Preparation of High Specific Induced Radioactivity by Neutron Bombardment of Metal Chelate Compounds

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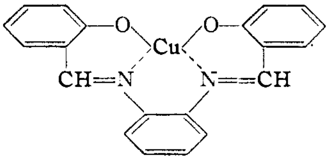
The Szilard–Chalmers method[1a] has been used for some time in the production of radioactive tracers of high specific activity in those cases where the active isotope is made by an η,γ reaction. The principal difficulty in many cases is to find a compound for the neutron bombardment such that there is no thermal exchange between the newly formed radioactive atoms and the inactive atoms in the original material and such that the chemical separation of the two is not too difficult.

Recently, two papers have appeared in which the use of metal chelated organic compounds for this purpose was reported. Drehman[2] attempted to separate Mn^{56} from neutron irradiated manganese acetylacetonate and manganese benzoylacetonate but failed because of the rapid thermal exchange. Starke[3] isolated U^{239} from neutron bombarded uranyl benzoylacetonate, but had some difficulties in the chemical separation because of thermal exchange.

In the course of some other work, we have found some metallic compounds similar to those mentioned above, but which show no measurable rate of exchange at room temperature.[4] We have used one of these compounds, copper salicylaldehyde-*o*-phenylenediimine to concentrate the 12.8-hour activity of copper, Cu^{64}. The formula for this compound is



For the production of Cu^{64} this process has no practical value since very high specific activities can be produced by deuteron bombardment of copper or by an n,p reaction on Zn^{64}. However, a great many metals form chelate compounds of the type in question[5] and this method should be applicable to the preparation of other activities. Some preliminary work has shown that the separation is quite successful for the corresponding uranyl compound.

Experimental

Preparation of Materials.—Copper salicylaldehyde-*o*-phenylenediimine was prepared by Pfeiffer's method[6] and its composition checked by analysis.

Pyridine.—J. T. Baker reagent grade pyridine was dried over potash and distilled through a Widmer column (b. p. 115.2–115.4°).

Neutron Bombardments.—All bombardments were done on the 60-inch cyclotron at Berkeley. The neutrons were produced by the action of 13.7 Mev. deuterons on beryllium. Each radioactive sample was followed on an electroscope to check its decay. Through a time equal to four half-lives, no measurable departure from the 12.8-hour half-life was found.

Szilard–Chalmers Experiments.—The data obtained are summarized in Table I. The specific activity obtained in experiment 3, where we got the best separation, was about a microcurie per milligram of copper.

Table I

Exp. no.	Material bombarded	Separation procedure	Activity[a] of Chelate compound	Activity[a] of Inorganic copper	Activity per g. of Cu Chelate compound	Activity per g. of Cu Inorganic copper
1	0.5 g. solid CuSal op.[b]	Chelate compound washed with water, 5 mg. CuAc$_2$ added to water and CuS precipitated	0.192	0.005	1.86	4.135
2	0.5 g. solid CuSal op.	Chelate compound dissolved in pyridine, 5 mg. CuAc$_2$-1Py[c] added as carrier. Chelate pptd. by dilution to ten volumes with 15% acetic acid. CuAc$_2$ pptd. as CuS	.239	.263	2.32	215
3	100 ml. of 0.0152 *M* pyridine soln. of CuSal op.	Five mg. of CuAc$_3$-1Py added to solution as carrier and solution treated as in exp. 2	.095	.443	0.99	366
4	100 ml. of pyridine soln. 0.0152 *M* in CuSal op and 0.0152 *M* in CuAc$_2$·1Py	Solution treated as in expt. 3 except that no carrier was added	.024	.721	0.25	7.5

[a] Activities are expressed in electroscope divisions per second. [b] CuSal op. = copper salicylaldehyde-*o*-phenylenediimine. [c] CuAc$_2$-1Py = copper acetate with 1 pyridine of crystallization.

A solvent extraction scheme could be worked out instead of the precipitation separation which we used. The addition of carrier would then be omitted and much higher specific activities obtained.

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