Heavy Elements: A Feud over 104 and a Future for 114

Of the several symposia held this year to celebrate the 100th anniversary of the development of the periodic table by Dmitri Mendeleev, the one that has attracted the greatest number of researchers in transuranium studies was sponsored by the Robert A. Welch Foundation and was held in Houston, Texas, from 17 to 19 November. Most of the conference time was spent reviewing recent developments associated with the possible existence of a group of stable elements centered around atomic number 114, but there was also news of new work on element 104.

Scientists from the Lawrence Radiation Laboratory of the University of California announced results of chemical experiments on element 104 and used the occasion to propose a new name. Albert Ghiorso, who reported the work, suggested that the element be called rutherfordium for Lord Rutherford "the great pioneer of nuclear science."

Since the honor of naming new elements is traditionally accorded to the discoverer, the Berkeley proposal is a direct challenge to Soviet claims of discovery. The Soviet work goes back to 1964 when a group under Georgii Flerov, Director of the Nuclear Studies Laboratory at the Joint Institute for Nuclear Research in Dubna, bombarded plutonium-242 with neon-22 and obtained a product to which they assigned atomic number 104 and mass number 260 ± 1. The isotope reportedly decayed by spontaneous fission with a half-life of 0.3 second. They named the element kurchatovium after Igor Kurchatov, the nuclear physicist who led the Soviet atomic energy program during World War II and did much of the pioneering work with their cyclotrons.

Soviet and American Research

The Dubna group conducted physical experiments until the summer of 1966, when they began a series of chemical studies that lasted until the summer of 1968. The chemistry of element 104 is especially interesting because it begins a new series in the periodic table and therefore should display different properties from the 11 actinide elements that have been produced artificially since 1940. The Soviet chemical experiments were designed to show that element 104 formed a volatile chloride that behaved like the chlorides of hafnium and zirconium rather than those of the actinide elements.

The Dubna team has reported that a mixture of chlorides [primarily element 104, nobelium (102), and lawrencium (103)] was formed in the target area and passed through a gas filter that adsorbed the actinide compounds. Fission tracks in mica detectors were cited as evidence that element 104 passed through the filter.

Ivo Zvara, a Czechoslovakian chemist now working at Dubna, reported at the Welch conference that an improved measuring technique resulted in a lowering of the half-life to 0.1 second. The earlier chemical experiments could not have been obtained with atoms having a half-life of 0.1 second, and so there is now some confusion surrounding the work. Zvara said in Houston that an isotope with a longer half-life must have been present, but he presented no physical evidence to support this statement.

The first results by the Berkeley team on element 104 were obtained in July 1968 when they began a series of bombardments of californium-249 with carbon-12 and carbon-13 ions. Results of this and later work were published in *Physical Review Letters* last 16 June. From the carbon-12 bombardment they reported synthesis of element 104^{257}, which decayed by emission of alpha particles to nobelium-253 with a half-life of about 4.5 seconds. From carbon-13 bombardment they reported element 104^{255}, which decayed by alpha particle emission to nobelium-255 with a half-life of about 3 seconds. In both cases the nobelium daughter nuclei that were formed after element 104 recoiled from the target were identified. Tentative identification of element 104^{258} produced by bombardment with a mixture of carbon isotopes was reported, and the team of researchers estimated that it has a half-life of 11 milliseconds for decay by spontaneous fission.

At the meeting of the American Chemical Society held in New York last September, the Berkeley team reported synthesis of element 104^{261} by oxygen-18 bombardment of curium-248. The element decays by alpha particle emission with a half-life of 70 seconds, and the nobelium-257 daughter nuclei produced by this decay were identified.

With the facilities at hand for producing the isotope with long half-life, the Berkeley group began preparing to run chemical experiments about 8 weeks ago. Like the Soviets, they wanted to show that element 104 behaved like hafnium rather than like the actinides. Earlier work had shown that the actinides form complexes in ion exchange columns and are difficult to wash out with an organic eluting agent, and theory predicted that element 104 should wash out easily. In their experiments, atoms recoiling from the curium target were transferred to the ion exchange column and washed with the eluting agent. The eluted material was quickly dried, and its alpha spectrum was measured. In several hundred experiments performed over a period of about 3 weeks, they measured 17 alpha particles that could be attributed to the decay of element 104^{261}.

Burden of Proof

Since the Soviet claim for discovery of element 104 came first, they need only to make their work convincing. The Berkeley group, in addition to making their own claims seem reasonable, should show that the Soviet work before July 1968 was interpreted incorrectly.

Evaluation of experimental conditions—beam intensities, number of experiments, purity of targets, and the like—is a difficult matter, and many researchers outside Berkeley and Dubna prefer to refer to the difficulty of the experiments rather than to attempt to judge them. Likewise, it takes considerable technical expertise to evaluate the merits of the chemical work. However, measurements of spectra of alpha particles and detection of daughter nuclei are less ambiguous than analysis of fission tracks; therefore, on these grounds the Berkeley work is more convincing.

The attempt of the Berkeley team to confirm or disprove the Soviet work preceded their own synthesis. It consisted of a large number of bombardments including einsteinium-253 with boron-10, curium-248 with oxygen-16,

and curium-246 with oxygen-18. In their article last June they reported that in the bombardments they failed to find an isotope with a half-life as short as 0.3 second. After the Soviets lowered their value for the half-life to 0.1 second, the Berkeley group repeated many of the experiments and at the Mendeleev conference in Houston reported that they found nothing with a half-life in the range of 0.1 second.

The original Soviet experiment—bombardment of plutonium-242 with neon-22—was not done because the heavy ion linear accelerator (HILAC) at Berkeley cannot accelerate neon-22 with the desirable intensity. Ghiorso argues that reactions can be predicted well enough for Berkeley to be confident that they would have produced the spontaneous fission isotope with a half-life of 0.1 second with their reactions if such had been produced by the Soviets.

It is obvious in talking with the Berkeley researchers that they feel that there is only a remote possibility that the Soviets have produced element 104. They have allowed for this possibility by stating that they will withdraw their proposed name if the early Soviet work is verified.

There are still unresolved claims about elements 102 (nobelium) and 103 (lawrencium), and there are already conflicting experimental reports for element 105. Flerov's group at Dubna has reported that a few atoms of element 105 may have been produced, but they realize that the work was done under extremely difficult experimental conditions and have made no strong claims. The Berkeley team has repeated the experiment with negative results and has now embarked on its own program for the synthesis of element 105.

Superheavy Elements

At the Mendeleev centennial in Houston, there was also disagreement among theoretical physicists, but it was of a more scholarly nature and dealt with the possibility of finding stable elements centered around element 114.

Until a few years ago, most nuclear physicists thought that the study of synthetic elements that had been going on since the 1940's would end somewhere around element 110, because heavier elements would decay by spontaneous fission too rapidly to be detected. Recent theoretical work has altered this view.

The three modes of nuclear disintegration—beta decay, alpha decay, and spontaneous fission—are independent of one another and are treated theoretically by separate methods.

The liquid-drop model has been the backbone of theoretical work on neutron-induced fission. Although useful for studies of elements around uranium, it predicts that all superheavy elements would decay by spontaneous fission too rapidly to be studied, and it fails to account for certain regions of stable nuclei in existing elements.

The shell model accounts for these regions of stability by showing that "magic numbers" of neutrons and protons form stable shells analogous to Bohr electron shells. For example, the unusual stability of lead-208 is attributed to the double "magic number" of 82 protons and 126 neutrons. The shell model begins to fail as one moves away from the stable shells, and therefore several years ago Sven Nilsson of the Lund Institute of Technology, Copenhagen, developed a method of calculating orbitals for the shell model that made it applicable to deformed nuclei.

At about the same time the liquid-drop model was made useful for theoretical work on spherical nuclei by incorporation of some features of the shell model. This was done by William Myers and Wladislaw Swiatecki of the University of California (Berkeley), and further advances in the amalgamation of the two theories was accomplished by Vilen Strutinsky of the Kurchatov Institute near Moscow.

In 1966 Heiner Meldner, now at the University of California (San Diego), used the combined theories to predict that fission-stable nuclei should exist at atomic number 114. The predictive power of the nuclear calculations has been constantly checked against measured properties of newly discovered isotopes (now well over 100) of the actinide elements, and there is now considerable confidence in the method.

Actual predictions of half-lives of superheavy elements vary by several orders of magnitude, because it is possible for theorists to select any of several combinations of shell model and liquid-drop considerations for spontaneous fission decay and then to apply one of several methods of calculating alpha and beta decay. There is, however, general agreement that an island of stability should exist around element 114, and many theorists believe that element 110^{294} may be the most stable isotope in the region.

On the basis of even the most optimistic estimates of half-lives one would have to conclude that (i) if superheavy elements are on the earth or in cosmic rays as the result of primordial or stellar production, they are barely detectable; (ii) they would also be barely detectable if they are produced in the detonation of nuclear devices; and (iii) it is probably not possible to produce superheavy elements with existing accelerators.

In spite of this gloomy prognosis the possibility of detecting elements that were not conceived of a few years ago has prompted investigators from several countries to look for them in nature, and searches have been going on for about a year and a half. Although a few investigations have revealed fission tracks that could be attributed to heavy elements, track analysis is quite difficult and no one is claiming discovery of element 114 yet.

New Accelerators

Most researchers agree that if superheavy elements are found at all the discovery will not be in natural sources but as the result of ion bombardment. To do this accelerators are required that can obtain at least 5 million electron volts per nucleon by accelerating heavy elements that have been stripped of many electrons—a capability not possessed by existing devices.

Atomic Energy Commission Chairman Glenn Seaborg told *Science* that the AEC has received about ten proposals for heavy-ion accelerators as the result of interest prompted by the prospect of superheavy elements. With current budget conditions it is unlikely that any of the devices, which would cost up to $25 million, will be funded in the near future, but there are plans to modify two existing accelerators and to build several others. The Berkeley HILAC will be rebuilt during the first half of 1971, and the 3-meter cyclotron at Dubna will be converted to a 4-meter instrument. Several other countries, including France and West Germany, are also building machines.

A cyclotron being completed at Orsay, France, will be capable of accelerating ions as heavy as krypton, and the 4-meter Soviet instrument will be able to accelerate xenon. It may be that the products formed by bombardment with these particles will have too few neutrons to be stable. The new HILAC will be able to accelerate uranium ions, so it will have the potential of forming neutron-rich isotopes with reactions such as the bombardment of uranium-238 with uranium-238.—ROBERT W. HOLCOMB