Estimation of ^{63}Ni Production in Copper due to Cosmic Rays

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The determination of ^{63}Ni concentration in pure copper samples has been suggested as a means to assess fast neutron fluences in Hiroshima and Nagasaki [1,2]. In those samples, ^{63}Ni ($T_{1/2} = 100.1$ yr) was produced by MeV neutrons originating from the a-bomb explosions, via the reaction ^{63}Cu(n, p)^{63}Ni. Up to now several measurements of ^{63}Ni/Ni ratios in such copper samples from Hiroshima have been performed with the capability of the GAMS at the Munich Tandem Laboratory (see e.g. [3]).

However, at large distances from the hypocenter in Hiroshima a careful discussion of the fraction of ^{63}Ni produced by cosmic radiation is required [4].

A variety of reactions induced by neutrons, protons, muons and photons can contribute to the production of ^{63}Ni in copper (a detailed analysis is given in [5]). It is assumed that copper was exposed to cosmic radiation on the Earth's surface for a sufficiently long time to allow for the ^{63}Ni activity to reach saturation. The main production channels are due to:

i) **Neutrons:** Our estimates are based on the measured neutron spectrum of ref. [6]. Dependences on solar cycle and geomagnetic latitude and altitude are taken into account using two approaches, one by Yokoyama et al. [7] with an altitude correction by Roesler et al. [8] and another from Lal [9], respectively.

ii) **Protons:** High energy protons can contribute to the production of radionuclides via nuclear reactions. To estimate the proton energy spectrum at sea level, the proton/neutron ratio given by Lal and Peters [10] as a function of energy was used and folded with the neutron spectrum given in ref. [6].

iii) **Stopped muons:** High energy protons react with nuclei in the atmosphere and produce e.g. short-lived $\pi^{\pm}$ that decay ($\tau = 2.6 \cdot 10^{-8}$ s) and produce muons. The fraction f_D of negative muons which does not decay ($\tau = 2.2\mu$s), can finally be captured by a nucleus and may react with a proton to form a neutron (probability of capture in a copper nucleus $f_D^{Cu} = 0.928$ [11]).

A rate of stopped negative muons of 190 μ^- per gram copper and year is used [12], for the estimation of the number of ^{63}Ni produced due to stopped muons at sea level. The effective probability $f^*_{^{63}\mathrm{Ni}}$ that ^{63}Ni is produced from copper was not known and therefore determined experimentally. For this purpose, a high-purity copper target was used which had been irradiated previously with low-energy negative muons at the Paul-Scherrer-Institute in Villigen, Switzerland. Details of this experiment are described in [12,13]. The stable nickel content of the sample was determined at LLNL where also the sample was prepared. The number of ^{63}Ni nuclei in the sample ($N_{^{63}\mathrm{Ni}}$) was measured by means of AMS at the GAMS setup (for a description of the measurement procedure see [14]). With this data the effective probability $f^*_{^{63}\mathrm{Ni}}$ for the production of ^{63}Ni after μ^- capture in the copper target was calculated to (12.6±1.6)%.

iv) **Photonuclear reactions:** Photonuclear reactions might also contribute to the production of ^{63}Ni via the reaction ^{65}Cu(γ,np)^{63}Ni. The cross section was taken from [17] and folded with the differential energy spectrum of photons at sea level given in [18].

The results shown in tab. 1 correspond to a copper sample at sea level (37.0° N). In total we estimate as many as 10^4 ^{63}Ni/ g Cu if saturation is reached. For a piece of copper which was exposed to cosmic radiation for about 80 yr, which is typical for samples from Hiroshima, we estimate 4-5·10^3 ^{63}Ni/ g Cu.

reaction	isotope	isotopic abundance [%]	saturation concentration [^{63}Ni/g Cu]
(n,p)	^{63}Cu	69.16	4410
(n,γ)	^{62}Ni	3.59	1.6
(n,2n)	^{64}Ni	0.91	0.0017
(n,α)	^{66}Zn	27.9	0.0009
(n,p2n)	^{65}Cu	30.83	1720
(p,2pn)	^{65}Cu	30.83	164
(μ)	^{63}Cu,^{65}Cu	100	3 200
(photons)	^{65}Cu	30.83	230

Table 1: Saturation concentrations of ^{63}Ni per gram copper, produced due to neutrons, protons, muons and photons from cosmic radiation (for Hampton, 48.1° N geomagnetic latitude, 1999, sea level). For the Ni and Zn impurities typical values of 38 ppm and 5 ppm respectively were used.

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