^{41}Ca - a Biological Dosemeter for Neutrons?

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A project has been started in order to contribute to the neutron dosimetry calculations for A–bomb survivors in Hiroshima and Nagasaki. Within the present dosimetry system DS86 [1] thermal neutron fluences seem to be calculated lower than expected from the measurement of long–lived radionuclides produced via nuclear reactions during the atomic bomb explosions [2]. Beside another measurement already under way at this laboratory (the detection of ^{63}Ni ($T_{1/2}$ = 100 years) produced via fast neutron reactions on Cu [3], we initiated a feasibility study to measure ^{41}Ca ($T_{1/2}$ = 103 000 years) in teeth (produced via thermal neutron capture on ^{40}Ca). If successful, the detection of ^{41}Ca in teeth would represent the first biological fingerprint of neutron exposure of A–bomb survivors.

The main constituent of tooth material is calcium hydroxyapatite crystal. Tooth enamel itself consists to 36% of Ca. It shows little if any metabolism after the time, when the formation of that specific tooth is completed. Its crystalline structure offers the possibility to determine γ–ray doses by use of the electron spin resonance (ESR) method [4]. Thus, the measurement of ^{41}Ca in tooth material will deliver both, information on neutron and γ–ray exposure in the same biological material.

An essential prerequisite for the applicability of this method is the knowledge of the natural ^{41}Ca/^{40}Ca isotope ratio. Several years ago, these ratios were thoroughly investigated in order to establish dating with ^{41}Ca, which could work in an analogous way as the radiocarbon–method. Due to the longer half–life of ^{41}Ca compared to ^{14}C (about 20 times) dating back to the middle Pleistocene period would have been possible. It turned out, however, that the natural ^{41}Ca/^{40}Ca isotope ratio shows remarkable fluctuations depending on the lithosphere–constitution, erosion rate and shielding of the cosmic radiation by the earth's geomagnetic field. Differences in the natural calcium isotope ratio of more than two orders of magnitude were observed [5]. In our case the variations in this natural ^{41}Ca/^{40}Ca ratio are expected to be much lower for several reasons: a) only an isolated region around Hiroshima is examined; b) a significant amount of the food comes from the sea and its ^{41}Ca content should be low; c) around Hiroshima the erosion rate is high lowering the natural ^{41}Ca/^{40}Ca ratio, too. From these considerations both, a much lower natural ^{41}Ca background in tooth materials and much smaller fluctuations than observed in former investigations are expected.

Using the actual dosimetry system (DS86) for the Hiroshima–case an additional amount of ^{41}Ca/^{40}Ca of about 10^{-14} is expected for distances of 1 000 m from the hypocenter. The only method, which is sensitive enough to measure isotope ratios at this level is accelerator mass spectrometry (AMS) (decay counting suffers from the long half–life and the disadvantageous decay scheme of ^{41}Ca, whereas e.g. the sensitivity of resonant laser ionization mass spectrometry (RIMS) is a few orders of magnitude too low). Another advantage of AMS compared to decay counting is that few mg of sample mass is sufficient.

The interfering stable isobar for AMS measurements, ^{40}K, readily forms negative ions. To get rid of the isobar problem, molecular ions have to be used. In our case ratios below 10^{-13} are to be measured: We used Calcium hydride (CaH_2) which offers the advantage of sufficient ion currents (CaH_3^-) while potassium hydride does form negative ions such as KH_3^- only at an extremely low ratio [6]. Its drawback is the strong hygroscopic character of CaH_2 which requires sample handling under argon atmosphere and a costly sample preparation.

The enamel fraction is isolated from the rest of the tooth material and converted to CaC_2O_4 via standard oxalate chemistry. The subsequent steps are conversion to CaO, reduction to metallic calcium [7] and hydration [8]. Combining the high energies available with our MP tandem, a time of flight system, the gas filled magnet system (GAMS) [9] and a multianode ionization chamber meets the required high sensitivity for the Ca measurements.

Results obtained on a few teeth samples from unexposed survivors show no ^{41}Ca event, which corresponds to an upper background limit of $5{\cdot}10^{-15}$ for the ^{41}Ca/^{40}Ca isotope ratio. This result indeed confirms our first assumption of a low natural ^{41}Ca/^{40}Ca isotope ratio around Hiroshima. Further investigations of the natural background level and its fluctuations are necessary to quantify the contribution of A–bomb neutrons to the total production of ^{41}Ca.

References

[1] W.C. Roesch (ed.), U.S.-Japan Joint Reassessment of Atomic Bomb Radiation Dosimetry in Hiroshima and Nagasaki, Final Report. Radiation Effects Research Foundation, Hiroshima, 1987
[2] W. Rühm *et al.*, Radiat. Environ. Biophys. **37** (1998) 293.
[3] G. Rugel *et al.*, Nucl. Instr. Meth. **B172** (2000) 934.
[4] N. Nakamura and C. Miyazawa, J. Radiat. Res. **38** (1997) 173.
[5] D. Fink *et al.*, Nucl. Instr. and Meth. **B47** (1990) 79.
[6] G.M. Raisbeck *et al.*, Symp. On Accelerator Mass Spectrometry, eds. W. Henning *et al.*, Argonne National Laboratory, ANL/PHY-81-1, 1981, p. 426
[7] H.J. Maier and W. Kutschera, Nucl. Instr. Meth. **167** (1979) 91.
[8] G. Korschinek and W. Kutschera, Nucl. Instr. Meth. **144** (1977) 343.
[9] K. Knie *et al.*, Nucl. Instr. Meth. **B172** (2000) 717.