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STUDY AND DEVELOP FOR METHOD OF RADIOCHEMICAL NEUTRON ACTIVATION ANALYSIS (RNAA) TO DETERMINE THE CONCENTRATIONS OF TRACE AND ULTRATRACE ELEMENTS IN PURE MATERIALS (ALUMINUM, TIN, La_2O_3 AND ZrO_2)

Nguyen Van Minh, Le Tat Mua, Nguyen Mong Sinh, Nguyen Giang, Nguyen Ky Vong, Phan Ba Minh, Le Thi Ngoc Trinh, Nguyen Tien Dat and Le Ba Thuan

Nuclear Research Institute

I. INTRODUCTION

The pure materials are playing a very important role in scientific-technical research and technology. The concentrations of trace elements in these materials are very low (10-4-10-10%). The pure materials chosen in this research are aluminum (containers used for sample irradiation at Dalat reactor and two kinds of other pure aluminum), tin (pure Russian chemicals and refined tin from the Institute of Colored Metal Refinery in Ho chi Minh City), lanthanum oxides and zirconium oxides (pure Russian chemicals and some oxides that were refined at the Institute of Technology For Rare Elements, Vietnam Atomic Energy Commission).

Neutron activation analysis is one of the most sensitive, rapid, accurate methods for determinations of major, minor and trace elements in different materials. This method has been applying in Dalat Nuclear Research Institute since 1984. However, there are a lot of difficulties in the determination of trace elements in these pure materials by using instrumental neutron activation analysis method.

Therefore, the chemical separation in activation analysis is very necessary to reduce radiation background, increase sensitivity and accuracy. In this project, we used different separation techniques such as extraction, ion exchange and precipitation, which were carried out after the samples were irradiated in Dalat reactor. The radioactivity of the analytical elements such as Cu, Se, P, Mn, Fe, Sb, As, Cd, Sn, Sc, Sm, Eu, Ce, Cr, Co, Zn... was measured on the gamma multichannel analyzer connected to HPGe detector and PC.

The obtained studying results show that the method sensitivity ranges from 0.01-0.001 ppm and its error from 10 – 15%.

The radiochemical neutron activation analysis (RNAA) can develop to analyse many other trace elements in different pure materials.

II. EXPERIMENT

1. Matter Base

Dalat reactor, Mutilchannel analyzer

Equipments,

Chemicals, reagents,..,

2. Studying materials

Aluminum: old aluminum container, new aluminum container and thin aluminum.

Tin: pure Russian chemical PA and refined tin

La_2O_3: pure Russian chemical PA and refined La_2O_3

ZrO_2: pure Russian chemical PA and refined ZrO_2

The aluminum and tin samples were cleaned with diluted acid HNO_3 or HCl, then with ethanol and were dried at 105°C.

III. STUDYING OF PROCEDURES AND APPLICATION FOR ANALYSING SAMPLE

3.1. Studying of procedures

Procedures for analysis of each of trace elements in four kinds of studying materials were carried out as following steps:

- Determination of optimal conditions: medium, techniques, ...
- Determination of chemicals efficiency
- Determination of analytical sensitivity

Determination of accuracy:

- International Standards: 15A, Merck, Soil-7
- Artificial sample,...

3.2. Application for analysis of four kinds of studying materials

IV. RESULTS AND DISCUSSION

4.1. Analysis of trace elements in pure aluminum

In table 1 shown the analytical procedures, chemical efficiency (%), sensitivities and error (%) of 6 trace elements in pure aluminum samples. These procedures can apply for studying and analysing the trace elements in pure aluminum and other pure materials and can be carried out in the analytical laboratories at Vietnam.

Table 1: Analysis of trace elements in pure aluminum

Element	Separation Techniques			Sensitivity ppb	Error %
	Before irradiation	*After irradiation*	*Efficiency %*		
Cu		- HNO_3+HCl . On exchange column AG 1x8 . Extract with	84.2	9	15-20

		$PbDDC/CCl_4$ - γ measurement			
Se		- HNO_3+HCl - On exchange column AG 1x8 - Na_2SO_3 - γ measurement	87.1	10	10-15
P		- HNO_3+HCl - HCl, AG 1x8 - HCl, TDO - NH_4Mo - β measurement	85.2	10	15-20
Mn		- HNO_3 - MnO_2 - γ measurement	88.4	50	10-15
Fe As		- HNO_3+ H_2SO_4 - HCl 1:1+NH_4I - H_2S - filtered - HNO_3	86.8 79.9	8 10	15-20

Table 2: Concentrations of trace elements in pure aluminum (ppm)

	Element	Old container	New container	Thin aluminum
1	Cu	14974 ± 355	46.1 ± 3.1	3502 ± 24
2	Se	1.05 ± 0.12	<0.01	<0.01
3	P	1271 ± 58	0.26 ± 0.05	1.53 ± 0.15
4	Mn	3.3 ± 0.2	0.05 ± 0.007	4.3 ± 0.2
5	Fe	4069 ± 141	1001 ± 17	2960 ± 44
6	As	0.13 ± 0.02	0.04 ± 0.02	0.44 ± 0.03

Table 2 show that the cleanness of new aluminum container and thin aluminum is better than old aluminum container.

4.2. Analysis of trace elements in pure Tin

In table 1 shown the analytical procedures, chemical efficiency (%), sensitivities and error (%) of 4 trace elements in pure tin sample. These procedures can apply for

studying and analysing the trace elements in pure Tin and other pure materials and can be carried out in the analytical laboratories at Vietnam.

Table 1: Analysing of trace elements in pure Tin

Element	Separation Techniques			Sensitivity (ppb)	Error (%)
	Before irradiation	*After irradiation*	*Efficiency %*		
Cu		- H_2SO_4 - On exchange column AG 1x8 - Extraction with PbDDC/CCl_4 - γ measurement	84.6	9	10-15
Sb		- H_2SO_4 đ, - HCl 10M - On exchange colum Dowex 1x8 - Elute with H_2SO_4 12M+KI - Toluene extraction - γ measurement	84.7	30	10-15
As		- HCl - H_2SO_4-KI - ZnDDC/$CHCl_3$ - TDO - γ measurement	97	10	15-20
Cd		- Dilute HNO_3 - filtred $Sn(NO_3)_2$ - Cô dịch lọc - HCl - Oxime + $CHCl_3$/axêton - γ measurement	99	20	15-20

Table 2: Concentrations of trace elements in pure Tin (ppm)

	Element	PA Chemicals Tin (ppm)	Refined Tin (ppm)
1	Cu	25.8 ± 3.8	63.2 ± 4.9
2	Sb	0.45 ± 0.04	48.9 ± 3.3

3	As	0.053 ± 0.003	0.116 ± 0.007
4	Cd	1.65 ± 0.52	0.49 ± 0.26

Table 2 show that the cleanness of Tin refined from the Institute of Colored Metal Refinery in Ho Chi Minh City is very good.

4.3. Analysis of trace elements in pure La_2O_3

In table 1 shown the analytical procedures, chemical efficiency (%), sensitivities and error (%) of 6 trace elements in pure La_2O_3. These procedures can apply for studying and analysing the trace elements in pure La_2O_3 and other pure materials and can be carried out in the analytical laboratories at Vietnam.

Table 1: Analysis of trace elements in pure La_2O_3

Element	Separation Techniques			Sensitivity (ppb)	Error (%)
	Before irradiation	*After irradiation*	*Efficiency%*		
Sc		- HCl -TBP/Silica gel - 3N HNO_3 - γ measurement	90.2	15	10-15
Fe		- HCl -TBP/Silica gel - Elute with 0.1N HNO_3 - γ measurement	90.6	20	10-15
Sm Eu		- HCl - HDEHP/sili - Elute with 2M HCl - γ measurement	87.4 81.9	75 87	5-10
Ce		- HCl - HDEHP/sili - Elute with 0.2M HCl - γ measurement	84.2	80	5-10
Cr		- HCl - $HClO_4$ 1M - HMD - γ measurement	96.9	10	20

Table 2 show that the cleanness of refined la2o3 from the Institute of Technology for Rare Elements of Vietnam Atomic Energy Commission is very good.

Table 2: Concentrations of trace elements in pure La_2O_3 (ppm)

	Element	Pure La_2O_3 (ppm) Analytical results	Pure La_2O_3 (ppm) Certificated	Refined La_2O_3 (ppm)
1	Sc	0.0150 ± 0.002	0.02	<0.001
2	Fe	6.8 ± 0.9	7.40	<2
3	Sm	0.075 ± 0.002	0.08	0.05
4	Eu	0.087 ± 0.12	0.10	0.012
5	Ce	1.095 ± 0.131	1.20	0.83
6	Cr	0.075 ± 0.012	0.08	<0.001

4.4. Analysis of trace elements in pure oxide zirconium

In table 1 shown the analytical procedures, chemical efficiency (%), sensitivities and error (%) of 5 trace elements in pure ZrO_2. These procedures can apply for studying and analysing the trace elements in pure ZrO_2 and other pure materials and can be carried out in the analytical laboratories at Vietnam.

Table.1: Analysis of trace elements in pure ZrO_2

Element	Separation Techniques			Sensitivity ppb	Error %
	Before irradiation	*After irradiation*	*Efficiency %*		
Co		- $HF+HNO_3$ - H_2SO_4 - Elute with 4.5N HCl - γ measurement	89.9	10	10-20
Zn		- $HF+HNO_3$ - H_2SO_4 medium, - Anionit column - Elute with 0.5 N HCl - γ measurement	85.4	20	10-20
Sb		- $HF+HNO_3$ - H_2SO_4 + KI - Extract with MIKB - γ measurement	89	20	10-20
Eu, Sm		- $HF+HNO_3$ - H_2SO_4 + cuferon - HDEHP/silica gel	75.5 75.8	10 20	10-20

		column - Elute with 2N HCl - γ measurement			

Table 2: Concentrations of trace elements in pure ZrO_2 (ppm)

	Element	Refined ZrO_2 (ppm)
1	Co	0.28 ± 0.003
2	Zn	0.68 ± 0.04
3	Hf	1280 ± 48
4	Sb	92.3 ± 3.9
5	Eu	0.084 ± 0.008
6	Sm	0.125 ± 0.009

Table 2 show that the cleanness of refined ZrO_2 from the Institute of Technology for Rare Elements of Vietnam Atomic Energy Commission is very good.

V. CONCLUSIONS

Main results of this research:

1. Studying and application of 15 procedures for analysis of trace elements in four kinds of pure materials -such as aluminum, tin, oxide lanthanum, oxide Zirconium- with the range of 0.01 – 0.001 ppm of sensitivity and 10 – 15% range of error. Two of these procedures have been included in the report stated at the 2003 Conference at HoChiMinh City.
2. Re-established and equipped with hood-box a 40 m^2 RNAA laboratory for analysis of trace elements in pure material samples.
3. Equipped with specific equipments for RNAA in order to enhance the analytical capacity and to keep safe for radioactive operations.
4. The research results affirm the feasibility of using RNAA for analysis of trace elements in four chosen kinds of pure materials, also the ability of using RNAA for studying other pure materials.

This research is also a good basement for build – up a specific laboratory of RNAA in the next time.

It is necessary to maintain the research of using RNAA for other kinds of pure material, which popular used in sciencetific and technology application.

REFERENCES

1. J. H Zaaidi, M. Arif, I. Faaatima, Ahmed, I.H. Qureshi. J. Radioanal. Nucl. Chem. 241 (1999) p. p.123-127.
2. W. Maenhaut, F. Adams, J. Hose. J. Radioanal. Nucl. Chem. 6 (1970) p. p.83 - 95
3. K.S. Park, B. Kim, H. J. Woo. J. Radioanal. Nucl. Chem. 151 (1991) p. p.373 - 377
4. J.St-Pierre, G.Kennedy. J. Radioanal. Nucl. Chem. 234 (1998), pp.51 – 54
5. P.L. Paul. J. Radioanal. Nucl. Chem. 234 (1998), pp.55 – 58
6. V.P.Kolotov, I.I.Sadykov. J. Radioanal. Nucl. Chem. 168 (1993), pp.465 – 470
7. Hoàng Như Tố: Độc chất học, NXB Y học. 1970
8. Saclo. Các phương pháp hoá phân tích. Tập 1.NXB
9. Kreskop. Cơ sở Hoá học phân tích.
10. J. Chwaaastowsk, W. Zmijewska, E. sterlinska. J. Radioanal. Nucl. Chem. 196 (1995) p. p.3 - 9
11. W. Maenhaut, F. Adams, J. Hose. J. Radioanal. Nucl. Chem. 6 (1970) p. p.83 – 95.
12. E.Szabo. J. Radioanal. Nucl. Chem. 19 (1974) p. p.23 – 26
13. Nuclear activation techniques in the life sciences, 1967 – 1979.
14. Nuclear activation techniques in the life sciences 1972, :Determination of cadmium and mercury in biological samples and tap water" – page 309.
15. Handbook on Nuclear Activation Crossection – Vienna: IAEA, 1974 – page 558, R. Cher, "2200 m/s neutron activation cross – sections".
16. Journal of Radioanalytical Chemistry, Vol 51, No 1, 1979, page 63, J. Strary', A. Zeman, K. Kratzer, J. Pra'silova' "Selective extration and separation of cadmium using tri – n – octylamine".
17. Radiochemical Methods of Analysis – Proceeding of a symposium, 19 – 23 October 1964, Vol 1, A. Speecke and K. Maes "Determination of Cu, Zn and Mn on cobalt metal by activation analysis".