Resonance Neutrons in the Activation Analysis of Gold*

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Thermal neutrons are eliminated in activation analysis of gold. In the case of analysis of biological material the application of resonance neutrons enables the selective activation. In the case of cadmium telluride the error caused by neutron self-absorption is diminished by this way.

Up to now not enough attention has been given to the possibility of influencing favourably the distinguishing of the determined element already at the activation process itself. Such favourable influence can be brought about by selective activation using resonance maximum of activation cross section.

D. C. Borg and his associates [1—3] used selective activation in nondestructive manganese determination in biological material with very good results. During reactor activation they eliminated thermal neutrons by cadmium absorption, the lower energy resonance neutrons by boron absorption.

The next important aspect of the use of spectrum of resonance neutrons is the neutrons use in high activation cross section for thermal neutrons for material analysed, mainly in the analysis of samples containing cadmium. In such case it is advisable to eliminate thermal neutrons. The contribution of the specific activity of determined elements induced in the sample by thermal neutrons, could not be reproduced at different dimension of samples. This could otherwise become a source of error in the determination of the samples content of the given element.

From research performed in this field to date F. Baumgärtner [4] should be mentioned. This author proved experimentally that the specific activity of chlorine (in its determination in cadmium sulphide) practically equals not only the activity of the sample but also of the standard, assuming the activation is being done in cadmium-covered material (sample and standard).

The authors of this paper have been up to now mostly concerned with the determination of gold using selective activation. The cross section of the reaction of $^{197}Au(n, \gamma)^{198}Au$ shows a high resonance maximum at neutron energy of 4,906 eV.

Experimental

Determination of gold in biological material

Gold determination methods in biological material is of a practical importance in medicine, mainly in the evalution of colloidal gold or gold-containing drugs distribution.

The authors attempted to develope a nondestructive method based on selective acti-

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vation and integral measurement of gamma activity. Selective activation in the nuclear reactor core was performed by elimination of thermal neutrons using a 0.05 cm cadmium foil.

It was shown [5] by calculation that this particular cadmium filter thickness eliminates thermal neutrons with a 99,67 % effectiveness. It was proved experimentally that the ratio of the gold specific activity to the sodium specific activity (as well as other 1/v--absorbers) is six times higher than in the activation of bare samples in identical irradiation positions.

The absorption cross section of gold being relatively high, the question of a possible error caused by neutron flux perturbation inside the gold containing sample has consequently arisen. The perturbation of neutron flux is caused mainly by self-absorption inside the sample. (Detailed analysis of this perturbation has been described in the above cited paper [5].) However, it could be stated that the role of the neutron scattering in the sample and of the neutron flux depression in the vicinity of the sample (which are effective in the presence of thermal neutrons) is in this case negligible, the cadmium ratio at the given filter thickness being 1.005.

Fig. 1 and 2 will describe the relation of the neutron flux perturbation factor F to the samples radius r for human soft tissues and bone tissue. When activating without Cd-foil ($R_{Cd} = 2.450$), the thermal neutrons perturbation factor F_T [5] is not negligible — curves *5*.

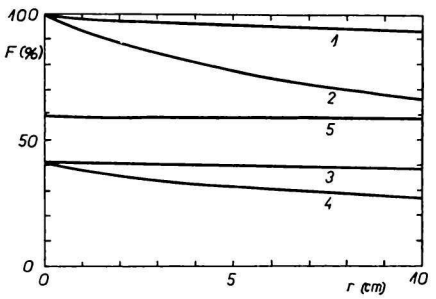


Fig. 1. Dependence of neutron flux perturbation factor F on sample radius r for average human soft tissue.
1. for resonance neutrons ($R_{Cd} = 1.005$, concentration of gold is 0 — 100 ppm.); *2.* for resonance neutrons ($R_{Cd} = 1.005$, concentration of gold is 1000 p m.); *3.* for resonance neutrons ($R_{Cd} = 2.450$, concentration of gold is 0 — 100 ppm.); *4.* for resonance neutrons ($R_{Cd} = 2.450$, concentration of gold is 1000 ppm.); *5.* for thermal neutrons ($R_{Cd} = 2.450$, concentration of gold is 0 — 1000 ppm.). $F = F_R$ in the case of curves *1* — *4*; $F = F_T$ in the case of curve *5*.

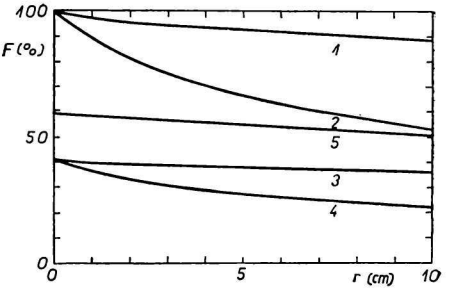


Fig. 2. Dependence of neutron flux perturbation factor F on sample radius r for human bone tissue.
For description of curves see Fig. 1.

The relations used for calculations of resonance neutrons are presented only:

$$F_R = \frac{J_{\text{eff}} + \frac{N_a^R}{N_0^R} 0.44\ \sigma_0}{(J + 0.44\ \sigma_0)\ R_{\text{Cd}}}$$

self-absorption factor for resonance neutrons only

$$J_{\text{eff}} = J\left(1 + \frac{\Gamma_\gamma\, n\sigma_t X_{\text{av}}}{\Gamma}\right)^{-1/2}$$

the effective resonance integral

$$\frac{N_a^R}{N_0^R} = f(X_{\text{av}}, \overline{\Sigma}_a)$$

the ratio of average resonance neutron density inside sample to the average resonance neutron density at the site of the sample prior to its insertion into the reactor (K. M. Case [6])

σ_0 = the absorption cross-section for neutrons a velocity of which is 2200 m/s,

J = the resonance integral,

R_{Cd} = the cadmium ratio,

Γ_γ = partial width for gamma emission,

Γ = total width for gamma and neutron amission,

$X_{\text{av}} = 4V/S$ = the average path of neutrons inside sample (V volume and S surface area of the sample),

$\overline{\Sigma}_a$ = the average macroscopic absorption cross-section for nuclides in the sample,

n = number of absorbing nuclides/cm³ sample,

σ_t = total cross-section in resonance maximum.

Samples of rat blood were prepared for the determination itself. The samples weighed approximately 0.1 g, their gold content varied. Activation was performed under the above described conditions for 20 hours in a nuclear reactor in Řež. The activity of the samples was measured by a scintillation counter at different times following activation.

The half life of the evaluated gold is 2.71 days. It could have been assumed that in integral measurements of activated biological material of the relatively short lived

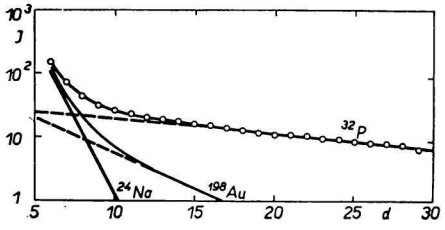


Fig. 3. The decay curve of the activated sample of blood containing 0.05 ppm Au.

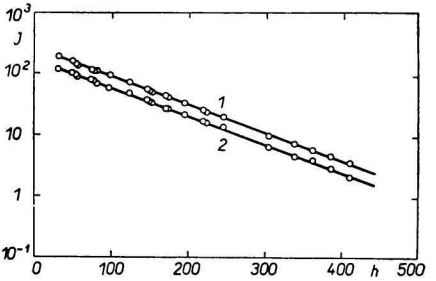


Fig. 4. The decay curve (*1*) of the gold precipitate obtained from activated CdTe (according the given scheme) in comparision with the decay curve (*2*) of standard.

radionuclides, radiosodium will play the most important role and of the long lived radionuclides, radiophosphorus.

The radiophosphorus (pure beta emitter) desintegration rate was reduced by copper filtres of 2640 mg/cm³ thickness. The individual decay curves and their decomposition will be described now.

The radiogold activity in the sample containing 5 ppm Au is predominant.

At 0.5 ppm concentration the decomposition of the decay curve enables the evaluation of radiogold, radiosodium and radiophosphorus content in the sample.

At 0.05 ppm concentration (Fig. 3) the gold content in the sample can still be evaluated by the analysis of the decay curve.

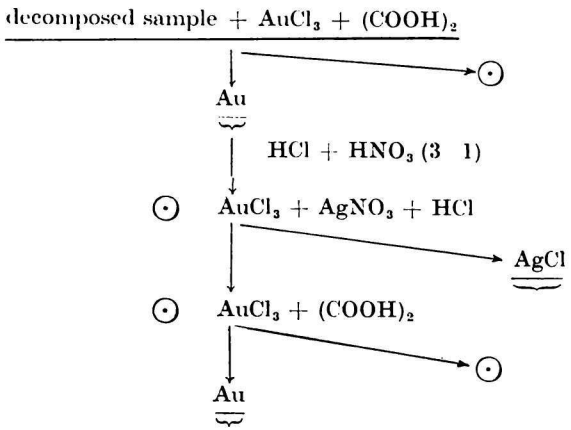
The many kinds of biological materials contain several times less sodium than blood. Thus it can be assumed that the limit concentration for biological materials in the above described method is 0.01 — 0.05 ppm. Preliminary experiments show that with the use of gamma spectrometer it will be probably possible to determine the gold content in biological material at a concentration one order lower than the above.

Determination of gold in cadmium telluride

The authors have been asked by the Mathematics and Physics Faculty of Charles University in Prague, to develope a method for gold determination in cadmium telluride by activation analysis. (The research department of this Faculty is working on a study of gold diffusion in cadmium telluride.)

The aspect of self-absorption of thermal neutrons in the sample renders the problem quite interesting. In the activation of a bare sample the error caused by self-absorption of thermal neutrons according to the authors calculations would be 17.98 % (under the assumption that the sample has a radius of 0.155 cm and weighs 0.1 g). By elimination of thermal neutrons using a 0.05 cm Cd-filter this error is reduced to 0.15 % only and can be corrected by calculation. The presented results have been reached by calculation of neutron flux perturbation the same as in gold determination in biological material.

Because a very complex mixture of radionuclides developes during cadmium and



Scheme 1.

tellur activation in nuclear reactor, it was necessary to perform a chemical separation and prove radiochemical purity of the specimen, on the basis of the decay curve.

In the first experiment following decomposition and addition of a carrier, gold was precipitated by oxalic acid. It was obvious that the chemical separation was insufficient.

The result of the second experiment showed that by reprecipitation of the resulting precipitate a product is obtained, the activity of which falls with the half life of about 7 days. This half life is responsible to radiosilver ^{106}Ag which arises from cadmium by (n, p) reaction (as has been proved by chemical experiment).

In the last experiment the separation of radiosilver in chloride form was included to prevent the coprecipitation of silver in the final precipitate. Fig. 4 shows, that by this way practically absolutely pure radiogold is obtained. The decay curve (sample) corresponds in this case to the natural gold content in cadmium telluride. Gold was determined through comparison with the desintegration rate of the standard. The concentration of gold was 8.8 ppm.

Conclusion

The gold content of biological material with concentration limit of 0.05 ppm can be determined by use of selective activation, integral measurements and decomposition of the decay curve.

In cadmium telluride the gold content can be evaluated by a method based on the activation in the nuclear reactor core inside a cadmium foil, on a chemical separation consisting of three steps (metal gold precipitation, silver chloride precipitation, additional metal gold precipitation) and further based on integral measurements of gamma activity using a scintillation counter.

RESONANČNÍ NEUTRONY V AKTIVAČNÍ ANALÝZE ZLATA

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Při stanovení zlata v biologickém materiálu a v telluridu kademnatém neutronovou aktivační analýzou byly vzorky obaleny kadmiovou folií a vloženy do jádra reaktoru. Kadmiová folie eliminovala vliv tepelných neutronů, takže tvorba radiozlata ^{198}Au probíhala pouze následkem reakce s resonančními neutrony. Tím bylo u biologického materiálu dosaženo selektivní aktivace a u telluridu kademnatého byla vyloučena chyba způsobená samoabsorpcí tepelných neutronů. Jako vzorků biologického materiálu bylo užito krve s přídavkem zlata o různé koncentraci. 0,05 ppm zlata lze snadno stanovit nedestruktivně na základě integrálního měření aktivity gama aktivovaného vzorku a rozkladu rozpadové křivky. V případě telluridu kademnatého bylo třeba provést chemickou separaci. Kromě dvojnásobné redukce kovového zlata bylo třeba zařadit separační stupeň k odstranění radiostříbra vysrážením ve formě chloridu. Ve vzorku telluridu kademnatého bylo nalezeno 8,8 ppm zlata.

РЕЗОНАНСНЫЕ НЕЙТРОНЫ В АКТИВАЦИОННОМ АНАЛИЗЕ ЗОЛОТА

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При определении золота в биологическом материале и в теллуриде кадмия нейтронным активационным анализом образцы были завернуты в кадмиевую фольгу и помещены в ядро реактора. Поскольку кадмиевая фольга устранила влияние термических нейтронов, образование радиозолота ^{198}Au происходило только вследствие взаимодействия с резонансными нейтронами. Таким путем было достигнуто в биологическом материале селективное активирование, а для теллурида кадмия устранилась ошибка, вызываемая самопоглошением термических нейтронов. В качестве образца биологического материала использовали кровь, в которую прибавлялось золото в различных концентрациях. 0,05 *ppm* золота можно легко определить без разрушения образца с помощью интегрального измерения гамма-активности активированного образца и разложением кривой распада. Для теллурида кадмия необходимо произвести химическое разделение. Кроме двухкратного восстановления металлического золота, необходимо было включить разделительную операцию для устранения радиосеребра осаждением в виде хлорида. В образце теллурида кадмия нашли 8,8 *ppm* золота.

Перевела Т. Диллингерова

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