

## Synthesis and Chemical Characterization of the Transactinides

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The synthesis and characterization of elements with atomic numbers greater than 103 has been an active area of study since 1964. Theories describing an "island of stability" for elements with around 114 protons and 184 neutrons were first proposed in the 1960's. Such proton and neutron configurations are predicted to be particularly stable due to the filling of the proton and neutron shells in a fashion analogous to the filling of electron shells in the noble gases [1-3]. As of yet these superheavy elements have not been synthesized, and the island of stability has not been reached. However, the periodic table has been extended through element 110 [1, 2].

Recently there has been much interest in the chemistry of the heaviest elements. At very high atomic numbers, relativistic effects can influence the electronic structure of the atoms, and can affect the energies of the valence electrons [4]. For example, element 104 may actually have a ground state of  $7s^2 7p_{1/2}^2$  rather than the ground state  $6d^2 7s^2$  expected by extrapolating from Hf. Since the binding energies and radii of the electronic orbitals play an important part in the chemistry of an element, it is clear that these relativistic effects will affect the chemistry of the heavier elements [5].

There are a few obstacles that must be overcome in order for the chemistry of these elements to be explored. Production of these elements requires both beams of charged particles at suitable energies and, often, radioactive actinide targets [6]. Even with the optimal parameters, the production cross section is often small: on the order of  $10^{-34}$  cm<sup>2</sup> [7]. Also, even if the target is free of impurities, the nuclear reaction does not give a single product. For example,  $^{249}\text{Bk} + ^{18}\text{O} \rightarrow ^{262}105 + 5n$  also yields Md and Fm isotopes. Often small lead impurities exist in the target material, leading to the formation of isotopes of Bi and Po [4, 8-14].

The most specific means of detecting and identifying an element is through the use of the  $\alpha$ -activities of the isotope and its daughters [13]. However, sometimes the impurities can mask the  $\alpha$ -activities of the isotope being explored. For example, in the search for the isotope  $^{263}105$ , the  $\alpha$ -activities of polonium isotopes masked the  $\alpha$ -activities of the  $^{263}105$  isotope [14] so that identification of the latter among the nuclear reaction products is extremely difficult.

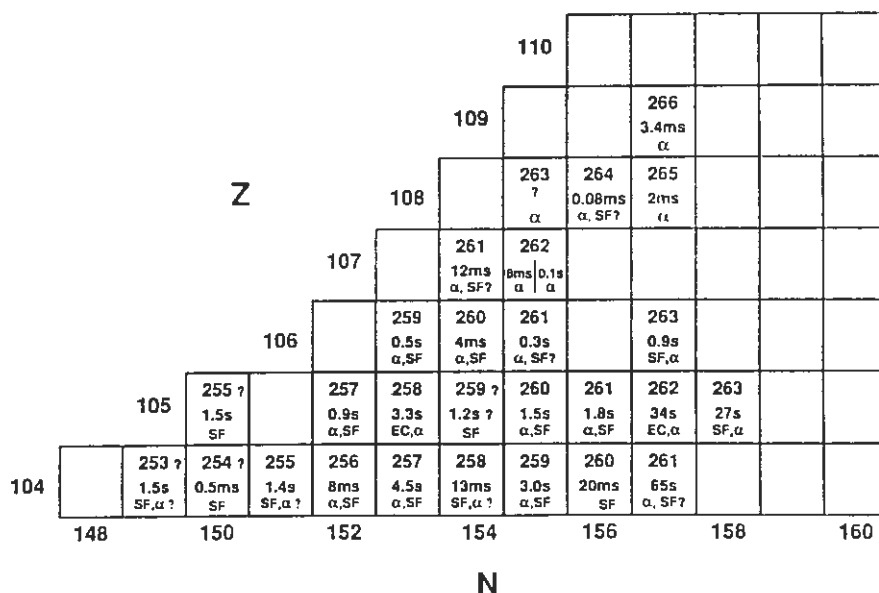


Fig. 1. Isotopes of transactinide elements.

The half-lives of the isotopes are also rather short. The longest known isotope of element 105 has a half-life of only 34 seconds [6]. To overcome the short half-lives, several rapid assay techniques have been developed; among these are the OLGA II on-line gas chemistry apparatus [15], the automated chromatographic chemical element separation system (ACCESS), the automated injection system (AIS) [16], and the heavy element volatility instrument (HEVI) [17].

Despite these obstacles, exploration of the chemistry of elements 104 and 105 has been done. An early study of element 104 by Zvara *et al.* utilized frontal gas thermochromatography [18]. His results led him to conclude that the chemistry of 104 resembles that of its lighter homologs. Later extraction experiments also confirm the group 4, or pseudo-group 4, behavior of element 104 [6]. Recent gas phase chromatographic experiments by Turler *et al.* [10] have recently found that the volatility of the bromides of 104 are higher than those of its homologs.

Zvara also utilized frontal gas thermochromatography [7,18] to study isotope  $^{261}\text{105}$ . Zvara interpreted the data to prove that 105 follows the trend for the group 5 elements and has less volatile bromides than its homologs. Turler *et al.* [10] also performed gas chromatography tests on the bromides of element 105. It was found that, contrary to the result found for element 104, bromides of element 105 are less volatile than the lighter group 5 homologs. Gäggeler *et al.* [4] calculated a sublimation enthalpy for  $\text{105Br}_5$  of about 140 kJ/mol. In more recent years, the aqueous chemistry of element 105 has been explored [8,9,11-13]. Comparison of the sorption ability and extraction ability of element 105 with both its lighter homologs and the group 4 elements shows that 105 does belong to the group 5 family. Some other studies found deviations from the chemistry expected from the position of 105 in the periodic table [8,13].

## References

1. Seaborg, G. T.; Loveland, W. D., *The Elements Beyond Uranium*; Wiley & Sons, Inc: New York, 1990, pp. 51-61.
2. Ghiorso, A.; Münzenberg, G.; Oganessian, Y. T.; Loveland, W., in *Conference on Chemical Research: XXXIV. Fifty Years With Transuranium Elements*; The Robert A. Welch Foundation: Houston, 1990, pp. 51-217.
3. Kaplan, I., *Nuclear Physics*; Addison-Wesley Publishing Co.: Reading, 1962, pp. 209-218.
4. Gäggeler, H. W.; Jost, D. T.; Kovacs, J.; Scherer, U. W.; Weber, A.; Vermeulen, D.; Turler, A.; Gregorich, K. E.; Henderson, R. A.; Czerwinski, K. R.; Kadkhodayan, B.; Lee, D. M.; Nurmia, M.; Hoffman, D.C.; Kratz, J. V.; Goyer, M. K.; Zimmerman, H. P.; Schädel, M.; Bröchle, W.; Schimpf, E.; Zvara, I., "Gas Phase Chromatography Experiments with Bromides of Tantalum and Element 105," *Radiochim. Acta* **1992**, *57*, 93-100.
5. Keller, O. L. Jr., "Chemistry of the Heavy Actinides and Light Transactinides," *Radiochim. Acta* **1984**, *37*, 169-180.
6. Hoffman, D. C., "Atom-at-a-Time Chemistry," *Radiochim. Acta* **1993**, *61*, 123-128.

7. Zvara, I.; Belov, V. Z.; Domanov, V. P.; Shalaevskii, M. R., "Chemical Isolation of Nilsbohrium as Ekatanalium in the Form of the Anhydrous Bromide: II. Experiments with a Spontaneously Fissioning Isotope of Nilsbohrium," *Sov. Radiochem.* **1976**, *18*, 328-334.
8. Kratz, J. V.; Zimmerman, H. P.; Scherer, U. W.; Schädel, M.; Brüchle, W.; Gregorich, K. E.; Gannett, C. M.; Hall, H. L.; Henderson, R. A.; Lee, D. M.; Leyba, J. D.; Nurmia, M. J.; Hoffman, D.C.; Gäggeler, H. W.; Jost, D. T.; Baltensperger, U.; Nai-Qi, Y.; Türler, A.; Lienert, C., "Chemical Properties of Element 105 in Aqueous Solution: Halide Complex Formation and Anion Exchange into Triisocetyl Amine," *Radiochim. Acta* **1989**, *48*, 121-133.
9. Schädel, M.; Brüchle, W.; Schimpf, E.; Zimmerman, H. P.; Guber, M. K.; Kratz, J. V.; Trautmann, N.; Gäggeler, H. W.; Jost, D. T.; Kovacs, J.; Scherer, U. W.; Weber, A.; Gregorich, K. E.; Türler, A.; Czerwinski, K. R.; Hannink, N. J.; Kadkhodayan, B.; Lee, D. M.; Nurmia, M. J.; Hoffman, D.C., "Chemical Properties of Element 105 in Aqueous Solution: Cation Exchange Separations with  $\alpha$ -Hydroxyisobutyric Acid," *Radiochim. Acta* **1992**, *57*, 85-92.
10. Türler, A.; Gäggeler, H. W.; Gregorich, K. E.; Barth, H.; Brüchle, W.; Czerwinski, K. R.; Guber, M. K.; Hannink, N. J.; Henderson, R. A.; Hoffman, D.C.; Jost, D. T.; Kacher, C. D.; Kadkhodayan, B.; Kratz, J. V.; Kovacs, J.; Kreek, S. A.; Lee, D. M.; Leyba, J. D.; Nurmia, M. J.; Schädel, M.; Scherer, U. W.; Schimpf, E.; Vermeulen, D.; Weber, A.; Zimmerman, H. P.; Zvara, I., "Gas Phase Chromatography of Halides of Elements 104 and 105," *J. Radioanal. Nucl. Chem. (Articles)* **1992**, *160*, 327-339.
11. Guber, M. K.; Kratz, J. V.; Zimmerman, H. P.; Schädel, M.; Brüchle, W.; Schimpf, E.; Gregorich, K. E.; Türler, A.; Hannink, N. J.; Czerwinski, K. R.; Kadkhodayan, B.; Lee, D. M.; Nurmia, M. J.; Hoffman, D.C.; Gäggeler, H. W.; Jost, D. T.; Kovacs, J.; Scherer, U. W.; Weber, A., "Chemical Properties of Element 105 in Aqueous Solution: Extractions into Diisobutylcarbinol," *Radiochim. Acta* **1992**, *57*, 77-84.
12. Zimmerman, H. P.; Guber, M. K.; Kratz, J. V.; Schädel, M.; Brüchle, W.; Schimpf, E.; Gregorich, K. E.; Türler, A.; Czerwinski, K. R.; Hannink, N. J.; Kadkhodayan, B.; Lee, D. M.; Nurmia, M. J.; Hoffman, D.C.; Gäggeler, H. W.; Jost, D. T.; Kovacs, J.; Scherer, U. W.; Weber, A., "Chemical Properties of Element 105 in Aqueous Solution: Back Extraction from Triisocetyl Amine into 0.5 M HCl," *Radiochim. Acta* **1993**, *60*, 11-16.
13. Gregorich, K. E.; Henderson, R. A.; Lee, D. M.; Nurmia, M. J.; Chasteler, R. M.; Hall, H. L.; Bennet, D. A.; Gannet, C. M.; Chadwick, R. B.; Leyba, J. D.; Hoffman, D.C., "Aqueous Chemistry of Element 105," *Radiochim. Acta* **1988**, *43*, 223-231.
14. Kratz, J. V.; Guber, M. K.; Zimmerman, H. P.; Schädel, M.; Brüchle, W.; Schimpf, E.; Gregorich, K. E.; Türler, A.; Hannink, N. J.; Czerwinski, K. R.; Kadkhodayan, B.; Lee, D. M.; Nurmia, M. J.; Hoffman, D.C.; Gäggeler, H. W.; Jost, D. T.; Kovacs, J.; Scherer, U. W.; Weber, A., "New Nuclide  $^{263}\text{Ha}$ ," *Phys. Rev. C* **1992**, *45*, 1064-1069.
15. Gäggeler, H. W.; Jost, D. T.; Baltensperger, U.; Weber, A.; Kovacs, J.; Vermeulen, D.; Türler, A., "OLGA II, an On-line Gas Chemistry Apparatus for Applications in Heavy Element Research," *Nucl. Instr. Meth. A* **1991**, *309*, 201-208.

16. Kreek, S. A.; Nurmia, M. J.; Kadkhodayan, B.; Gregorich, K. E.; Lee, D. M.; Hoffman, D. C.; Hall, H. L.; Palmer, C. E. A.; Baisden, P. A.; Henderson, R. A., "An Automated, On-line Rapid Chemistry System," *Nucl. Instr. Meth. A* **1992**, *317*, 251-253.
17. Kadkhodayan, B.; Türler, A.; Gregorich, K. E.; Nurmia, M. J.; Lee, D. M.; Hoffman, D.C., "The Heavy Element Volatility Instrument (HEVI)," *Nucl. Instr. Meth. A* **1992**, *317*, 254-261.
18. Zvara, I.; Aikhler, V.; Belov, V. Z.; Zvarova, T. S.; Korotkin, Y. S.; Shalaevskii, M. R.; Shchegolev, V. A.; Yussonnua, M., "Gas Chromatography and Thermochromatography in the Study of Transuranium Elements," *Sov. Radiochem.* **1974**, *16*, 709-715.