DETERMINATION OF TANTALUM IN HIGH-TANTALUM FERRO-ALLOYS

D. GIBBONS and H. SIMPSON

Isotope and Research Division, A.E.R.E., Harwell, U.K.

Une methode est décrite pour la détermination rapide de forte concentration de tantale dans les alliages de fer utilisant l'activation neutronique, suivit par la mesure du rayongamma choisi. L'échantillon est irradié en solution pour éviter les erreurs de blindage contre les neutrons.

The determination of tantalum in ferro-alloys containing niobium is quite difficult, particularly on a routine basis. Since tantalum has a large slow neutron capture cross-section, and the radio-tantalum (¹⁸²Ta) produced on irradiation emits high-energy gamma-rays (1·12, 1·22 MeV), it should be an ideal element for non-destructive analysis by neutron activation and direct gamma-ray spectrometry.

The large slow neutron capture cross-section (22 barns) does, however, introduce neutron self-shielding errors in both samples and standards, and it was expected that some sort of "solid dilution" technique¹ using powdered graphite or sucrose as diluent would be necessary.

SELF-SHIELDING EFFECTS

Two samples were obtained for analysis and were originally supposed to contain about 5 per cent and about 20 per cent of tantalum. To assess the self-shielding effects, the samples (in the form of turnings) were irradiated as received, together with tantalum pentoxide standards. The induced activity at about 1.2 MeV was measured on a gamma spectrometer, the resolution of which was reduced so that the high energy gammas of tantalum-182 combined into a single flat-topped peak. As was expected, high results were obtained due to self-shielding in the tantalum standard. The results for the 5 per cent sample were, however, much higher, relatively, than those for the 20 per cent sample, thus indicating that there was also some self-shielding in the 20 per cent sample.

Attempts were made to eliminate these effects by mixing the samples and standards with powdered graphite or sucrose (previously shown to contain a negligible amount of tantalum), but at best this only partially reduced the self-shielding effects. This was attributed to the comparatively large particle size of the sample and standard; although the particles were dispersed in the graphite, self-shielding still occurred within the individual particles.

This was confirmed by preparing a series of tantalum solutions of increasing concentration (up to 30 mg/ml) and irradiating equal volumes of each. A plot of induced activity against weight of tantalum then gave a typical self-shielding curve with an observed reduction in activity of 30 per cent at 30 mg/ml of tantalum. It is interesting to note that the formula of Keyes² indicates that the self-shielding should be negligible at this dilution. From the graph it was possible to calculate the specific activity which would have been obtained in the absence of self-shielding, and this was shown to be greater than that obtained from a single grain of tantalum pentoxide. When a solution of tantalum, of such dilution that the self-shielding error was less than 1 per cent was used as standard, low results were obtained. The results for the 20 per cent alloy were much less, comparatively, than those for the 5 per cent alloy, thus confirming that self-shielding effects were still operating even in these samples.

Two methods of approach were now available: to prepare standards of the same tantalum content as the samples (either in solution or, preferably, in alloy form); or to dissolve both standards and samples and to dilute the solutions to the same concentration. The latter procedure was in fact chosen. If the samples and standards are both diluted to a concentration less than that giving 1 per cent self-shielding error, the two concentrations need not be matched exactly; but if a preliminary analysis is made to enable the concentrations to be matched fairly closely, much higher concentrations and therefore shorter irradiation times can be used.

The alloy samples and the tantalum metal standard were dissolved in hydrofluoric acid, containing the minimum of nitric acid, in platinum dishes. The solutions were diluted by weight in polythene bottles. A thulium-170 source was used as a static eliminator to facilitate these weighings. The first set of solutions for irradiation were heat-sealed in polythene tubes (pill-packs) but variable results (\pm 10–15 per cent) were obtained. This effect was attributed to incomplete sealing, although usually this was satisfactory, and in all subsequent irradiations silica ampoules were used, giving a reproducibility better than 5 per cent. Since high concentrations of tantalum were involved, reagent blanks were not important, but it was established that the acids used contained negligible amounts of tantalum.

METHOD

Prepare matched solutions of standard and sample in the range of 5-10 mg/ml of tantalum. If solutions cannot be matched, dilute to less than 2 mg/ml of tantalum for less than 1 per cent self-shielding. Irradiate weighed 0.2 ml portions of these solutions in a neutron flux of 10^{12} neutrons cm⁻² sec⁻¹ for 30 min, or proportionately longer for the more dilute solutions or lower fluxes. Allow 24 h for short-lived activity to decay, then determine the tantalum activity on a gamma-ray spectrometer using a scaler connected into a fairly wide channel (about 5 V). If a spectrometer is not available, use a normal gamma-ray counter biased so that it just counts the high energy tantalum activity. Compare the activities of standards and samples, and calculate the results in the usual way.

Results

	Ta present	Ta found
Sample 1	13.18%	$13\cdot1\pm0\cdot3\%$
Sample 2	19.4%	$20.0\pm1.0\%$
Sample 3	5.2%	$5.7\pm0.2\%$
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All errors represent maximum experimental variations.

Sample 1 is a standard alloy with an agreed average value.

Sample 2. The value 19.4 per cent for the tantalum content is the mean of 18.4 per cent (Swedish referee), 18.6 per cent (British referee) and 21.3 per cent (industrial analysis).

Sample 3. The value 5.2 per cent is from an industrial analysis, and no explanation has so far been found for the discrepancies in the results for this sample.

References

¹ J. V. P. Long, Chemical Research Laboratory, Teddington, England. Report No. CRL/AE **60** (1950)

² R. Keyes, United States Atomic Energy Commission. Report No. AECD 3000 (1950)