An alpha irradiator for ESR dating

Rainer Grün† & Oliver Katzenberger-Apel§

†Quaternary Dating Research Centre, Australian National University, Canberra ACT 0200, Australia.
§Stupp Str. 3, 50823 Köln, Germany.

Due to the large sample size required for ESR measurements (10-20 mg), ESR alpha irradiation experiments on thin fine grain layers have rarely been reported (e.g. Yokoyama *et al.* 1981, Grün 1985, Mudelsee 1990). Some studies have used an accelerator to determine alpha radiation effects on speleothems (Lyons & Brennan 1989, 1991) others have experimented with ^{210}Po-doping (DeCanniere *et al.* 1986).

In order to perform alpha irradiation on thin fine grain samples, we have constructed a large ^{241}Am-irradiator (see Fig. 1). The irradiation chamber is constructed from a 150 mm ID stainless steel tube which is cut in two halves. The top contains the alpha source and a valve to flush the chamber with argon when not in use in order to avoid oxidation of the ^{241}Am foils. The bottom half contains an adjustable plate for holding samples and the collimator. The maximum distance between the source and the samples is 100 mm, the positioning can be read from a steel ruler. A side valve is used to evacuate the chamber to about 10^{-3} Torr.

The alpha source (Fig. 2) has a diameter of 100 mm and is built by four ^{241}Am smoke detector foils (Amersham glued the foils into holder that we supplied). Since the foils have an inactive strip of 1mm at each side, the alpha emission is not homogeneous. Therefore the sample holder has four fixed sample positions for disks with a diameter of 31.5 mm. In order to guarantee a steep impact angle (>65°) of the alpha particles, we have constructed a 64 mm high brass collimator. The use of metal material avoids electrostatic charging which may occur in plastic. Figure 3 shows the sample holder and the collimator.

The energy of the alpha particles at the surface of the foils is 4 MeV (according to Amersham). Each fixed sample position was calibrated against the alpha source of the Max Planck Institute in Heidelberg by TL using $CaSO_4$:Dy. The dose rates for position 1 to 4 in a distance of 65 mm using the collimator are 31.64±0.69, 27.73±1.05, 29.44±1.48 and 29.91±0.09 Gy h^{-1}, respectively. The errors result from repeated measurements.

We have carried out some alpha efficiency measurements. Fine grains were separated from ground samples with deposition in acetone as described by Huxtable (1979). About 20-22 mg were effectively deposited on the aluminium sample holders (diameter: 31.5 mm; thickness: 5 mm) which corresponds to an average thickness of about 9 to 10 µm. The range of 4 MeV alpha particles is about 15 µm in a matrix with a density of 2.95 g cm^{-3} (Rytz 1979). This energy is high enough for the alpha particles to pass completely through a 10 µm layer of constant thickness (with the above configuration).

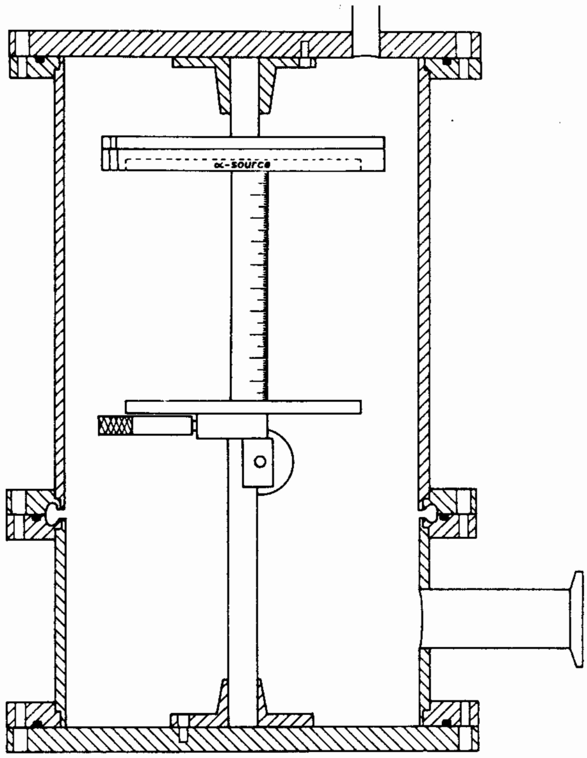
Gamma irradiation was carried out with a calibrated ^{60}Co source at Louvain-la-Neuve. Alpha and gamma irradiations were performed on aragonitic mollusc shells, corals and on tooth enamel (hydroxyapatite). ESR measurements were performed with a Bruker ER 200 tt ESR spectrometer at room temperature.

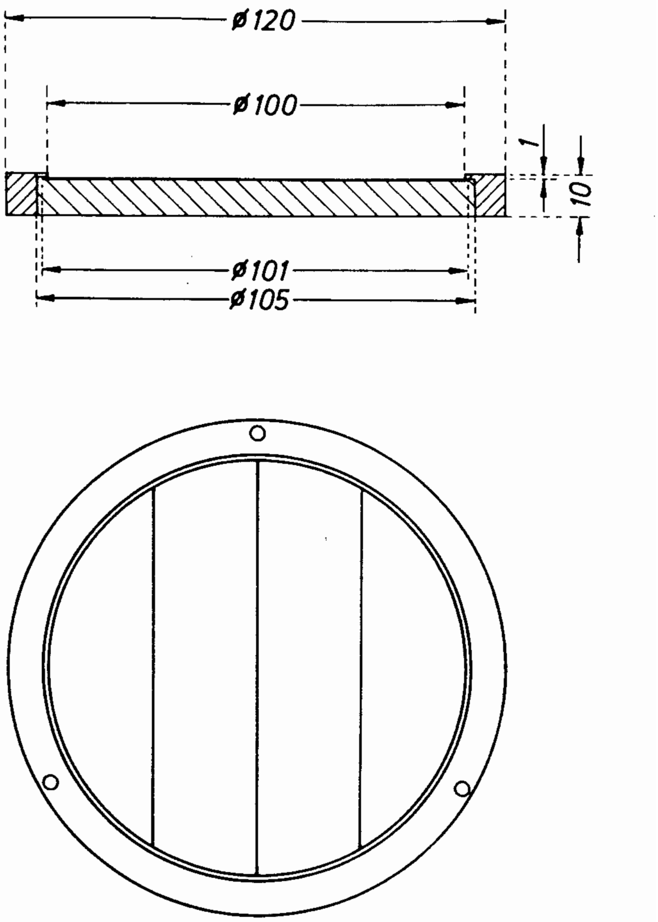
Figure 4 shows the dose response curves for gamma and alpha irradiation on recent enamel (g=2.0018) and mollusc shells (over-modulated signal with 0.63 mTpp; see Katzenberger, 1989), respectively. The best fits using a single saturating exponential function are shown in Figure 4. These curves have not been forced through zero. The scatter of the data points around this best fit may be used to assess the random errors that are

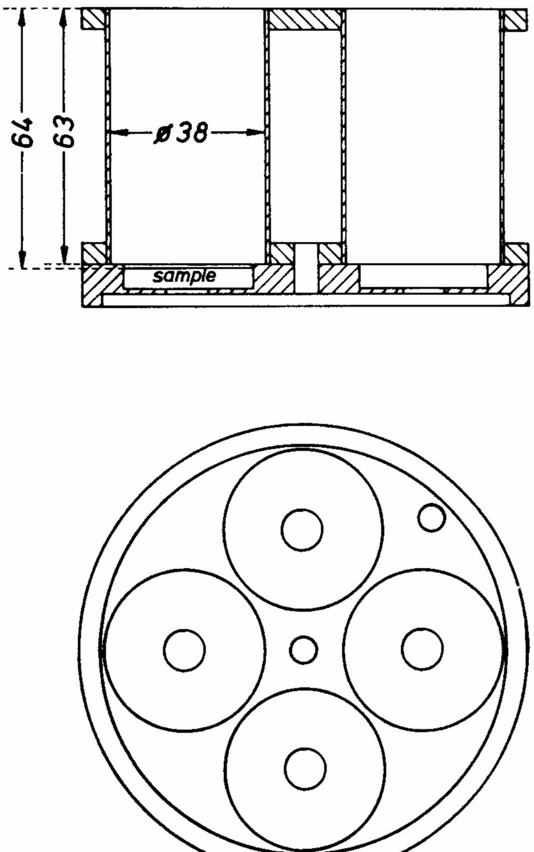
Figure 1. (Right)
Alpha irradiator for ESR samples: the alpha source (see Fig. 2) is mounted in the upper half of the stainless steel tube. The samples and collimator (Fig. 3) are placed on the adjustable plate. The chamber can be evacuated (top valve) and flushed with argon (side valve).

Figure 2. (lower right)
Alpha source and source holder (top: side view, below: frontal view). The source has a diameter of 100 mm and is constructed from four smoke detector foils.

Figure 3.(lower left)
Sample holder (top and below) and collimator (top). Dimensions are in mm. The two smaller holes in the sample holder are for fixing the positions of the samples.







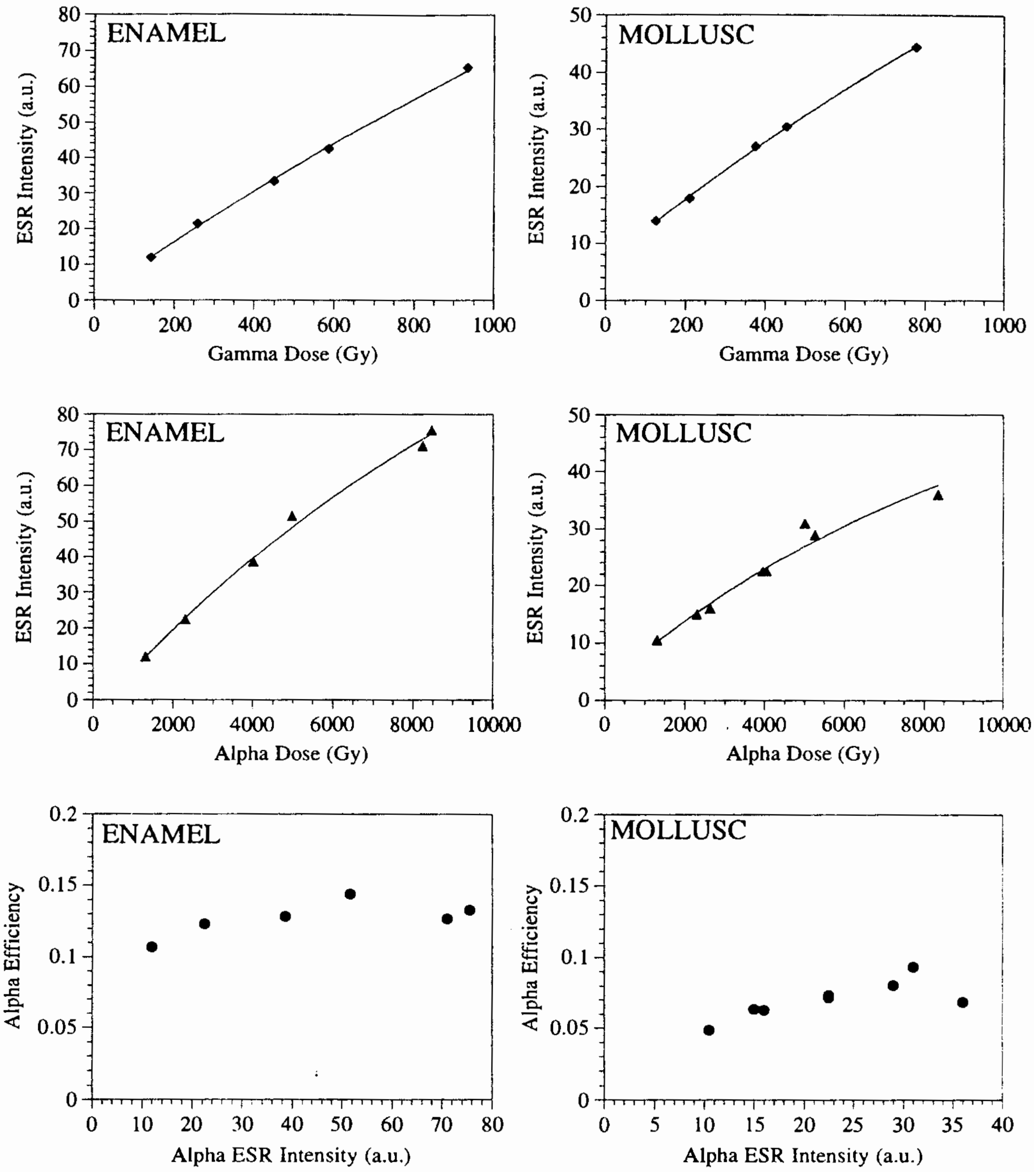


Figure 4.
Gamma (top) and alpha (middle) dose response curves for enamel (left) and mollusc shells (right). The lowermost diagrams show the alpha efficiencies by projecting the measured alpha intensity values onto the gamma dose response curves and calculating the ratio of projected gamma dose over alpha dose.

involved in the sample preparation and irradiation procedures. For molluscs the data points show a scatter of about 1.2% and 5.8% around the best fit for gamma and alpha irradiation (using the same sample pretreatment procedures), respectively. For enamel the corresponding values are 2.2% and 4.9%. This shows that the alpha irradiation causes an additional error compared to gamma irradiation which may be attributed to agglomeration of the fine grains. Considering a random error of up to 5% in the calibration plus a possible systematic calibration error of 5%, the precision of this configuration is in the range of 7% and the accuracy for dose measurements is probably in the range of 12%.

The lowermost diagrams of Figure 4 show the calculated alpha efficiencies for the measured samples. The corresponding gamma doses were calculated by projecting the measured alpha intensities onto the fitted gamma dose response curve. The efficiency value results from the ratio of projected gamma dose over alpha dose. Both data sets imply a more or less constant alpha efficiency over the whole dose range although the lowermost intensity values seems to display slightly lower efficiency values than the higher ones. This may be due to a somewhat larger signal to noise ratio. The average efficiency values are 0.127±0.011 for enamel and 0.070±0.012 for mollusc shells. The alpha efficiency values of corals presented by Radtke and Grün (1988) and Grün et al. (1990) of 0.06±0.01 and 0.05±0.01, respectively, were also obtained with this source.

Experiments with alternating alpha and gamma doses (Katzenberger, 1989) on mollusc shells do not show changes in the gamma sensitivity which implies that damage by alpha rays does not create additional trap sites in this material.

The alpha efficiencies of secondary carbonates measured with this source and obtained by others are all below 0.1 (for a compilation see Grün, 1992). This means that the contribution of the alpha dose rate to the total dose rate is relatively small and therefore uncertainties in the assessment of the total dose rate introduced by an unknown alpha efficiency (e.g. 0.07±0.03) are also relatively small (see also Lyons and Brennan, 1989; 1991). However, more measurements should be carried out before such a low alpha efficiency value can be assumed for all ESR signals in secondary carbonates.

The alpha efficiency of tooth enamel which has been repeatedly measured to be in the range of 0.11 and 0.15 is not negligible, particularly for samples with high uranium concentrations.

Acknowledgments
This work would have been impossible without the very active help of R. Berndt, head of the workshop at the former Institut für Kernchemie, Universität zu Köln, who constructed the alpha irradiation chamber. The alpha chamber is now located at the Laboratoire de Chimie Inorganique et Nucleaire, Louvain-la-Neuve, where also sample preparation and gamma irradiation was carried out. For providing their laboratory facilities and helpful discussions we are grateful to D. Apers, R. Debuyst, P. DeCanniere and F. Dejehet. We wish to thank Prof. G. Wagner and Dr. L. Zöller, Heidelberg, for the calibration of the source.

References

DeCanniere, P., Debuyst, R., Dejehet, F., Apers, D. & Grün, R. (1986) ESR dating: A study of ^{210}Po-coated geological and synthetic samples. *Nuclear Tracks* **11**, 211-220.

Grün, R. (1985) Beiträge zur ESR-Datierung. Sonderveröffentlichungen des Geologischen Instituts der Universität zu Köln **59**, 1-157.

Grün, R., Radtke, U. and Omura, A. (1990) ESR and U-series analyses on corals from Huon Peninsula, New Guinea. *Quaternary Science Reviews* **11**, 197-202.

Grün, R. (1992) Suggestions on minimum requirements for reporting ESR age estimates. *Ancient TL* **10**, 37-41.

Huxtable, J. (1979) Fine grain dating (F.G.D.). *Eur. PACT* **2**, 7-11.

Katzenberger, O. (1989) Experimente zu Grundlagen der ESR-Datierung von Mollusken-Schalen. Sonderveröffentlichungen des Geologischen Instituts der Universität zu Köln **72**, 1-71.

Lyons, R.G. and Brennan, B.J. (1989) Alpha-particle effectiveness in ESR dating: energy dependence and implications for dose rate calculations. *Applied Radiation and Isotopes* **40**, 1063-1070.

Lyons, R.G. and Brennan, G.J. (1991) Alpha/gamma effectiveness ratios of calcite speleothems. *Nuclear Tracks and Radiations Measurements* **18(1/2)**, 223-227.

Mudelsee, M. (1990) ESR-Datierung karbonathaltiger, quartärer Tiefseesedimente. Unpublished Diplomarbeit. Universität Heidelberg, 73pp.

Radtke, U. and Grün, R. (1988) ESR dating of corals. *Quaternary Science Reviews* **7**, 465-470.

Rytz, A. (1979) New catalogue of recommended alpha energy and intensity values. *Atomic Data and Nuclear Data* **23**, 507-533.

Yokoyama, Y., Quaegebeur, J.P., Bibron, R., Leger, C., Nguyen, H.V. and Poupeau, G. (1981) Electron spin resonance (ESR) dating of fossil bones of the Caune de l'Arago at Tautavel. In: DeLumley, H. and Labeyrie, J. (eds): *Absolute dating and isotope analysis in prehistory - methods and limits*, Proceedings Pretirage, 457-492.

PR Albrecht Wieser