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Extension of the NPL Alanine Reference Dosimetry Service to 100 kGy.

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ABSTRACT

NPL operates a mailed alanine reference dosimetry service for absorbed dose to water in the range 20 Gy to 70 kGy. This report describes the work and findings to extend this range to 100 kGy.
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1 Introduction

NPL has provided a mailed alanine reference dosimetry service since 1991. The service originally operated over the range 70 Gy to 70 kGy and has been predominately used by the medical device sterilisation industry. In 1996 NPL extended the range to doses below 70 Gy for use in radiotherapy applications. Currently the minimum dose offered is 5 Gy and the maximum 70 kGy.

As part of the 2004-2007 NMS programme an investigation to extend the range upwards was undertaken. This short report describes the work and findings of this investigation.

As this extension will be used as part of the standard NPL mailed alanine reference dosimetry service it was decided to avoid altering the pellet composition or measurement parameters from those normally used. Above 50 kGy increased curvature of the response function causes an increase in uncertainty and the extension was therefore restricted to 100 kGy to ensure a minimal effect of the overall uncertainty budget.

Studies were made of the overall dose response, temperature effects and fading.

2 Equipment

All work in this report used the standard NPL alanine dosimeters[2]. The cylindrical (NPL 'G type') polyacetal holders were used each containing four alanine pellets. Alanine pellets from three batches (Batch 60, 61 and 63) were used. Pellets are of 5 mm diameter and 2 mm height consisting of 90% alanine powder and 10% paraffin wax (98 °C melting point). The pellets mass range was within 52-58 mg.

All irradiations were made in the standard NPL four position holder (levels one and two only) using the NPL Gammacell 220 high dose cobalt-60 irradiator. Irradiation dose is determined by timing and a known dose rate. The dose rate is traceable to the primary standard and during the period of this work ranged from ∼6-10 kGy / hour. The uncertainty on the irradiation dose is 2.2% (k=2). Irradiation temperature ranging from 25 to 55 °C was controlled (by use of a Peltier junction and liquid carbon dioxide) to better than ±2 °C throughout irradiation.

On irradiation alanine produces stable free radicals and the relative concentration of these radicals can be determined by electron paramagnetic resonance (EPR) spectrometry. The free radical concentration is proportional to dose over a wide range, is relatively stable and has a small temperature coefficient. EPR is non-destructive readout.

NPL has two EPR machines for alanine measurements. The machines are Bruker EMX models, X-band (9.75 GHz, Bres 3480 G) with 8 and 9" magnets. The microwave bridges are software controlled and feature automatic tuning. Both machines (NPL identifier EMX1 and EMX2) were used for this work. The EMX1 uses a standard Bruker st4102 rectangular cavity and the EMX2 uses a standard Bruker
Measurements were made both manually and, for comparative measurements only, with the slider automatic pellet handling system. All pellets were measured individually and corrected for their measured mass.

Response has been calculated in terms of the peak-to-peak height of the central feature of the spectrum (henceforth called pp in this report) - the standard NPL method for doses above 20 Gy.

3 Initial studies

The first step was to determine the general viability of extending the service to 100 kGy, in particular to check the suitability of current instrument parameters and the shape of response curve. A set of sixteen dosimeters were irradiated to 50, 60, 70, 80, 90, 100, 110, 120 kGy, two dosimeters at each dose, irradiation temperature 25 °C. These were measured on EMX1 using the standard measurement parameters. Initial results from these measurements looked promising. The response up to 120 kGy was sufficiently large to enable measurement without saturation of the signal.

Figure 1 shows the results fitted with a general 3rd order weighted fit. Figure 2 shows the same results as fractional residuals about the fitted line and figure 3 the same data but in terms of dose percentage residual.

At the same time as the above measurements were made a set of standard checks (0.5 - 72 kGy) were included with the extension irradiations.

Figures 4 and 5 show, as fitted line and residual respectively, the standard check data and extension data combined.

The graphical results confirm the practicability, in general, of extending the line to higher doses without needing to alter the principal measurement parameters.

Following on from these results it was decided to plan a series of irradiations of nearly the complete current range for standard NPL alanine extended upwards to 120 kGy.
Figure 1: Initial studies, fitted third order line

Figure 2: Initial studies, fitted third order line, fractional residuals
Figure 3: Initial studies, fitted third order line percent residuals

Figure 4: Full line (standard plus extension), fitted third order
4 Full line irradiations

4.1 Determination of response over a “Full” calibration line.

As part of the pre-issue procedure[2] for NPL alanine reference dosimeters a series of dosimeters are irradiated to a range of doses between 0.07 to 72.4 kGy. The measurement data (EPR pp) from these irradiations providing the calibration coefficients used to determine doses.

Due to the inherent nature of EPR machines it is not possible to assume pp measurements on one day will necessarily match those on another or that pp measurements on one machine will match those on a different machine. Hence the critical need for reference and / or check dosimeter measurements for every measurement run. This also means it was not possible to directly compare pre-issue measurements with new measurements without some form of normalisation.

To evaluate the response of irradiations at doses greater than 72 kGy a set of twenty four dosimeters (Batch 63) were irradiated to 9.1, 12.8, 18.1, 25.6, 36.2, 51.2, 72.4, 78.9, 86.1, 93.0, 102.4, 111.7 kGy, two dosimeters at each dose, irradiation temperature 25 °C. The doses up to 72.4 kGy were selected to match the higher range part of the standard pre-issue irradiation doses for the standard NPL alanine - these progress in a ratio of $\sqrt{2}$. To increase, relatively, the number of dose points in the range from 72 kGy to 120 kGy a smaller progression ratio was selected (of 1.19) to give dose points at 78.9, 86.1, 93.0, 102.4, 111.7 kGy.
These were measured on both EMX1 and EMX2 instruments using the standard measurement parameters.

Based on the respective EMX1 and EMX2 pre-issue lines the expected pp can be calculated (conveniently done using NPL Scilab function alXXpp) for doses up to 72 kGy. These calculated pp values can then be compared to the actual values measured. This provides a normalisation factor, which can be applied to all the measurements from 9.1 kGy to 111.7 kGy i.e. over the whole range. This method of normalisation was used for both EMX1 and EMX2 results.

The current standard NPL alanine pre-issue line i.e. ranging from 0.07 to 72.4 kGy is currently fitted, using a third order weighted fit (weighting = 1/dose^2), with two spans for doses <= 10^4 Gy and doses >= 10^3 Gy respectively using a weighted least squares procedure.

Analysis of the extension results indicated that a fitting function based on three spans would be better than attempting to span the full dose range in just two. The spans decided on are for doses less than 10 kGy, between 1 kGy and 73 kGy and between 10 kGy and 112 kGy.

Figures 6 and 7 show the fitted line and data for measurements on EMX1 and EMX2 respectively. Shown in red is the expected response based on the previously determined calibration lines.

Figures 8 and 9 show the fitted line and data for measurements on EMX1 and EMX2 respectively in terms of fractional dose residual about the fitted line.

In terms of dose the pellet-to-pellet residual variation is 0.45% (k=1, EMX1) and 0.42% (k=1, EMX2). This is well within the accepted coefficient of variation 0.8%
Figure 7: Full line irradiations, fitted line, EMX2

Figure 8: Full line irradiations, fitted line, EMX1, percent dose residual
(peak-to-peak values)[2]. As such the current uncertainty budget is valid for the extension to 100 kGy.

5 Temperature studies

Previous studies[3] at NPL have shown a small but not insignificant temperature effect of about +0.143 % per °C (pp relative response to 25 °C response). The previous work did not show any dose dependence effects over the dose range studied i.e. 1 - 40 kGy. Currently this value is used for all NPL standard reference alanine dosimeters. To test this linearity with respect to dose at higher doses was still applicable a series of high dose irradiations at various temperatures were made. Irradiations to doses of 51.2, 72.4, 86.1 and 102.4 kGy each to temperatures of 25, 35, 45, and 55 °C were made.

Analysis of the results by comparing the pp at temperature t to that at 25°C are shown graphically in Figure 10.

As can clearly be seen from graph the previous linearity assumption with dose is not valid for doses above 50 kGy. The dashed line shows the standard 0.143% per °C coefficient. The 51 kGy irradiated dose shows good agreement with this value. However the 72, 86 and 102 kGy values show an increasing variation to the standard value.

The respective coefficients (1st order plot: pp relative response to 25 °C versus irradiation temperature) for the doses are shown in table 1 below. Also included
Figure 10: Irradiation temperature coefficient study

<table>
<thead>
<tr>
<th>Dose / kGy</th>
<th>Temperature coefficient</th>
</tr>
</thead>
<tbody>
<tr>
<td>102.4</td>
<td>0.00210</td>
</tr>
<tr>
<td>86.1</td>
<td>0.00189</td>
</tr>
<tr>
<td>72.4</td>
<td>0.00171</td>
</tr>
<tr>
<td>51.2</td>
<td>0.00138</td>
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<tr>
<td>70</td>
<td>0.00158</td>
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</tr>
<tr>
<td>10</td>
<td>0.00128</td>
</tr>
<tr>
<td>1</td>
<td>0.00156</td>
</tr>
<tr>
<td>0.1</td>
<td>0.00142</td>
</tr>
</tbody>
</table>

Table 1: Bold is data from this report. Other data from earlier work[3]
in the table are similar values for doses below 70 kGy from earlier work at NPL[3].
This data is listed in the form required for NPL Scilab routines i.e. \( pp(25) = pp(t)[1+c(25-t)] \) where \( c \) is the tabulated data.

A polynomial function (2nd order) fitted to the tabulated data has been made to allow for temperature correction to be made at any dose up to 100 kGy. This data is shown in figure 11.

Similar temperature effects found in this work are also described in work by other authors[1]

### 6 Fading studies

An allowance for fading of about 0.25% per month (i.e. \( \sim \)3% per year), in terms of dose, at 20 kGy is made for the NPL reference alanine dosimeter. The fading is dose dependent and to determine the amount of fading at doses above 70 kGy a series of measurement sequences were made at varying times post irradiation.

Pre-conditioning and storage conditions can be critical to the response of alanine dosimeters especially with regard to post-irradiation fading[4]. All dosimeters were pre-conditioned as normal[2] and all dosimeters were kept together at all times under laboratory conditions (temperature controlled 22 °C ±2 °C, humidity uncontrolled.)

As mentioned earlier due to instrument instability on separate measurement runs it was necessary to make relative measurements using dosimeters irradiated at different
times and measured during the same measurement run.

The results from the experiments below are described in terms of change in peak-to-peak height. The effect on measured dose is greater than the effect on peak-to-peak height, as shown in figure 13.

Three sets of experiments are described, the first two (initial and 55 °C) in summary and the main fading experiments in detail.

6.1 Initial studies

Initial experiments involved two sets of dosimeters irradiated (25 °C to doses of 51.2, 72.4, 78.9, 86.1, 93.9, 102.4, 111.7, 121.8 kGy) approximately 60 days apart. The most recently irradiated set when measured two days and eight days after irradiation showed evidence of 0.4% growth. This indicated that growth effects could still be seen even after two days. The new and old sets were measured together eight days after the end of the most recent irradiation, i.e. after the initial changes were complete, to determine the fade over 60 days. These results indicated a fade of about 0.4%.

6.2 High temperature fading experiments.

This experiment was to determine if high temperature caused a significant change to the short term fading expected at lower temperatures. A set of dosimeters were irradiated to 98 kGy at 55 °C. These dosimeters measured with a set that had been irradiated 80 days previously, i.e. long enough for short term changes to have stabilised. No significant effect was seen between the 0.5 hour and 208 hour measurements post-irradiation. This contrasts to studies made with dosimeters irradiated at room temperature (see below), indicating that short term changes may be accelerated at higher temperatures and effectively take place during the irradiation itself.

6.3 Main fading experiments

For this study two sets of dosimeters were irradiated at 25 °C to doses of 51.2, 72.4, 78.9, 86.1, 93.9, 102.4, 111.7, 121.8 kGy approximately one year apart. By comparison of the response of older dosimeters to those of the new it was possible to determine the fading characteristics for a series of post-irradiation times.

A series of measurement sequences were made. Each measurement sequence as a series of interleaved pairs i.e. old dosimeters followed by new dosimeters for each dose point. The data were analysed by taking the ratio of the peak-to-peak heights of pairs of dosimeters irradiated to the same dose, but one year apart. In this way, variations in spectrometer response would cancel out and the known absence of short term changes in the dosimeters irradiated a year previously enabled changes in the newly irradiated dosimeters to be determined.
Figure 12: Variation of pp ratio up to 1500 hours

Over the first 2 to 3 days a maximum increase in pp of about 1.5% is seen. After about 3 days the ratio between the two sets of dosimeters stabilises, indicating a fade over the year of $\sim 1.6\%$. In terms of dose a 1.6% change in pp equates to approximately with 3-3.5% in terms of dose at 100 kGy. However this conversion is dependent upon the dose and is graphically represented in figure 13.

7 Conclusions

The study has confirmed the viability of extending the NPL standard alanine reference service from 70 kGy to 100 kGy. The extension does not require an alteration to the measurement parameters and the overall response is within the expected existing uncertainty. The change in response of the temperature coefficient with dose adds another level of calculation but does not effect any practical aspects. The fading results are slightly greater than expected but still within acceptable limits.
Figure 13: Relationship between change in pp and change in measured dose

References


