Pollution of liquid Argon due to neutron radiation

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Abstract

The purpose of the neutron facility installed at SARA is to investigate the behaviour of various materials to be used in the ATLAS electromagnetic calorimeter and the pollution of the cryostat liquid, under fast neutron radiation. So far, pieces of FR4, G10, prepreg adhesive and some epoxy have been irradiated and the charge signal of a α-cell immersed in the liquid argon has been measured before and after irradiation. Preliminary results concerning pollution are presented.

With a luminosity equal to $10^{34} \text{cm}^{-2}\text{s}^{-1}$, very high dose rates are expected in the ATLAS detector. In the electromagnetic calorimeter, the neutron fluence integrated over 10 years should figure out at $2 \times 10^{14}$ up to $4 \times 10^{15}$ in the barrel and in the endcaps, respectively, and one fears the liquid to become polluted with electronegative elements released in the active medium through the irradiation. Since no one wishes to have to empty the cryostat and refill it with cleaner argon too often during the ATLAS lifetime, it is therefore desirable to make sure that the argon will not get contaminated too quickly over certain threshold, say a few ppm equivalent oxygen.

1 EXPERIMENT

A neutron facility has been installed at the SARA accelerator in Grenoble. Its features have been fully described in an earlier paper [1]. A thick beryllium target is bombarded with a 20 MeV deuteron beam. Its intensity can reach a steady $7 \mu\text{A}$. A mean fluence up to $3 \times 10^{14}\text{cm}^{-2}$ can be obtained in about two days, over an area $8 \times 8 \text{cm}^2$ and a 8
Figure 1: Charge spectra before irradiation: the smooth lines are the result of a gaussian fit. On the top figure, the peak on the left is the noise, cut with the threshold.

cm depth. The neutron mean energy is 6 MeV. The pieces of materials to be tested are immersed in a liquid argon cryostat close to the neutron source. It is assumed that the pollution rate is a surface more than a volume effect. Therefore the relevant parameter will be the total area of the samples normalised to the liquid volume. In order to measure the pollution more easily, this normalised area has been made larger than in the calorimeter itself, by a factor sometimes greater than ten. An ionisation chamber is placed in the bottom of the cryostat. Its gap is 0.8 mm and a $^{241}$Am α-source has been deposited on its cathode. The collected charge is measured at several electric field values ranging from 5 to around 20 kV/cm.

Three sets of data are collected on the same analog channel, from the charge preamplifier to the QDC: the α signal, the pedestal (enhanced on purpose) and a calibration signal. The latter is obtained by feeding the test input of the preamplifier with a calibrated charge pulse. Thus one defines a relative charge for the α signal: $Q = \frac{\alpha\text{-signal}}{\text{test}}$. In this way, one gets rid of possible gain fluctuations over the run period. An example
The ratio of the charge signals before and after the irradiation (attenuation factor) is related to the pollution through the expression:

$$\frac{Q(\text{after})}{Q(\text{before})} = \frac{\lambda}{d} \times (1 - \exp\left(-\frac{d}{\lambda}\right)),$$

where $\lambda$ is the charge carrier absorption length and $d$ is the gap width of the $\alpha$-cell. In the case of oxygen pollution, $\lambda$ has been shown [2] to be best fitted with:

$$\lambda(\text{cm}) = (0.14 \pm 0.03) \times \frac{E(\text{kV/cm})}{\rho(\text{ppm})}.$$  \hspace{1cm} (2)

Much care is taken to fill the cryostat with very clean argon. Then, the relative charge is measured several times over a few days to make sure that everything is stable, at one or two field values. Eventually, its dependence on the field is recorded. It will be the zero-pollution reference. After the irradiation has started, it is measured again regularly at the same field values to observe the evolution of the pollution. At this stage, the charge spectrum exhibits a stronger and stronger contribution from $\beta^-$ signals mostly due to the activation of Argon: $n + {}^{40}\text{Ar} \rightarrow {}^{41}\text{Ar} \rightarrow {}^{41}\text{K} + \beta^-$. After the irradiation, the evolution is checked again and once the $\beta^-$ rate has gone very low, a new field curve is recorded again.

The charge as collected in the $\alpha$-cell during a particular run is displayed on figure 1.
2 OXYGEN AND NITROGEN TEST

The whole setup was tested by introducing known contaminants into the cryostat and the attenuation of the charge signal was determined each time. They were oxygen (about 3 and 6 ppm) and nitrogen (about 400 ppm). A few hours were necessary each time to reach an equilibrium. Results are shown on figures 3 and 4. If the oxygen data are well

![Figure 3](image)

Figure 3: Signal loss due to oxygen and nitrogen impurities. The curves are the result of a fit according to equation 1.

![Figure 4](image)

Figure 4: Absorption length of charge carriers in liquid argon with oxygen and nitrogen impurities. The lines are drawn according to equation 2.

fitted using the relations of reference [2], the nitrogen data were rather a surprise, since the first idea that comes to mind is that the absorption length should increase when the electric field is raised.

The nitrogen pollution of liquid argon has been studied before [2, 3], but not in the same range of field or concentration and the variation of the absorption length as seen here in presence of nitrogen could not be evidenced then. In fact, such a behaviour is not unique and it has been explained many years ago [4, 5]. The absorption length \( \lambda \) is related to the attachment rate of the drifting electrons \( k_s \): \( \lambda = d/k_s[S] \), where \([S]\) is the impurity concentration. The attachment rate on \( O_2 \), \( N_2O \) and \( SF_6 \) in liquid argon has been studied by Bakale et al. [4]. It is seen that \( N_2O \) is a much stronger absorber than oxygen and that contrary to oxygen, the attachment rate increases with the field. There is no universal law about the variation of the absorption length versus the electric field, it depends on the nature of the impurity.

If one extrapolates the \( O_2 \) \( k_s \) curve of ref. [4] to a field of 10 kV/cm and one uses the 2.8 ppm derived from the fit, the calculated absorption length value is exactly as measured in this test.
3 RESULTS

The materials which have been irradiated so far are FR4, G10 (von Roll Isola 64220 and 64060), prepreg (CTMI 5512-1383-1808-50/110) and epoxy Ciba-Geigy 5052. Although neither fluences nor normalised areas were identical during these four runs, the results have been summarised on figures 3 and 3.

Figure 5: Signal loss due to impurities released after irradiation. Error bars are purely statistical.

The very first observation which can be made is that the main contaminant released in the liquid is not oxygen, and nothing at this point can tell us about its nature. At the end of some of the runs, part of the liquid argon was evaporated into a bottle and sent off for chromatography analysis. Traces of oxygen and nitrogen were identified but in no case compatible with the observed attenuation. One may have noticed on figure 1 that, while evaporating the liquid, the impurity level in the remaining liquid seems to concentrate accordingly. The very last litre of liquid was recovered once and analysed, but although impurities were more abundant, they could not explain the absorption length data.

We made an attempt to normalise all our data at a field value of 10 kV/cm as in ATLAS, for a fluence equal to $1.6 \times 10^{14}$ neutrons/cm$^2$ and a value of the normalised sample area equal to 70m$^2$/m$^3$LAr, in order to compare the pollution rates coming from the
Figure 6: Absorption length of charge carriers in liquid argon after irradiation. Error bars are purely statistical.

Various materials tested so far. The results are shown in Table 1. The quoted uncertainties are estimates. They nevertheless indicate that the comparison of the attenuation factors is meaningful.

The contamination was suspected to come possibly from halogens, but it must be pointed out that FR4, which contains bromine whereas G10 does not, gives the most encouraging results after the epoxy 5020. The prepreg tested contains 4% chlorine and it has not been possible so far to make a test with a lower percentage product.

4 CONCLUSION

As already stated, the main source of contamination is not oxygen and its real nature is not known yet.

The signal loss expected in the EM calorimeter for a given amount of pollution can be derived from these data. If one takes into account the difference in gap thicknesses and in shaping times between the present test and the calorimeter, the signal loss is found to be a factor 3 lower when calculated using Mathematica software. Keeping in mind that the normalised area of the samples was sometimes a factor 10 larger in the test, one is entitled to conclude that the pollution level due to neutron irradiation in ATLAS should be bearable even after 10 years running and that the accuracy in deriving the energy in the calorimeter should be preserved.
Table 1: Alpha-signal loss after irradiation. Data are normalised to a fluence of $1.6 \times 10^{14}$ neutrons/cm$^2$ and to a ratio (open area of samples/liquid argon volume) equal to 70m$^{-1}$.

<table>
<thead>
<tr>
<th>material</th>
<th>signal loss(%)</th>
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<tbody>
<tr>
<td>epoxy</td>
<td>0. ± 1</td>
</tr>
<tr>
<td>FR4</td>
<td>4. ± 1</td>
</tr>
<tr>
<td>G10</td>
<td>8. ± 2</td>
</tr>
<tr>
<td>prepreg</td>
<td>15. ± 3</td>
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</tbody>
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References


