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Authors

Kratz, J.V.

Liljenzin, J.O.

Seaborg, G.T.

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A CHEMICAL GROUP SEPARATION PROCEDURE FOR SUPERHEAVY ELEMENTS AND VARIOUS
OTHER REACTION PRODUCTS FROM HEAVY-ION BOMBARDED URANIUM TARGETS*

J. V. Kratz[†], J. O. Liljenzin^{††} and G. T. Seaborg

Lawrence Berkeley Laboratory
and Department of Chemistry
University of California
Berkeley, California 94720

INTRODUCTION

Attempts to synthesize superheavy elements through heavy-ion acceleration have been started in several laboratories. Among the various techniques for an identification of superheavy elements (with half-lives longer than a few seconds) chemical separations are likely to be the most sensitive approach because (i) chemical separations can be performed quantitatively, (ii) they can take advantage of the increased yields of reaction products in thick targets and (iii) thick targets have markedly better heat conduction properties than thin foils and can thus be bombarded with the maximum particle currents available at present.

Because it is an open question which, (if any), of the superheavy elements will be formed in heavy-ion reactions, the first step in a chemical search for these elements should include a separation characterized by high decontamination from actinides and, at the same time, by a high general sensitivity for all superheavy elements around element 114. Such group separations have to take advantage of common features in the chemistry of these elements, where individual differences among these elements are of little importance.

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[†]On leave from Institut für Kernchemie, Universität Mainz, with a fellowship from Gesellschaft für Schwerionenforschung GSI, Darmstadt, Germany.

^{††}Department of Nuclear Chemistry, Chalmers University of Technology, Göteborg, Sweden.

Predictions concerning the chemical properties of superheavy elements (1-4) have been evaluated with respect to these requirements (5). It appears that complex formation, preferentially with heavy halide ions, provides a convenient means to isolate the superheavy element-group from actinide elements and the target material. Different chromatographic separation techniques have been tested to separate the superheavy element complex anions from the cationic actinides and uranium in dilute HBr/Br₂ acid systems (5), and it was concluded from these studies that a simple cation exchange separation is the most suitable method. This separation is presently used to process uranium targets bombarded at the Berkeley SuperHILAC. In addition to the search for superheavy elements, the same targets are used to obtain information basic to an understanding of heavy-ion reactions by measuring cross sections of various other reaction products. This is done by adding further chemical separation steps to the elution of a superheavy element fraction followed by α -particle and γ -ray spectroscopy.

EXPERIMENTAL

The cation exchange resin used is AG 50W \times 8 (Bio-Rad) in the hydrogen form with a particle size of 230 - 400 mesh. Heavy-walled glass tubing, 3 mm i.d., is used for the columns. The columns are fitted at the top with a 5 cm long reservoir with a standard taper 18/9 ball joint. Quartz wool plugs are used as bed supports. The columns are prepared by slurry packing in 2 M HCl and compacted by applying 15 lb/in² pressure from a N₂ tank. To process \sim 30 mg uranium targets, 1 ml of resin is used; part of the resin then extends up into the lower part of the column reservoir (i.d. 8 mm). This part of the resin is used to absorb the large amount of target material; actually 1/7 of the capacity of the column is used by the uranium. Columns with such (stepwise) decreasing inner diameter are known to offer certain advantages in the chromatographic separation of small quantities from bulk materials (6). After packing, the columns are clamped into a thermostated brass block and eluted with several bed

volumes of 0.1 M HBr at 50°C. Mixtures of known amounts of tracer activities are loaded onto the column in 100 µl 0.1 M HBr/Br₂ solution and then eluted with HBr/Br₂ and HCl solutions of increasing molarity. The mobile phase velocities (typical 1 drop per 6 seconds) are adjusted by applying N₂ pressure to the top of the column reservoir. The effluent volume is controlled by an electronic drop counter. Fractions are collected in plastic cones and measured in constant geometry with a Ge(Li) diode using standard pulse height analysis and, if necessary, half-life corrections. The activities of individual tracers in the eluate fractions in percent of the total activities are evaluated as a function of the effluent volume. To check for complete elution, the column material is also counted for remaining activities. The preliminary elution scheme obtained from the tracer studies was checked by processing several uranium targets previously bombarded with ⁴⁰Ar ions. Results and the final separation scheme are discussed below:

RESULTS

Cation Exchange Group Separation for Superheavy Elements

Figure 1 shows some examples for the elution of homologs of the superheavy elements from a 3x20 mm AG 50Wx8 column at 50°C and an elution rate of one drop per 5 seconds. Os, Ir, Pt, Au, Hg, Tl, Bi and Po are eluted in 4 column volumes

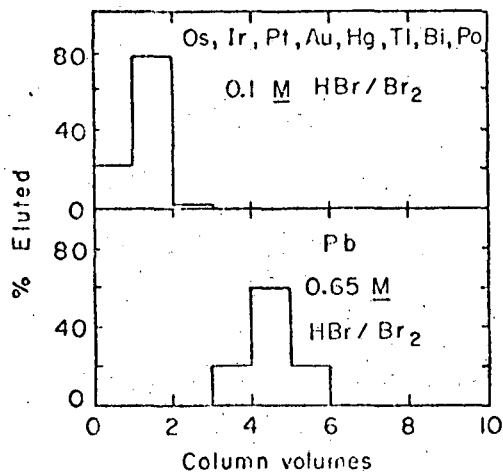


FIG. 1

Elution of homologs of the SHEs from a 3x20 mm AG 50Wx8 column. Elution rate 1 drop/5 sec. 50°C

of 0.1 M HBr/Br₂ whereas UO₂²⁺ and Eu³⁺ are strongly absorbed on the resin. To elute Pb(II), 6 column volumes of 0.65 M HBr/Br₂ are necessary. It is not possible to start the elution immediately with 0.65 M HBr/Br₂ because the elution of Au(III) would then be markedly delayed. Under the described conditions the elution of Os through Po is quantitative. The method seems to be suitable for a fast automated group separation of superheavy elements; by using very small columns the superheavy element

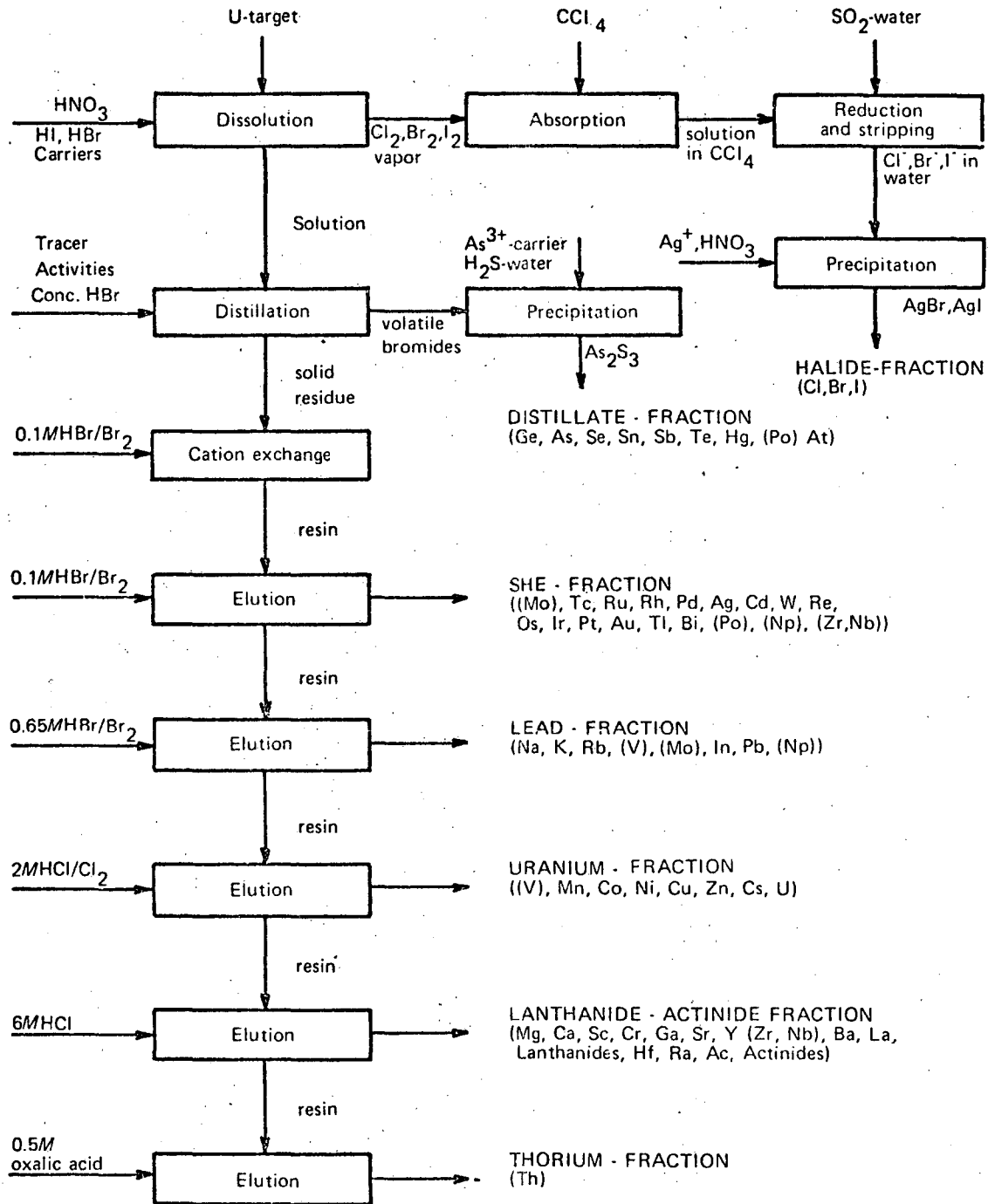
fractions could be eluted within eluate volumes of the order of 5 drops. Together with a fast transportation system for recoil atoms from a thin target separation times of 50 to 60 seconds are certainly achievable.

Processing of Heavy-Ion Bombarded Uranium Targets

A flow diagram describing the chemical processing of heavy-ion bombarded uranium targets is given in Fig. 2. Thick natural uranium foils (~ 30 mg) are bombarded in a water-cooled target holder. Dissolution of the target takes place in a closed glass apparatus in conc. HNO_3 . After addition of 35 μl each of 1 M HBr and HI carriers, bromine and iodine vapors are distilled into a CCl_4 trap. After backextraction into SO_2 -water, AgBr and AgI are precipitated with 1 M $\text{HNO}_3/\text{AgNO}_3$, filtered onto a membrane filter (15 mm diameter) and mounted for γ -ray counting. Chemical yields for Br and I (typically 95% and 70%, respectively) are later determined by activation analysis.

The nitric acid in the dissolver is destroyed by fuming with conc. HBr and evaporating to dryness three times. Volatile bromides are collected together with the distillate in an empty glass trap. After the addition of 0.1 mg of As^{3+} carrier to the distillate, a sulfide precipitate from this solution is filtered onto a membrane filter. The latter is mounted uncovered, dried, and counted for α -particle, spontaneous fission and γ -ray activities. Chemical yields were found to be 60 - 80% for Ge, As, Se, Sn, Sb and Hg. These yields are determined by adding known amounts of tracer activities to the solution prior to the bromide distillation.

The solid residue in the dissolver is then dissolved in 0.1 M HBr/ Br_2 and transferred to the preconditioned cation exchange column. The eluate fractions are collected in 10 ml glass beakers and condensed nearly to dryness. These solutions together with successive portions of HBr washing solution are then transferred onto 15 mm circular microscope cover glass discs and evaporated to dryness. The glass discs are mounted for α -particle, spontaneous fission and γ -ray counting. The samples are acceptably free of residue as evidenced



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FIG. 2

Flow diagram for the chemical processing of heavy-ion bombarded uranium targets. Elements identified in the chemical fractions are listed below the respective fraction names.

by good resolution in the α -particle spectra. The following eluate fractions are eluted from the column (see Fig. 2):

Superheavy element fraction	in 4 ml 0.1 <u>M</u> HBr/Br ₂
Lead fraction	in 6 ml 0.65 <u>M</u> HBr/Br ₂
Uranium fraction	in 4 ml 2 <u>M</u> HBr/Br ₂
Lanthanide-Actinide fraction	in 9 ml 6 <u>M</u> HCl
Thorium fraction	in 2 ml 0.5 <u>M</u> oxalic acid.

Superheavy element fraction, lead fraction, and distillate are expected to contain the superheavy elements. Except for the halide fraction and the distillate, where chemical yield determinations have to be carried out, the separations are quantitative. In Fig. 2 elements identified in a certain fraction are listed below the respective fraction names. Complications arise for a few elements, e.g. Mo, Po, Np, V, Zr, Nb, are distributed over two or more fractions. For isotopes of these elements the initial activities from different fractions are summed to obtain the total cross sections. A number of elements are not listed in Fig. 2. Isotopes of these elements have not been detected in the separations performed to date because of unfavorable half-lives, low quantum yields or low formation cross sections.

This separation scheme has been used extensively during the past year in the search for superheavy elements at LBL. The fractions relevant for the superheavy element search are ready for counting within 40 - 50 minutes after the end of bombardment. The complete separation has been carried out by two chemists within 150 minutes. Through use of this separation scheme combined with γ -ray spectroscopy, yields of more than 100 isotopes distributed among 49 elements have been determined in one experiment.

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