

## $^{108m}\text{Ag}$ in Neutron Irradiated Silver Coins

By A. Wytttenbach<sup>1</sup>, L. Tobler<sup>1</sup> and H. U. Geiger<sup>2</sup>

<sup>1</sup> Paul Scherrer Institut, 5232 Villigen PSI, Switzerland

<sup>2</sup> Institute of History, University of Berne, 3012 Bern, Switzerland

(Received June 9, 1998; accepted in final form September 30, 1998)

*Silver-108m / Neutron activation analysis / Coins / Residual activity*

### Summary

Six coins, which were irradiated with neutrons 33 years ago, were investigated for their residual activity. Only traces of  $^{108m}\text{Ag}$  could be detected. An estimation of the formation cross section of  $^{108m}\text{Ag}$  is in accord with published values.

### 1. Introduction

The residual activity of samples analysed by instrumental neutron activation analysis (INAA) is usually of little importance, as most samples are discarded after the analysis. An exception is samples that are kept after analysis because of their valuable or unique nature. Archaeological objects, especially coins, belong to this category. INAA of coins was quite popular during the 60s and 70s, as this method is not restricted to the coin surface and is nondestructive in the sense that it does not cause visible changes of the samples. Already in 1972 the number of coins analysed by INAA was estimated to be more than 5000 [1]. If the residual activity was considered at all, it was assumed to be due to  $^{110m}\text{Ag}$  ( $T_{1/2} = 250$  d) because most ancient coins contain Ag, and was therefore assumed to vanish completely within a few years. An experimental verification of this point has never been conducted. We lately came across some coins that were irradiated 33 years ago and determined their present residual activity.

### 2. Experimental and results

The sample analysed in 1964 comprised about 400 coins. These were irradiated in subgroups of 30 coins on a spinning wheel in front of the nuclear reactor SAPHIR. All coins were exposed to a mean flux density of  $2 \cdot 10^9$  n/cm<sup>2</sup>s. Subgroups were formed according to the coin weight. Irradiation time of the subgroups varied between 40 min for the heaviest and 240 min for the lightest coins, in order to keep activities as small as possible [2, 3]. Analysis for the major components Cu and Ag, and for traces of Au, was done by measuring  $^{64}\text{Cu}$ ,  $^{110m}\text{Ag}$ , and  $^{198}\text{Au}$ ; results were reported in [4].

Six of these coins were recovered in 1997 and measured 90 h each with a HPGe-detector with known counting efficiency. The only  $\gamma$ -lines not belonging to the detector background were due to  $^{108m}\text{Ag}$ : 433.7, 614.4 and 722.9 keV, each with 90% abundance [5]. Sample activities calculated from the three lines varied in a nonsystematic way by 2.8% (with 12 degrees of freedom); this variation is completely explained by counting statistics. Mean  $^{108m}\text{Ag}$  activities are given in Table 1, column 4.

### 3. Discussion

The theoretical consideration of the  $^{108m}\text{Ag}$  residual activity in silver coins was for a long time impossible due to the lack of necessary parameters. It was later discouraged by discrepancies in published values. The half-life of  $^{108m}\text{Ag}$  has experienced a dramatic increase with time, being  $\geq 5$  y in 1960 [6] with no indication

Table 1. Activity of  $^{108m}\text{Ag}$  in coins<sup>a</sup>

Sample number	Coin weight g (1)	Silver conc. % (2)	Silver weight g (3)	Activity of the coins		Specific silver activity Bq/g Ag (6)
				Bq (4)	Bq/g (5)	
1	0.894	37.5	0.335	1.309	1.464	3.893
2	0.916	29.5	0.270	1.004	1.096	3.715
3	0.985	35.0	0.345	1.152	1.170	3.343
4	1.812	64.0	1.160	0.939	0.518	0.809
5	2.199	44.2	0.972	0.760	0.346	0.782
6	2.265	48.0	1.087	0.811	0.358	0.746

<sup>a</sup> Coins are ordered according to their weight. They contain only Ag and Cu, except ~1% of unidentified constituents. Activities have a measuring error (reproducibility) of 2%.

how this value was obtained,  $(127 \pm 7)$  y in 1970 [7], and  $(418 \pm 15)$  y in 1992 [8]. A recent compilation adopts a value of  $(433 \pm 15)$  y [9]. The reaction cross section for  $^{107}\text{Ag}(n,\gamma)^{108\text{m}}\text{Ag}$  with thermal neutrons was given as 3 b or 10 b in 1976 [10] and determined later as 0.35 b [11], 0.37 b [12] and 0.48 b [13]. All cross section determinations were done on samples enriched in  $^{107}\text{Ag}$  and assuming  $T_{1/2} = 127$  y, but they are sometimes erroneously combined with  $T_{1/2} = 418$  y [14].

The specific Ag activities in Table 1, column 6 show a decrease with increasing sample weight, as might be expected from the irradiation protocol. The allocation of the individual coins to the subgroups is no longer available. Assuming an irradiation time of 240 min for coin #1 and of 40 min for coin #6 leads to identical reaction cross sections of 1.0 b (with  $T_{1/2} = 433$  y) or of 0.36 b (with  $T_{1/2} = 127$  y), which is in good agreement with the values quoted above.

The activities reported in Table 1 are all well below the exemption limit set by legislation for  $^{108\text{m}}\text{Ag}$  [15], meaning that there are no legal restrictions as to the storage or the exhibition of these coins. Indeed, their activity is roughly equal to the naturally occurring  $^{40}\text{K}$  activity in a glass object of equal weight, for which no objections to display are met.

#### 4. Conclusions

The only residual activity found in Cu-Ag coins 33 years after their irradiation with neutrons is  $^{108\text{m}}\text{Ag}$ . Albeit the activity is very small and below the legal exemption limit, its  $T_{1/2}$  is very long, even on the time scale relevant to museums. It is therefore questionable if INAA of coins is as nondestructive as one might

wish. In any case it seems reasonable to restrict the applied neutron dose to an absolute minimum when investigating coins by INAA.

#### References

1. Meyers, P.: Activation Analysis Methods Applied to Coins, a Review. In: *Methods of Chemical and Metallurgical Investigation of Ancient Coinage* (E. T. Hall, D. M. Metcalf, eds.). Royal Num. Soc. Spec. Publ. No. 8, London (1972).
2. Wyttenbach, A., Hermann, H.: *Archaeometry* **9**, 139 (1966).
3. Wyttenbach, A.: *Helv. Chim. Acta* **49**, 2555 (1966).
4. Geiger, H. U.: Der Beginn der Gold- und Dickmünzenprägung in Bern. Thesis, University of Berne 1968, p. 217.
5. Erdtmann, G., Soyka, W.: *The Gamma Rays of the Radionuclides*, Verlag Chemie, Weinheim (1979), p. 862.
6. Wahlgren, M. A., Meinke, W. H.: *Phys. Rev.* **118**, 181 (1960).
7. Harbottle, G.: *Radiochim. Acta* **13**, 132 (1970).
8. Schötzig, U., Schrader, H., Debertin, K.: Precision Measurements of Radioactive Decay Data. In: *Nuclear Data for Science and Technology* (S. M. Qaim, ed.). Springer, Berlin (1992), p. 562.
9. INDC Report: Activation Cross Sections for the Generation of Long-Lived Radionuclides of Importance in Fusion Reactor Technology (A. B. Pashchenko, ed.). INDC (NDS)-344, IAEA, Vienna 1997.
10. Erdtmann, G.: *Neutron Activation Tables*, Verlag Chemie, Weinheim (1976), p. 146.
11. Rao, D. V., Govelitz, G. F., Mallams, J. T.: *Int. J. Appl. Rad. Isotopes* **29**, 405 (1978).
12. Ryves, T. B.: *Nucl. Sci. Eng.* **72**, 357 (1979).
13. Gavrilas, M., Guinn, V. P.: *J. Radioanal. Nucl. Chem.* **113**, 327 (1987).
14. Pfennig, G., Klewe-Nebenius, H., Seelmann-Eggebert, W.: *Karlsruher Nuklidkarte*, 6<sup>th</sup> edition, Forschungszentrum Karlsruhe 1995.
15. Swiss Federal Council: Radiological Protection Ordinance, 22 June 1994.