^{63}Ni Concentrations in Copper Samples from Japan Relevant for Neutron Dosimetry

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Extensive concentration measurements of radionuclides induced by a-bomb neutrons in samples from Hiroshima and Nagasaki has been used to help to validate the neutron exposures received by a-bomb survivors. The significance of those efforts is emphasized by the fact that the a-bomb survivor data serve as the worldwide basis for safety standards and cancer risk estimates for humans exposed to ionizing radiation (see e.g. [1,2]). The fluence of fast neutrons, which dominate neutron doses in Hiroshima and Nagasaki, can be determined retrospectively by the measurement of the ^{63}Ni ($T_{1/2}$ = 100.1 yr) concentration in copper samples, where it has been produced by fast neutrons (above ≈ 1 MeV) via the reaction ^{63}Cu(n,p)^{63}Ni [3,4]. At distances of greatest epidemiological relevance (between 800 m and 1500 m from the hypocenter), the resulting ^{63}Ni/Ni ratios in copper samples from Hiroshima are expected to be 10^{-12} and below. After enormous efforts we could demonstrate that such small concentrations are measurable by means of accelerator mass spectrometry (AMS) with the GAMS setup at Munich [5,6].

For the determination of the ^{63}Ni content via AMS, the interfering isobar ^{63}Cu, which is about 18 orders of magnitude more abundant than ^{63}Ni in distant samples, has to be removed effectively and nickel has to be quantitatively extracted from the copper matrix. The chemical procedure and the dedicated setup for the AMS measurements in Munich have been described elsewere in detail [5,6,7,8].

Suitable copper samples had to meet the following requirements: a) known location at the time of the bombing, b) line-of-sight with no or minimal intervening shielding, c) located at distances most relevant to the a-bomb survivor health studies, d) low in stable nickel to keep the competing production channel due to the thermal reaction ^{62}Ni(n,γ)^{63}Ni as low as possible. To date, seven copper samples were collected in Hiroshima with distances to the hypocenter ranging from 380 m to more than 5000 m. Each of these samples has been measured by AMS multiple times. The samples from 380 m to 1461 m provide a direct determination of the fast neutron fluences, while samples from 1880 m and 5062 m provide information on ^{63}Ni in copper samples not exposed significantly to a-bomb neutrons. The results of the latter samples approach a constant background level which is in the order of 7×10^4 ^{63}Ni/g Cu [9]. This background level obtained from the distant samples was subtracted for samples with distances less than 1 500 m. Fig. 1 shows the ratio of the measured ^{63}Ni/g Cu values and calculations using the present dosimetry system for Hiroshima (DS86) [10,11]. The only existing data for fast neutrons in Hiroshima before the ^{63}Ni data are the results for ^{32}P concentrations produced via ^{32}S(n,p)^{32}P. These measurements had been done in the weeks after the bombing and these results are shown too. (e.g. [10]). The results for ^{63}Ni are the mean of at least two measurements. To avoid any bias in the measurement, stable nickel content and amount of stable nickel carrier added at LLNL was not disclosed to other measurers until the final AMS result was obtained. The data suggest a somewhat lower fast neutron activation near the hypocenter and good agreement with the calculations applying DS86 [10,11] is observed for larger distances, which are most relevant to the a-bomb survivor data. The constant background beyond about 1800 m appears to originate from a combination of in-situ production of ^{63}Ni due to sample processing, AMS measurement and cosmic rays. The latter contribution is discussed in [12].

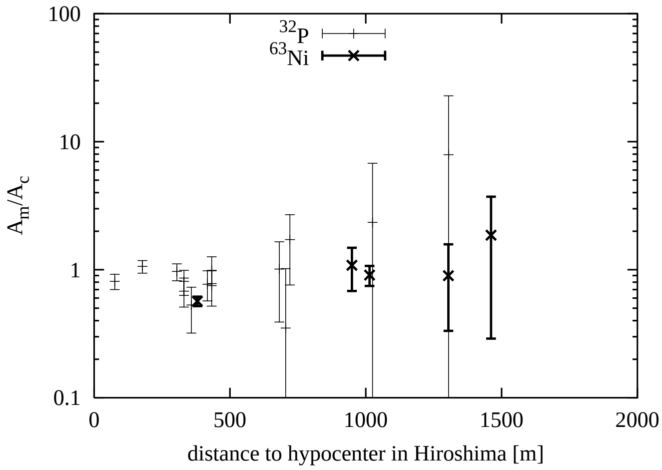

Fig. 1: Ratio of the measurement of ^{63}Ni in copper samples and the ^{32}S data [10] (A_m: result of the measurement) over the calculation of DS86 (A_c: DS86 calculation). The DS86 calculation includes a correction for the specific sample location and shielding situation. Error bars depict the 68% confidence interval.

References

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