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Chemical Identification of a Long-Lived Isotope of Dubnium, a Descendant of Element 115

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The recognition criterion for discovery of a new chemical element includes two aspects, the characterization properties and the assignment properties. In this paper, we will discuss the status of element 115 experiments that have been performed in Dubna, Russia, highlighting the characterization and assignment properties as they specifically relate to a recent experiment. After discussing the status of what is known about the decay properties of element 115 [1], observed previously using the Dubna Gas-Filled Recoil Separator, we will discuss the prior chemical studies that have been performed on the Db descendant of element 115 [2]. Following the success of that experiment, some additional chemical information was desired. Two separation chemistries were then developed at LLNL and JINR. LLNL utilized reversed phase chromatography and JINR utilized anion exchange chromatography to perform not only +4/+5 separations, but also intra-group separations, where Nb-like and Ta-like fractions were eluted. The results from an experiment using these chemistries for the first time during December 2005 in Dubna, Russia, will be compared with prior chemical results. We will conclude with a discussion of possible enhancements to the work already performed and the current status of the future experimental plans.

1. INTRODUCTION

In 2003, an experiment was performed at JINR using the Dubna Gas-Filled Recoil Separator (DGFRS) to determine the production and decay properties of Element 115. The details of this experiment are reported in [1]. To summarize: Thin (0.36 mg/cm²) rotating ²⁴³Am (99.9%) oxide targets that were electroplated onto 1.5-mm Ti backing were bombarded with ⁴⁸Ca⁵⁺ ions supplied by the U400 cyclotron. This reaction was run at two different energies (248 and 253 MeV) with a total beam dose of 8.6×10^{18} particles. The suppression factors for beam- and target-like particles at DGFRS are $\geq 10^{15}$ and

$\geq 10^4$, respectively. The transmission efficiency is 35 to 40% for $Z = 114$ to 116 nuclei. In this experiment, four time and position correlated event chains were observed that have been assigned to Element 115. At the lower energy ($E_{lab} = 248$ MeV) three decay chains were observed which consisted of five alpha decays followed by a fission. At the higher energy ($E_{lab} = 253$ MeV) one decay chain was observed that consisted of four alpha decays followed by a fission. The chains observed at the lower beam energy had similar energies and lifetimes and were assigned to $^{288}115$. The chain observed at the higher beam energy was different from the other three and assigned to $^{287}115$. The production cross section for $^{288}115$ was estimated to be 2.7 pb based on these three events. The probability that these decay chains were due to a random coincidence is very small. All of the alpha decay events assigned to $^{284}113$, $^{280}111$, ^{276}Mt , and ^{272}Bh occurred while the beam was switched off; thus the background was much lower than during beam-on periods. The SF occurred much later. There were not any time and position correlated alpha events in the 60 seconds before the SF events that have been assigned to ^{268}Db . There were only seven SF events observed during the entire $E_{lab} = 248$ MeV experiment; the three SF events assigned to ^{268}Db , three SF events assigned to $^{244m,f}\text{Am}$ (EVR-SF correlation within 5 ms), and one assigned to the known background level of long-lived spontaneously-fissioning Cf isotopes that are a result of a previous experiment that used the same set of detectors. The interesting thing about these chains was that the first 5 alpha decays occurred rather quickly (within 1 minute) and the spontaneous fission that terminated the sequence had a half-life of about 20 hours. A half-life of 20 hours is long enough to allow significant aqueous chemistry to be performed using that nuclide.

Understanding the chemical properties of each element is the fundamental basis of chemistry. It is these chemical properties that make an element unique and determine how it can be used. The periodic table was created when Mendeleev observed a periodicity in the behavior of chemical elements as mass increased. Later it was determined that the mass wasn't the physical property that determined the chemical behavior but rather the number of protons in the nucleus, so that is what defines an element. Observations of the current, more complex, periodic table indicate some deviations from simple extrapolations. At the upper end of the periodic table these deviations are generally due to a phenomenon called the relativistic effect. Simply described, relativistic effects result when the innermost electrons approach relativistic speeds because of the large positive charge of the nucleus. This relativistic speed causes the electrons to increase their mass ($E = mc^2$) which results in a reduction in the average orbital diameter. This change to the innermost electrons has a cascading effect on all of the other electrons surrounding the nucleus. These changes can alter the valence electrons and the electron orbitals that are involved in bonding, which can alter the expected chemistry. These relativistic effects can alter simple extrapolations from periodic trends upon which the early part of the periodic table was created. There are models that predict the chemical behavior of an element that include these relativistic effects. Data on chemical behavior can be used to validate these models which would then give more support to the model for predicting the chemical behavior of other elements or the same element under different chemical conditions. An understanding of the alteration in periodic trends for those elements that exhibit relativistic effects can also help in interpreting the chemical properties at the upper end of the periodic table. A chemical experiment to confirm the chemical identity of

the spontaneously fissioning species was desired since it would offer independent proof of the production of Element 115 by identifying the 5- α -decay daughter.

2. EXPERIMENTAL DETAILS

To pursue the chemical identification of the spontaneously-fissioning progeny of $^{288}115$, an experiment was performed in June 2004. If the assignment of Element 115 is correct for the parent, the 5- α -decay descendant would be dubnium. It has been shown experimentally that Db behaves like the +5 species, Nb and Ta (see, for example, [3,4]). Therefore, a chemical experiment was designed to separate +5 species from the abundant reaction products produced. The details of this experiment are presented in [2]. In summary, the reaction $^{243}\text{Am} + ^{48}\text{Ca}$ was performed using a beam energy of 247 MeV to produce $^{288}115$. Twenty-four hour irradiations were performed with $2.5 - 7.5 \times 10^{17}$ particles on target. The reaction products were collected in a copper block. The first five alpha decays occurred within the copper block leaving ^{268}Db . After the irradiation was over, the top 10 μm of the surface was mechanically removed and dissolved in aqua regia. Tracers and La carrier were added. The +3, +4, and +5 species were precipitated with the La carrier upon addition of NH_4OH . The copper remained in solution. The precipitate was washed and dissolved in HCl. After conversion of the solution to nitrate form it was loaded onto an AG 50W-X8 cation exchange column and the +4 and +5 species were eluted using 2M HF. The +3 ions (which include actinides) remained on the column (no ^{88}Y , a +3 species, was detected in the eluant). The sample was dried on a thin polypropylene film and counted. The detection system consisted of solid state detectors surrounded by neutron detectors to look for coincident fission fragments and neutrons from spontaneous fission events. The chemical yield of the +5 elements was about 80% (^{92m}Nb tracer) and the +4 elements was about 60% (^{89}Zr tracer). At the end of the chemistry experiment 15 spontaneous fission events were observed with an average of 4.2 neutrons. The results of this chemical experiment were in agreement with the earlier production experiment performed with DGFRS. The cross section, half-life, and TKE were all within error bars for both of these experiments (see Table 1, this table is derived from Table 2 in [2]).

The June 2004 chemistry experiment provided additional evidence that the spontaneously-fissioning species was ^{268}Db since the fissions occurred in the fraction that contained the +5 species. However there was a significant amount of +4 species also in that fraction, so an additional chemical separation was desired. We wanted to chemically separate the +4 and +5 species to further specify the elemental identification of the spontaneously-fissioning species. In addition, we could also consider creating Nb-like and Ta-like fractions from the +5 species to begin determining if Db behaves more like Nb or Ta under these chemical conditions. With these two ideas in mind JINR chemists and LLNL chemists created separation schemes to use for the next experiment in December 2005.

3. CHEMICAL PROCEDURES

Each group developed a chemical procedure; JINR's procedure was based on anion exchange chromatography and LLNL's procedure was based on reverse phase chromatography. The experimental set up for the December 2005 chemistry experiment was similar to the set up in June 2004. The target/Cu catcher block configuration is shown in Figure

Table 1

Comparison of DGFRS Experiment in 2003 and Chemical Experiment in 2004. Table is derived from Table 2 in [2].

| Property | DGFRS Experiment (2003) | Chemical Experiment (2004) |
|------------------------------------|------------------------------|---|
| Separation method | Kinematic separator | Radiochemical separation |
| Separation efficiency | 35% | 80% |
| Detection method | Decay chains of Z=115 nuclei | SF of Z=105 nuclei |
| ^{48}Ca beam energy | 248 MeV | 247 MeV |
| ^{48}Ca total beam dose | 4.3×10^{18} | 3.4×10^{18} |
| ^{243}Am target thickness | 0.36 mg/cm ² | 1.2 mg/cm ² |
| Number of SF events | 3 | 15 |
| Cross section (Z=115) | $2.7_{-1.6}^{+4.8}$ pb | $4.2_{-1.2}^{+1.6}$ pb |
| Half-life (Z=105) | 16_{-6}^{+19} h | 32_{-7}^{+11} h |
| TKE of SF fragments | ~ 225 MeV | ~ 230 MeV, $\langle \nu \rangle = 4.2$ |

1. The same reaction was used but the irradiation time was 40 hours for the December 2005 experiment. The reaction products were again collected in a copper block and a 10 μm layer was removed and dissolved in aqua regia. Tracers and carriers were also added. This time the tracers were ^{177}Ta , ^{175}Hf , ^{92m}Nb , ^{89}Zr , and ^{173}Lu . The carriers were La, Ta, Hf, Nb, and Zr. A hydroxide precipitate was also used to remove the copper from the +3, +4, and +5 species. The sample was then processed using either the LLNL chemistry or JINR chemistry.

The LLNL procedure consisted of dissolving the hydroxide precipitate in HCl and then adding concentrated HF to precipitate LaF_3 along with the +4 species. The precipitate was further processed to generate a +4 fraction. The supernatant (which contains the +5 species) was then loaded onto a column of Kel-F that was pre-conditioned with methyl isobutyl ketone. The column was then washed with four different eluents and each was collected in separate fractions (sometimes multiple fractions per eluent). The eluents in order of use were 6N HCl:6N HF, 3N HCl:1N HF, 1.5N HCl: 0.5N HF, and H_2O . Each fraction was assayed to determine the locations of the Nb and Ta species. The fractions containing Nb and Ta were then combined to create the Nb and Ta samples, respectively. These samples were then counted in the same detector system that was used in the June 2004 chemistry experiment. The chemical yields were about 80% for each fraction and the total chemistry time from target removal until samples were on the counters was about 5 hours. During development at LLNL the procedure was tested using tracers. After arrival at JINR, the procedure was tested using reaction products in addition to tracers to confirm that the reaction products did not interfere with the separation of +4 species from +5 species nor with the Nb and Ta separation.

The JINR procedure consisted of dissolving the hydroxide precipitate in HCl and then adding concentrated HF to precipitate LaF_3 along with the +4 species. The supernatant (which contains the +5 species) was loaded onto an anion exchange column in 0.3M

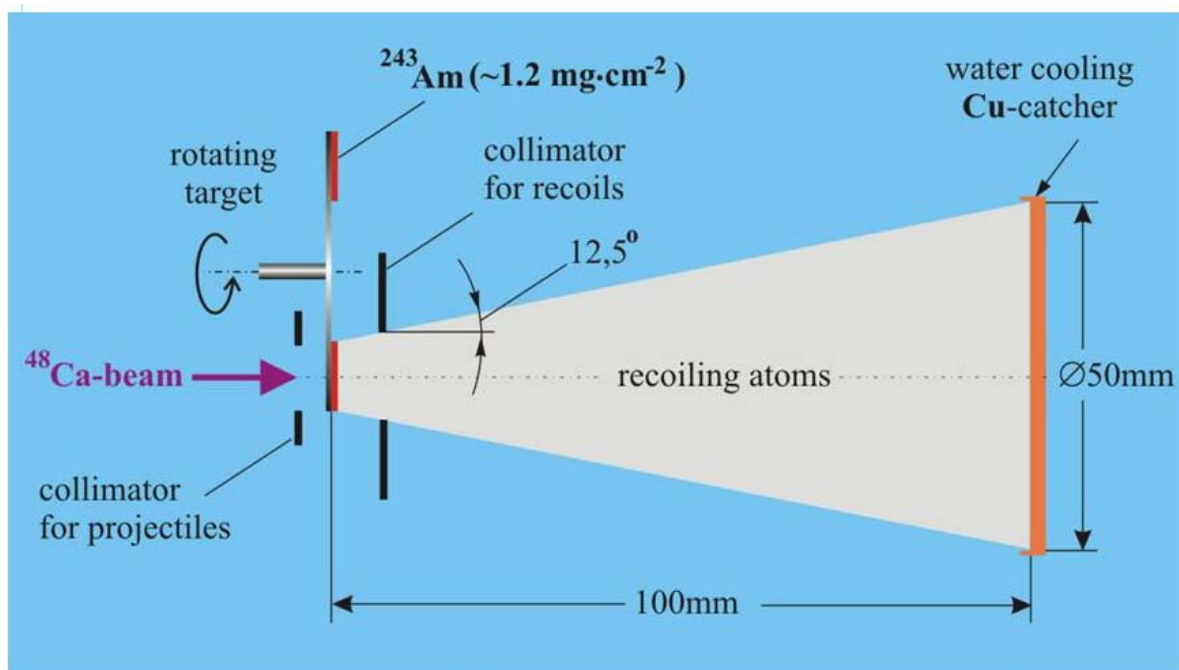


Figure 1. The target/Cu catcher block configuration for both the June 2004 and December 2005 chemical experiments.

HCl:4M HF. For four of the irradiations, a separate Nb fraction was obtained by eluting using 1.5M HF: 3M HNO₃ with an 80% yield. A Ta fraction was then obtained by eluting with 1.5% H₂O₂:0.5M HNO₃ with a 90% yield. For the other two irradiations, both the Nb and Ta were eluted with 1.5% H₂O₂:0.5M HNO₃ with an 80% yield. All elutions were collected as several small fractions which were assayed to determine the appropriate fractions to combine for the counting samples. These samples were counted using the same counters that were used in the June 2004 experiment. Development of this chemical procedure was performed using a combination of tracers and reaction products.

4. RESULTS

A total of eight irradiations were performed in December 2005 where these chemistries were employed. For two of the irradiations, the LLNL procedure was used and for the other six irradiations, the JINR procedure was used. For three of the irradiations, a +4 fraction was generated in addition to the +5 fractions to determine if the spontaneously fissioning activity was in the +4 fraction. From five of the irradiations, a spontaneous fission was observed in a counting sample (two from the LLNL chemistry and three from the JINR chemistry). All of the spontaneous fissions were observed in a +5 fraction; three of the +5 fractions had been further partitioned into Nb and Ta fractions and in each of these cases the spontaneous fission occurred in the Ta fraction. At least one spontaneous fission was observed for each of the chemistry procedures used. It is unfortunate that the counting

samples produced using these new chemistries were thick with respect to expected fission energies, which resulted in rather low observed spontaneous fission energies (the fission fragments observed ranged from 3 to 66 MeV, but they were obviously spontaneous fissions since all were accompanied by at least 1 neutron). The December 2005 experiment is summarized in Table 2. If the spontaneous fission events were background events, one would expect them to occur randomly through all fractions rather than only in the +5 or Ta fractions.

5. CONCLUSIONS

The 2003 DGFRS experiment identified an isotope of element 115 which decayed by five α -decays to a spontaneously fissioning nuclide with a long half life that was assigned to ^{268}Db . The June 2004 chemical experiment provided evidence that the spontaneously fissioning species behaved like a +4 or +5 element, consistent with Db. The December 2005 chemical experiment provided evidence that the spontaneously fissioning species was indeed a +5 element, strengthening the Db assignment and also the element 115 parent assignment.

Although in the December 2005 chemical experiments the Db species appeared in Ta-only fractions three times and not in Nb-only fractions at all, one cannot make any conclusive statements about the behavior of Db since two different chemical systems were used. One or two events is not enough to make conclusive statements about chemical behavior.

In 2003, there were 3 events observed using DGFRS. In 2004, there were 15 spontaneous fissions observed following chemical separations. In this experiment, there were 5 spontaneous fissions observed following chemical separations. Using all of these events, the half life of ^{268}Db is determined to be 28_{-4}^{+11} hours.

6. FUTURE

Currently, there are several different directions being pursued to study the chemical behavior of heavy elements. If these same or similar chemistries are to be performed in the future, thinner counting samples need to be made to improve the measurement of the fission fragment energies. There is also interest in continuing to pursue Element 112 chemistry and possible Element 114 chemistry using the COLD apparatus and the reaction $^{244}\text{Pu} + ^{48}\text{Ca}$ (see Robert Eichler's paper from this conference). While we have shown that the spontaneously-fissioning species is Db, which would then indicate that the 5-alpha parent is Element 115, we have not measured the exact mass of the isotope. To perform that task, JINR is continuing to pursue measuring the mass of the element 105 (Db) using MASHA (Mass Analyzer for Super Heavy Atoms). We are also pursuing the possibility of performing automated chemistry using a gas-jet transfer system on Element 105. LLNL is also pursuing the possibility of developing an automated chemistry apparatus for Element 114 chemistry experiments. The direction that this work proceeds is dependent upon the results of the COLD apparatus Element 114 experiments that are planned for 2007.

7. ACKNOWLEDGEMENTS

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Table 2: Summary of December 2005 Chemical Experiment.

| Date | Beam Integral (particles) | Nb Fraction | +5 Fraction(s) | Ta Fraction | +4 Fraction | # FF | SF Information FF energies (MeV) | # n | Chem. Proced. |
|-----------|------------------------------|-----------------|-----------------|-------------|-------------|------|-------------------------------------|-----|---------------|
| 10Dec2005 | 4.5×10^{17} | No SF* | 1 SF at 30 hrs* | No SF† | No SF† | 2 | 22 + 3 | 6 | LLNL |
| 12Dec2005 | 5.2×10^{17} | 1 SF at 27 hrs* | | No SF* | No SF* | 1 | 51 | 1 | JINR |
| 14Dec2005 | 4.2×10^{17} | No SF* | 1 SF at 16 hrs* | | - | 2 | 45 + 5 | 2 | LLNL |
| 16Dec2005 | 4.2×10^{17} | 1 SF at 37 hrs* | | No SF* | No SF* | 2 | 40 + 53 | 3 | JINR |
| 18Dec2005 | 4.2×10^{17} | No SF* | No SF* | No SF* | - | - | - | - | JINR |
| 20Dec2005 | 3.9×10^{17} | No SF* | 1 SF at 18 hrs* | | - | 2 | 66 + 8 | 2 | JINR |
| 22Dec2005 | 4.4×10^{17} | No SF† | No SF* | No SF† | - | - | - | - | JINR |
| 24Dec2005 | 4.4×10^{17} | No SF* | No SF† | No SF† | - | - | - | - | JINR |

* Sample counted for 96 hours.

† Sample counted for 48 hours.

‡ Two samples generated, one counted for 48 hours, the other was counted for 1200 hours.