 10 .

The expected effect of neutron irradiation is even smaller. If we assume, following [6] (Fig. 1), that the effect of 10^{11} neq/cm² is roughly equivalent to 1 Gy, then the expected radiolysis yield of the total fluence of $\sim 0.13 \cdot 10^{12}$ neq/cm² will be equivalent to the effect of 1.3 Gy, that is $1.3 \text{ Gy} \cdot 0.82 \text{ ppm/Gy} \approx 1 \text{ ppm}$.

We can try to obtain an upper limit for the neutron effect differently, by computing the total effective surface S_c of the coolant exposed to the neutron flux and taking an exaggerated assumption that all neutrons crossing this surface will be absorbed in the coolant. The biggest pipes (the feed, or return, or vertical connection pipes, see the previous Section) in the exposed area will each have the effective square inner cross-section of $(0.87 \text{ cm})^2$. The two pipes will have the combined width of < 2 cm. For safety, we take 5 cm and obtain $S_c = 0.05 \text{ m} \cdot 2 \cdot (5 \text{ m(H)} + 6 \text{ m(W)}) \cdot 12 \text{ planes} \approx 13 \text{ m}^2$. Assuming 1 MeV/n, the total “guesstimated” deposited energy will be $13 \cdot 10^4 \cdot 0.15 \cdot 10^{11} \cdot 10^6 \approx 2 \cdot 10^{21} \text{ eV}$ which will dissociate $G \cdot 2 \cdot 10^{21} / 100 \approx 5 \cdot 10^{20}$ molecules out the total of $540 \cdot 19 \cdot 10^{23} \approx 10^{27}$ molecules of the coolant. This corresponds to **0.5 ppm**, which qualitatively agrees with the above estimate.

Thus, the expected combined radiolysis effect on the SiPM coolant will be microscopic. Furthermore, only a fraction of the radiolysis products will represent a potential hazard (e.g., acids), especially taking into account that most of such products will be removed by inline coolant rectification facilities (filters, ad/absorbers). ~~In addition, one can speculate that a significant part of non-acid C6K radiolysis products will be similar to those of C₆F₁₄ radiolysis considered to be harmless or tolerable.~~

The above estimates refer to a hypothetical pure compound in vacuum. A presence of dissolved gases and, especially, water will increase the chemical impact of the radiation due to the high chemical activity of the radicals and, especially, the important role of oxygen in their fate¹. ~~However, these effects should be largely similar for C6K and C₆F₁₄.~~

For C6K, the hydrolysis effect of liquid water, which is soluble in C6K at the level of **20 ppm** by weight [1] and with which C6K can directly react producing PFFA, *appears to be more important than the radiolysis at the doses expected at SciFi.*

3. Conclusion

Simple estimates, under most pessimistic assumptions, show that the effect of irradiation of the C6K with the doses expected in the SciFi application will be negligibly small, with the total radiolysis yield of 1 to 10 ppm by weight. Given that only a fraction of the radiolysis product will represent potential hazards (e.g., PFFA), it seems that the pilot validation study should mainly focus on the chemical reactivity of C6K with expected contaminants (first of all, with water) and methods of inline rectification. However, using C6K in other applications, with orders of magnitude higher expected doses, will certainly require a detailed radiolysis study.

References

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 - b. G-value (is) defined as the number of product molecules formed (or initial molecules changed).
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Appendix: Trivia

$$\text{Gy} = 1 \text{ J/kg}$$

$$1 \text{ R} = 0.00877 \text{ Gy}$$

$$N_A = 6 \cdot 10^{23}$$

$$1 \text{ J} = 6.24 \cdot 10^{18} \text{ eV}$$

$$\text{Average C-C bond energy} = 3.6 \text{ eV [1]}$$

$$E = hc/\lambda \approx 1242 \text{ (eV)/}\lambda(\text{nm})$$

$$1 \text{ eV} = 1.6 \cdot 10^{-19} \text{ J}$$

$$1 \text{ eV/ion} = 96 \text{ kJ/mol}$$

$$\text{C6K} = \text{C}_6\text{F}_{12}\text{O} (M_w = 316)$$

Wikipedia, "[Bond-dissociation energy](#)"