Determination of Uranium, Molybdenum and Aluminum of U-Mo Alloy by Isotope Dilution Mass Spectrometry and Inductively Coupled Plasma Atomic Emission Spectrometry

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1. Introduction

The quality assurance of the uranium compound in nuclear industry is very important because that is useful for a reactor work as well as for the effective utilization of a nuclear fuel. Several analytical approaches have been applied to the determination of component elements of uranium compound. Thermal ionization mass spectrometry (TIMS) is quite useful for analyzing very small amounts of uranium compound, due to its high accuracy and precision for isotopic measurement. Uranium and molybdenum in uranium alloy can be measured using isotope dilution mass spectrometric techniques (IDMS), and can be also determined with a detection limit of 0.1-0,2 μg/mL by inductively coupled plasma atomic emission spectrometry (ICP-AES) [1,2].

The aim of the present work is to determine simultaneously the contents of U, Mo and Al, and the their isotopic compositions for U and Mo in a U-Mo alloy sample manufactured at KAERI using the TI-IDMS and ICP-AES techniques, to determine the respective validity of the methods

2. Experiments

2.1 Chemicals

The Certified ^{233}U (99.470atom%) and ^{96}Mo (95.78atom%) spikes were obtained from Oak Ridge National Laboratory (ORNL). These spike solutions were prepared by dissolving their oxides in 8 M HNO_3-0.01 M HF. The concentrations of the spike solutions were prepared by calibrating them with a standard solution. Uranium, molybdenum, aluminum and silicon standard solutions were obtained from AccuTrace and Spex Industries Inc.

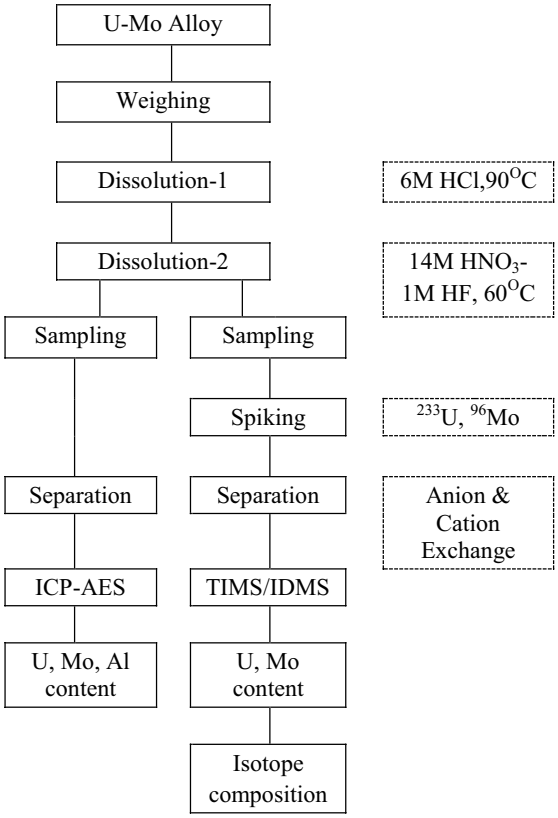

Fig. 1. Analytical processes for the determination of U, Mo and Al in the U-Mo alloy sample.

2.2 Sample preparation, separation and isotopic measurement

The U-Mo alloy consists of an aluminum matrix dispersion of uranium 7wt.% molybdenum (U-7Mo) metallic alloy. The matrix contains a little of silicon and the cladding material of the U-Mo alloy consists of aluminum alloy. The dissolution procedure of the U-Mo alloy sample was composed of two-steps for sequential dissolution of the cladding material and the alloy with 6 M HCl and 14 M HNO_3-1M HF mixture [2]. Chemical separations for IDMS and ICP-AES were carried out by the separation precedures using two sequential anion and cation exchange resins. The basic processes in the analytical laboratory for the

determination of U, Mo and Al are shown in Figure 1. Chemical separation for IDMS was carried out for both the unspiked and the spiked sample solutions in the same experimental conditions. The isotopic compositions of U and Mo in the unspiked and spiked samples were determined using a thermal ionization mass spectrometer (Finnigan TRITON).

2.3 Determination of U and Mo by IDMS

The concentrations of U and Mo in the sample solution were determined by the IDMS according to Eq. (1):

$$C_n = C_a \cdot G_a/G_n \cdot M_n/M_a \cdot (R_a-R_m)/(R_m-R_n) \cdot (\sum nRi)/(\sum aRi) \quad \text{---------- (1)}$$

Where,

C_n : concentration of Mo in sample soln. (μg-Mo/ mL)
C_a : concentration of Mo in spike soln. (μg-Mo/mL)
G_n : volume of sample soln. taken (mL)
G_a : volume of spike soln. taken (mL)
M_n : mean atomic weight of Mo in sample
M_a : mean atomic weight of Mo in spike
R_a : ratio of two basic isotopes in spike ($^{96}Mo/^{98}Mo$)
R_m: ratio of two basic isotopes in mixture($^{96}Mo/^{98}Mo$)
R_n : ratio of two basic isotopes in sample ($^{96}Mo/^{98}Mo$)
$\sum nRi$: sum of ratios of total isotopes for basis isotope in sample
$\sum aRi$: sum of ratios of total isotopes for basis isotope in spike

3. Results & Discussion

3.1 Isotopic compositions of U and Mo

Table 1 shows the isotopic compositions of U and Mo in the U-Mo alloy sample measured by the TIMS .

Table 1. Isotopic compositions of U and Mo separated from the U-Mo alloy

Element	Isotope	Atom%
U	U-234	0.0010±0.0002
	U-235	0.223±0.002
	U-238	99.78±0.04
Mo	Mo-92	14.67±0.22
	Mo-94	9.22±0.01
	Mo-95	15.95±0.02
	Mo-96	16.82±0.03
	Mo-97	9.56±0.02
	Mo-98	24.16±0.04
	Mo-100	9.62±0.02

3.2 Total quantities of U and Mo by IDMS

Table 2 shows the contents of U and Mo in a U-Mo alloy sample determined by the IDMS.

Table 2. Determination of U and Mo in the U-Mo alloy by IDMS

Element	g /g-sample
U	0.529±0.011
Mo	0.0406±0.0008

3.3 Determination of U, Mo and Al by ICP-AES

The analytical results of U and Mo in the U-Mo alloy sample determined by the ICP-AES compared with those by the IDMS, and those estimated from the manufacturing history.

4. Conclusion

The contents of U, Mo and their isotopes in the U-Mo alloy can be determined simultaneously by the isotope dilution mass spectrometric method using ^{233}U and ^{96}Mo as spikes. The contents of U, Mo and Al in the U-Mo alloy can be determined simultaneously by the inductively coupled plasma atomic emission spectrometry. A comparison between independently determined values provides a check on the validity of the results

REFERENCES

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[2] J. S. Kim et al., “Dissolution and Simultaneous Determination of U, Mo and their Isotopes of U-Mo Alloy Nuclear Fuel”, Korean Radioactive Waste Society AUTUMN 2014, 155-156 (2014).