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How to improve calibration for analytical accuracy in ESR spectrometry

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1. Introduction

Electron spin resonance (ESR) has proved to be a very effective tool of dosimetry. Characteristics of Alanine/ESR dosimetry system is better suitable than routinely used personnel dosimeters for the long term radiation measurement in harsh condition of Nuclear power plant : This system is not significantly affected by temperature and humidity and also has low fading rate.[4] About 2 years ago, alanine and lithium compound dosimeters were installed at Wolsung unit 1 for the environment monitoring as a part of equipment qualification program. Afterward, the dosimeters were also installed at the other nuclear power plants.

The recovered alanine dosimeters in the period of maintenance were measured by E-scan alanine analyzer system or EMX ESR spectrometer. For the accurate measurement of the dosimeters, we have studied various source of errors. This paper discusses sources of inaccuracy related to data processing. Errors in data processing were researched by E-scan alanine analyzer system. E-scan alanine analyzer system proposes the fit function with best f-statistics but the user may choose a different one. This fact is useful for our research.

2. Methods and Results

L-a-alanine is a crystalline amino acid in which free radicals are formed at irradiation. These radiation induced radicals can be detected by means of electron spin resonance (ESR) spectroscopy. The signal of the irradiated alanine dosimeter has multiple peaks, but only the central peak (with the largest amplitude) in the blue region is used for calculating the intensity (Fig. 1). This signal is proportional to the amount of radicals in the measured sample.

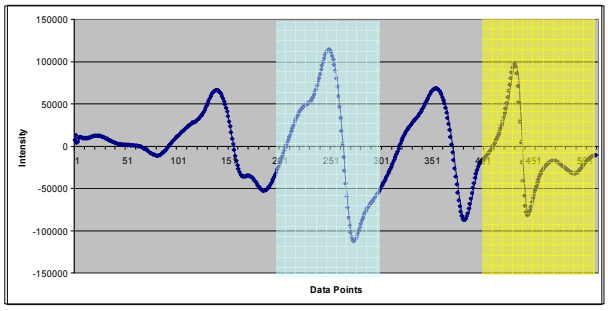

Fig.1 An ESR-Spectrum of 512 datapoints is acquired which is represented by 512 pairs of magnetic field strength and signal intensity.

The e-scan is a benchtop ESR (Electron spin resonance) spectrometer dedicated to the evaluation of absorbed dose in alanine dosimeters (either film or pellet). With the appropriate accessories the e-scan measures absorbed dose from a few Gray to about 200 kGy. The e-scan alanine system is equally suited for use in electron beam, X-ray, or gamma irradiation facilities.

The e-scan has a set of default parameters that are specifically optimized for each type of dosimeter insert. And, the e-scan needs a proper dosimeter calibration for the dosimeter insert (The dose ranges that are applicable for each e-scan dosimeter insert : PU - 20Gy – 500Gy, PL - 250Gy – 10kGy, PH - 2kGy – 80kGy).

2.1 Comparison of the calibration files.

To make a meaningful dose assessment the dosimeter measurements need to be compared with measurements made using certified dosimeters of known dose. This is achieved by performing the calibration routine which creates a calibration curve and corresponding fit coefficients from measurements made with a set of dosimeters with known certified dose. Therefore, the measurement accuracy is affected by calibration curve.

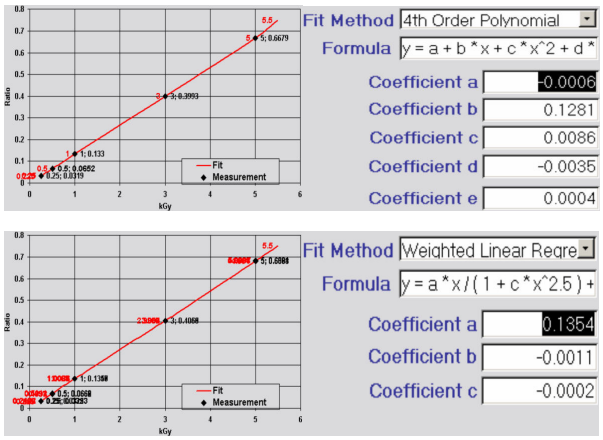

Fig. 2 Calibration files of the PL inserts (a)upper : First calibration file (with the typical procedure) (b) down : Second calibration file (with the reorganized procedure)

First, it makes the calibration file with the typical procedure. Second, it makes the calibration file with the reorganized procedure. In the case of the PL insert, this calibration files are Fig. 2.

First calibration file which was measured of the one time per calibration point was fitted fourth-order polynomial. But, the alanine dosimeter has a linear dose response from well below 1Gy up to 10^4 Gy [5]. First

calibration file result doesn't consider systematic errors. So, the calibration file needs a reorganized procedure. The calibration curves in ESR dosimetry are usually constructed by means of the least-squares technique in its simplest variant. The recommended alternative linearity test (Nalimov, 1963, Draper and Smith, 1981) requires several replicate measurements of Y at each used X value [2]. Also, measurement is subject to error, so repeat measurements will not be identical. A technical description of a linear calibration is assumed that the dispersion of the measurements is the same for each standard. But, in some cases, the standard deviation has to be specified separately for each value of concentration. In this case, it used the WLS (Weighted Least-Squared Regression) method instead of the OLS (Ordinary Least-Squared Regression) method.

According to the passage, it generalizes down to the reorganized procedure. This procedure is as follows:

1. Measurements of the five times per calibration point.
2. It draws a distribution chart.
3. Check of the linearity and the dispersion.
4. Outlier diagnosis with the static program.
5. Confirmation of the Calibration file.

2.2 The test with the NPL samples.

The dosimeters which used in the PL insert were irradiated surrounded by water equivalent material in a calibrated cobalt-60 gamma-ray field at a dose rate of 40 Gy per hour. The uncertainty associated with the dose to water measurement at the point of irradiation is estimated to be ±2.4 %. This was proven by the NPL (National Physical Laboratory) which issued certificate.

Alanine dosimeters which used in the tests are all the same. The below table is the test results of the PL inset, and the details for making dosimeter calibration files are described in the 2.1 of this paper.

Dose (Gy)	1st Calibration	Diff. (%)	2nd Calibration	Diff. (%)
250	252.76	1.10	242.77	- 2.89
500	513.18	2.64	497.75	-0.45
1000	1021.337	2.13	1002.11	0.21
3000	3074.117	2.47	2998.29	-0.06
5000	5124.26	2.49	5025.69	0.51

Table 1.Results of the PU insert test.

Results show differences by the calibration file, 2nd calibration which used reorganized procedure is mostly near the true values. In the case of the 250Gy dosimeter, the result deviates from the true value than the 1st calibration case. This result needs a complementary explanation of the other overlapped calibration file. Anyway, second calibration file with the reorganized procedure expressed an improved accuracy of the measurement value and a linearity of the calibration file.

Also, the other inserts made the calibration files with the same method. Especially, the calibration files of the PU insert and the PH insert divided in two. In the case of the PU insert, it considered a low radiation level (under 20Gy). And, the alanine dosimeter has a linear dose response from well below 1Gy up to 10^4 Gy [5]. So, In the case of the PH insert, it considered a high radiation level (over 10^4Gy).

3. Conclusion

ESR (Electron spin resonance) is the best of the available methods for determining dosimetry. However, like any other sophisticated high-precision tool, this method requires delicate handling. Its careless application without due attention to potential sources of errors may produce inaccurate results.

Above all, the data processing errors in the potential errors are incorrect model selection for fitting the data, and unsuitable design of the calibration experiment. We adopted the reorganized procedure, so the analytical accuracy of ESR spectrometry is improved. But, these results will be repeatedly cross-checked. Forward, to improve the accuracy, we will research into the lithium compound and the minor effect of temperature and fading. The more we study the method to enhance the measurement accuracy, we believe, the more we ultimately contribute to assessment of the level of radiation in NPP (Nuclear power plants).

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