Molecular Activation Analysis for Methylmercury Compounds

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Summary The use of neutron activation analysis for the selective determination of mercury in various mercury compounds is described as well as the application of this technique to the determination of methylmercury in fish tissue.

NEUTRON activation analysis gives information about the total quantity of selected elements, in most cases independent of the chemical form in which that element finds itself. The distribution of the radioactivity of the target element among its various possible chemical forms is, however, often strongly dependent on the nature of the target molecules. Especially among organometallic compounds it is observed[1] that the radioactivity can be recovered in a small number of identifiable compounds, with the target compound usually prominent among them. The fundamental principle to be applied to molecular activation analysis is that the observed pattern of radiochemical yields can give information about the identity of the target compound and, in favourable cases, can lead to quantitative estimation.

We have recently begun to study the application of this principle to distinguish between dimethylmercury, monomethylmercury, and ionic mercury targets. Previous work on alkylmercury compounds had been done by Heitz[2] and by Wheeler and McClin.[3] In the present study, samples of monomethylmercury chloride and dimethylmercury, sealed in quartz vials, were irradiated at 30° with a neutron flux of $1{\cdot}0 \times 10^{12}$ neutrons cm^{-2}.† Each sample was dissolved in a petroleum ether-chloroform solution of the other compounds (which acts as chemical carrier for any radioactive molecules formed) and separated by chromatography on silica gel column. Separate experiments showed no exchange between the mercury compounds during the time of the experiments (see also ref. 2), and no cross contamination in the separation.

Neutron-irradiated dimethylmercury gave a 13·4% yield of labelled dimethylmercury and no monomethylmercury, while irradiation of methylmercury chloride gave a 64·4% yield of the labelled methylmercury and no significant amount of dimethylmercury. From these data it is evident that an unambiguous identification of methyl- or dimethyl-mercury as target compound is possible.

In applying this principle to methylmercury compounds in other matrices, we have found that the values of the fundamental radiochemical yield ('retention') are not the same as in the pure compounds. We have measured this retention on two samples of freeze-dried fish protein concentrate doped with known small quantities of methylmercury. Results of several measurements indicate a marked difference between the two samples (3·1% for a codfish sample and 5·2% for swordfish). It is not yet known whether this difference in the retentions is characteristic of the species of the fish, or is rather dependent on its age or condition. These values are in reasonably good agreement with the results from gas chromatographic analysis[4] of the same samples. We anticipate that this principle will also apply to any element which forms non-exchanging molecular compounds and which can be activated by neutrons to give radionuclides with a satisfactory half-life.

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