BJRS

BRAZILIAN JOURNAL
OF
RADIATION SCIENCES
09-01A (2021) 01-18

Sociedade Brasileira de Proteção Radiológica

Direct determination of aluminum in low-enriched UAl_x targets (UAl_x-Al) by ICP OES

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ABSTRACT

The production of molybdenum-99 (^{99}Mo) using low-enriched uranium targets (< 20% ^{235}U) dispersed in aluminum (UAl_x) is a very promising strategy towards the independence in ^{99}Mo local production. A thorough control must be performed to ensure that these targets meet the regulatory requirements to achieve the expected efficiency in the reactor. The determination of the targets' composition is of high interest, because the distribution of Al in different phases may have an impact on the U concentration. Among the techniques used for this purpose, inductively coupled plasma optical emission spectrometry (ICP OES) stands out because of its high sensitivity and precision, allowing for simultaneous determination of several elements in a variety of samples and matrices. However, because U exhibits a complex emission spectrum, spectral interferences are prone to affect the analysis of Al, calling for time consuming preparation steps to remove the U from the matrix. This study proposes a method of direct determination of Al in UAl_x targets through the selection of specific emission lines enabled by the evaluation of the associated interferences on the recovery values.

Keywords: **UAlx, molybdenum, ICP OES, spectral interferences.**

ISSN: 2319-0612
Accepted: 2020-11-22

1. INTRODUCTION

Radioisotopes play a key role in modern medicine practices these days, being widely used for the diagnosis and treatment of many diseases. The demand of radioisotopes has notably increased over the last decade [1-4]. ^{99m}Tc, which is obtained through the ^{99}Mo decay, is the most extensively used radioisotope in nuclear medicine today (bone scans, cardiac perfusion studies and other diagnostic procedures). In Brazil, its use corresponds to approximately 80% of all radio-pharmaceutical applications [2]. It decays with a 6-hour half-life by emitting a high energy electron.

Most of the world's supply of ^{99}Mo for medical diagnostic imaging is produced from solid targets containing enriched uranium (^{235}U). After irradiation in the reactor, these targets are digested in acid or alkaline solutions and ^{99}Mo is recovered through a series of extraction (separation) and purification steps. Although most current ^{99}Mo production uses targets containing highly enriched uranium (HEU), several organizations have developed low-enriched uranium (LEU) targets for small to medium commercial scale ^{99}Mo production [5]. These targets consist of either the U-Al alloy dispersed in an aluminum matrix (UAl_x) or the uranium metal foil. In the first, the dispersion is clad between two thin aluminum plates, while in the latter, the foil is clad between aluminum tubes and separated from the cladding by a recoil barrier, which prevents the foil from bonding to the cladding [6].

Although the production of ^{99}Mo has not yet been implemented in a routine basis, a 2009 report from the USA National Academy of Sciences concluded that there are no technical reasons limiting an adequate production of ^{99}Mo from LEU targets in the future [7].

UAl_x is widely used as fuel to power research reactors worldwide [1, 4, 8]. The UAl_x-Al clad fuel plates consists of low enriched uranium aluminide dispersed in an aluminum matrix. The uranium aluminide is an alloy obtained by melting and casting operations. As a result of this process, the alloy usually contains a mixture of UAl_3, UAl_4 and UAl_2. A typical composition is around 63% UAl_3, 31% UAl_4 and 6% UAl_2 [9]. The composition is generally referred to as UAl_x. Due to its reactive nature, UAl_2 is likely to react with the excess aluminum of the plates to form UAl_3 and some of the UAl_3 subsequently reacts with the aluminum matrix to form UAl_4. Therefore, it's reasonable to assume that the finished fuel plates contain only UAl_3 and UAl_4 [10]. The actual fractions will vary from one manufacturer to another, depending on the different processes used to fabricate the powder, core compacts and fuel plates. Concerning the global process, it is possible to choose the best route and the best technique to achieve large scale production [11,12].

The U-Al alloy best suited for use in research reactor contains 14 to 16 wt% of aluminum [8,13]. For the production of ^{99}Mo, the UAl_x targets can be composed of different proportions of aluminum, varying from 5 to 25 wt% [14]. These inter-metallic compounds have been reported to exhibit a very good irradiation behavior.

In 2004, the International Atomic Energy Agency (IAEA) has developed and implemented a Coordinated Research Project (CRP) to support interested countries start a small-scale domestic ^{99}Mo production in order to meet the local demand (of these countries). This project provides all interested countries with access to non-proprietary technologies and methods for the production of ^{99}Mo using LEU targets of UAl_x dispersed in Al (UAl_x-Al) and targets of LEU metallic foils [2,4].

Since then, the technology to produce ^{99}Mo through fission process of LEU targets based on UAl_x-Al has been developed [2,13]. It consists of a metallurgical process involving UAl_x powders and UAl_x-Al mini-plates mounted as a nuclear fuel assembly comprising the plates and the coating [13].

Brazil, like several countries, still strongly depends on imports of ^{99}Mo to meet the internal supply of ^{99m}Tc generators. The ongoing ^{99}Mo world crisis translated into the shortage of its supply [15] has lead the Nuclear and Energy Research Institute (IPEN – CNEN/SP) in São Paulo to initiate efforts to produce ^{99}Mo through ^{235}U fission and officially join the Coordinated Research Project in 2009. The Institute currently provides radiopharmaceuticals to over 300 hospitals and medical clinics across the country. The use of radionuclides compounds in nuclear medicine has climbed at a rate of 10% per year over the last decade and IPEN is Brazil's main supplier of such compounds [1,2].

Several analytical techniques are used to perform characterization and quality control analysis of the uranium compounds used in reactors in order to secure the quality of the product for its intended activity. Among these, spectrometric techniques play an increasingly important part, being commonly used for the aforementioned purpose because they allow simultaneous determination of several elements with minimum amounts of sample [13,14,16].

For the present study, inductively coupled plasma optical emission spectrometry (ICP OES) was chosen for the determination of aluminum in targets of LEU (UAl_x-Al) samples. The analysis of uranium compounds by ICP OES, however, is strongly hampered by spectral interferences attributed to the complex emission spectrum of uranium [16] and usually requires previous steps of separation that may be accomplished by different techniques such as solvent extraction, precipitation, ion exchange chromatography, distillation and electrolysis.

Because UAl_x-Al is a mixture of uranium and aluminum dispersed in an aluminum matrix, one can also expect a spectral interference caused by high concentrations of uranium, leading to high background emission, which, in turn, decreases the signal-to-noise ratio and, consequently, worsens the final results [16-18]. This work aims to develop a method that enables the direct determination of aluminum in UAl_x targets of LEU as a monitoring tool of the production process of the pair ^{99}Mo-^{99m}Tc, by selecting usable wavelengths free of interferences and, thus, eliminating time-consuming preparation steps.

2. MATERIALS AND METHODS

2.1 Apparatus

A Spectro Arcos^{CCD} ICP optical emission spectrometer (Spectro Analytical Instruments Co, Kleve, Germany) equipped with axially viewed plasma and an air-cooled radio frequency generator based upon a free-running 27.12 MHz was used. This system allows adjustments of power between 700W to 1700W. The spectrometer is a Paschen–Runge mount arranged in an Optimized Rowland Circle Alignment (ORCA) with a holographic diffraction grating with two of 3600 and one of 1800 grooves mm^{-1}. It consists of two hollow section cast shells, optimized small volume, and 32 linear charge-coupled-device detectors (CCDs), which allow simultaneously measurements from 130 to 770 nm and complete spectrum capture within 2s. Because of the unique reprocessing capability of the system, new measurements are not required if additional elements or lines are to be determined later. The polychromator´s thermostat was adjusted at 15°C, the optic is hermetically sealed and argon is continuously circulated through a filter (Oxiclear) that removes oxygen, water vapor, and other species that can absorb electromagnetic radiation in the vacuum ultraviolet (VUV) range. Due to high optical transmission in the VUV region the instrument also allows determination of nonmetals. An end-on gas interface was used to minimize interference caused by self-absorption and recombination, providing a wide linear dynamic range and low background. The introduction system was composed of a double pass (Scott-type) spray chamber and a cross-flow nebulizer. All relevant ICP operating parameters are software-controlled, allowing easy selection of the optimum operating conditions.

2.2 Reagents and samples

All solutions were prepared using reagents of analytical grade unless otherwise specified. Ultrapure water was supplied by Milli-Q® water purification system (Millipore, Belford, MA, USA). A solution of 5000 mg L^{-1} of uranium was prepared weighting 0.601 g of U_3O_8 (0.847 gU/g; 0.721% U-235) from NBL (New Brunswick Laboratory,129-A, 99,9% of purity), which was further dissolved in 5 ml of concentrated nitric acid (HNO_3 65%, *Merck*, Darmstadt, *Germany*) and later made up to a final volume of 100 mL with ultrapure water. A stock solution of 1000 mg L^{-1} of $Al(NO_3)_3$ (Aluminum ICP standard Certipur®, Merck, Darmstadt, Germany) was used to prepare solutions for the interference tests and the standard calibration solutions ranging from 1 to 70 mg L^{-1} of Al in a 1% (v v^{-1}) nitric acid aqueous solution.

2.3 Procedure

All glassware and polypropylene flasks (Falcon tubes) used for sample and storage solutions preparations were cleaned with detergent solution and deionized water, with subsequent immersion in a 10% (v v^{-1}) aqueous solution of HNO_3 for 24 h, then repeatedly rinsed with high-purity deionized water, dried and stored in a closed polypropylene container.

The scanning of an aluminum standard solution of 5 mg L^{-1} in a 1 % (v v^{-1}) HNO_3 aqueous solution was performed to select the best emission line and signal-to-background ratio. The intensity of the Mg II 280.265 nm/Mg I 285.208 nm ratio emission lines was used to check the robustness of the instrument [19].

The observation zone was automatically set by the software Smart Analyzer Vision 3.01.0753. Optimization of the instrumental parameters was achieved with an Al solution of 5 mg L^{-1} in a 1 % (v v^{-1}) HNO_3 solution by varying RF power from 1.2 to 1.5 kW, auxiliary gas flow rate from 0.5 to

1.0 L min^{-1} and nebulizer gas flow rate from 0.5 to 1.5 L min^{-1} for optimum signal to noise ratio. The instrumental parameters and the analytical lines are displayed in Table 1.

Table 1: ICP OES instrumental parameters for Al determination in UAl_x samples

Parameter	Instrumental Conditions
RF Power (kW)	1.4
Plasma gas flow rate (L min^{-1})	13.0
Auxiliary gas flow rate (L min^{-1})	0.8
Nebulizer gas flow rate (L min^{-1})	0.8
Injector tube diameter (mm)	2.5
Spray chamber	Double pass
Nebulizer	Cross flow
Wavelengths (nm)	
Al	167.078(II[b]) 396.152(I[a]) 394.401(I) 176.641(I) 308.215(I) 309.271(I)

[a](I): atomic lines; [b](II): ionic lines

The limits of detection (LOD) were calculated according to Eq. 3 using the background equivalent concentration (BEC) and signal-to-background ratio (SBR), which were calculated by Eq. 1 and Eq. 2, respectively [19-21]:

$$BEC = \frac{C_{rs}}{SBR} \tag{1}$$

$$SBR = \frac{I_{rs} - I_{blank}}{I_{blank}} \tag{2}$$

$$LOD = \frac{(3\ x\ BEC\ x\ RSD)}{100} \tag{3}$$

where, C_{rs} is the multi-elemental reference solution in a concentration of 50 mg L^{-1}, I_{rs} and I_{blank} are the emission intensities for the multi-elemental reference (10 mg L^{-1}) and blank solutions, respectively, and RSD is the relative standard deviation for ten consecutive measurements of the blank solution.

In order to estimate the degree in which the concentration of uranium affects the intensity of the aluminum's emission lines, 7 replicates of a 5 mg L^{-1} aluminum solution and 7 replicates of a 50 mg L^{-1} aluminum solution were prepared in 1% (v v^{-1}) of HNO_3 and in increasing concentrations of uranium matrix, as follows: (i) none; (ii) 50 mg L^{-1}; (iii) 100 mg L^{-1}; (iv) 200 mg L^{-1}; (v) 400 mg L^{-1}; (vi) 800 mg L^{-1} and (vii) 1000 mg L^{-1} (Table 2).

Table 2: Al solutions with increasing concentrations of U.

Replicates	**U (mg L^{-1})**	**Al-5 (mg L^{-1})**	**Al-50 (mg L^{-1})**
(i)	-	5.0	50
(ii)	50	5.0	50
(iii)	100	5.0	50
(iv)	200	5.0	50
(v)	400	5.0	50
(vi)	800	5.0	50
(vii)	1,000	5.0	50

From the intensities and profiles of the Al emission signals it was possible to verify the interference caused by the presence of uranium in the solution.

3. RESULTS AND DISCUSSION

3.1. Influence of U concentration Al analytical signal

After complete dissolution of the UAl_x samples (100 mg) and dilution to a final volume of 50 mL, the concentration of uranium in the solution was 1700 mg L^{-1}, confirmed by the Davies & Gray method [22] for uranium determination. This concentration is enough to cause severe spectral interferences.

Results of the study to evaluate the influence of different concentrations of U (ranging from 50 to 1000 mg L^{-1}) on the analytical signal of Al are shown in Figure 1. Studies using Al concentrations of 5 mg L^{-1} and 50 mg L^{-1} are depicted in Figure 1a and 1b, respectively.

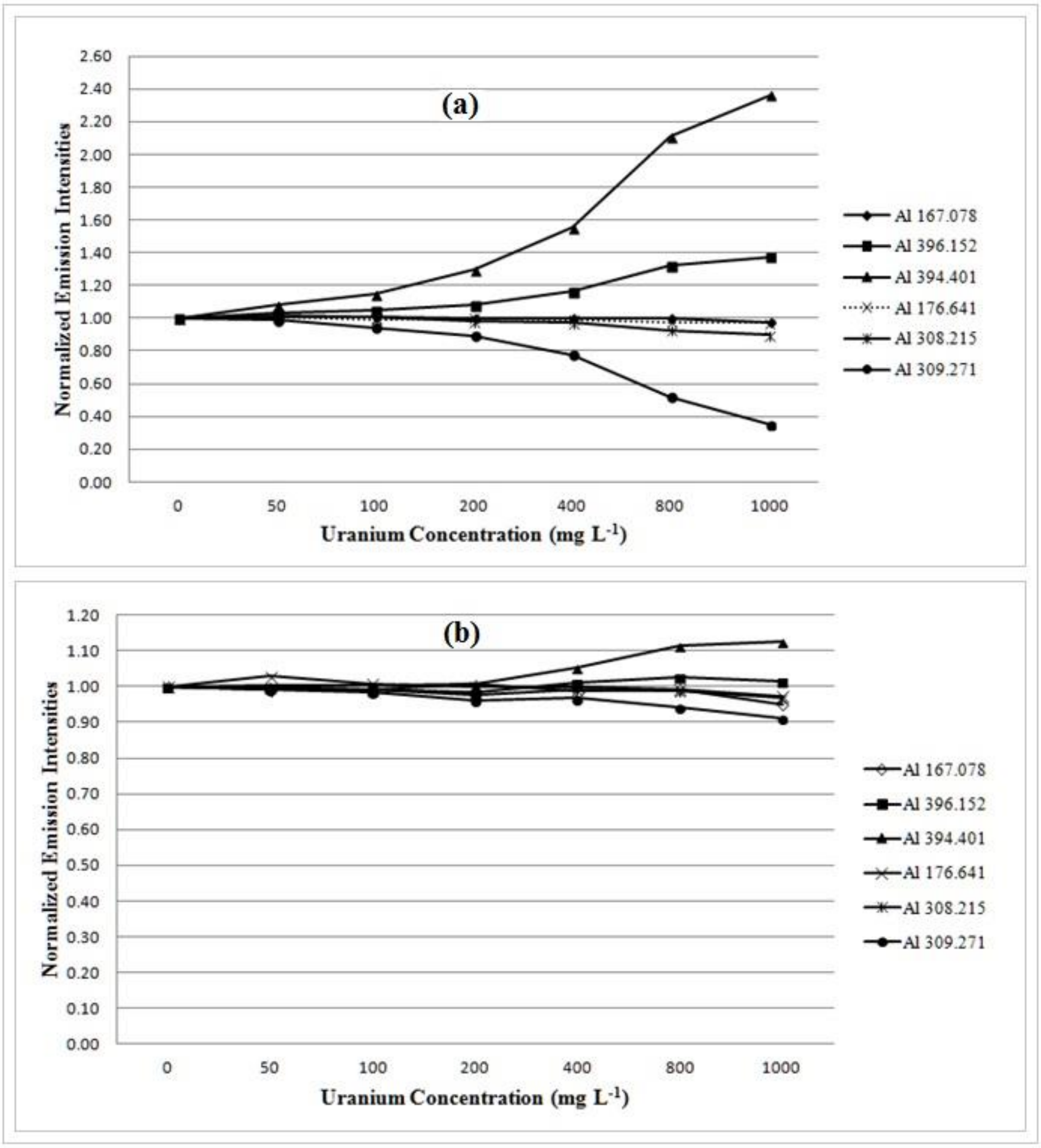

Figure 1: *Response of Al-5 (5 mg L^{-1} aluminum) and Al-50 (50 mg L^{-1} aluminum) solutions to increasing concentrations of uranium.*

Normalized emission intensities can be expressed as the ratio between the analyte intensity signals (Al) in the U matrix divided by the analyte intensity signal without the U matrix. Values approaching "1" are expected in the absence of spectral interference of the U upon the analytical signals. Below "1" values translate as negative interference, causing the signal to decrease, whereas above "1" values represent a positive interference, leading to an increase in the analytical signal.

As shown in Figure 1a, higher concentrations of U predictably exhibit greater influence on low concentrations of Al (5 mg L^{-1}). As the concentration of uranium is lowered, however, the attenuation of this effect is observed. On the other hand, milder effects were observed on the aluminum emission lines for an Al concentration 10 times higher (50 mg L^{-1}), as shown in Figure 1b. Even at a concentration of 5 mg L^{-1}, it is possible to choose three emission lines (167.078 nm, 176.641 nm and 308.215 nm) for aluminum which do not suffer significant interference by higher concentrations of uranium. Only two of the emission lines (394.401 nm and 309.271 nm) suffer pronounced effects caused by uranium in the concentration range of this study.

For these experiments, the concentration values of U were, at least, 10 times higher than those for Al. However, real UAl_x samples consist of an average proportion of 85% of U and 15% of Al where the U/Al ratio is of only 6 times, not 10. The conducted experiments thus prove that there's a safe margin considering that, even when U is present at concentrations 10 times higher than Al, satisfactory recoveries are achieved for the determination of Al. For instance, when the concentration of Al is 5 mg L^{-1}, the expected concentration for U is 30 mg L^{-1} (Al concentration multiplied by a factor of 6). At this concentration, the interference can be considered negligible, being only significant in concentrations approaching 100 mg L^{-1} of U, which would make up for a ratio of 20 times.

So, it is possible to select emission lines for Al that do not suffer from spectral interference caused by high concentrations of U in the sample, enabling a direct determination of aluminum through calibration with aqueous solutions without previous, time-consuming, extraction or separation of the uranium.

Figure 2a shows the scanning of Al at 50 mg L^{-1} prepared in aqueous solution without the addition of U. Comparatively, in Figure 2b, the same solution was prepared, however, in the presence of 100 mg L^{-1} of U. A high background signal ranging from about 190 nm to 470 nm is observed as the U concentration grows. This effect is more evident in Figure 2c, where the solution containing the same concentration of Al is in the presence of 1000 mg L^{-1} of U.

It is shown a large spectral shift due to high background signal from 250 nm on. The effect of U on the analyte emission intensities was more pronounced for higher wavelengths, near to the region of high uranium background emission (250 to 470 nm). This is verified in Figure 3c, in which the line 308.215 nm suffers from an intense spectral interference and an increase in the background signal. This effect does not occur in the line 167.078 nm and is minimum in the line 176.641 nm (Figures 3a and 3b, respectively).

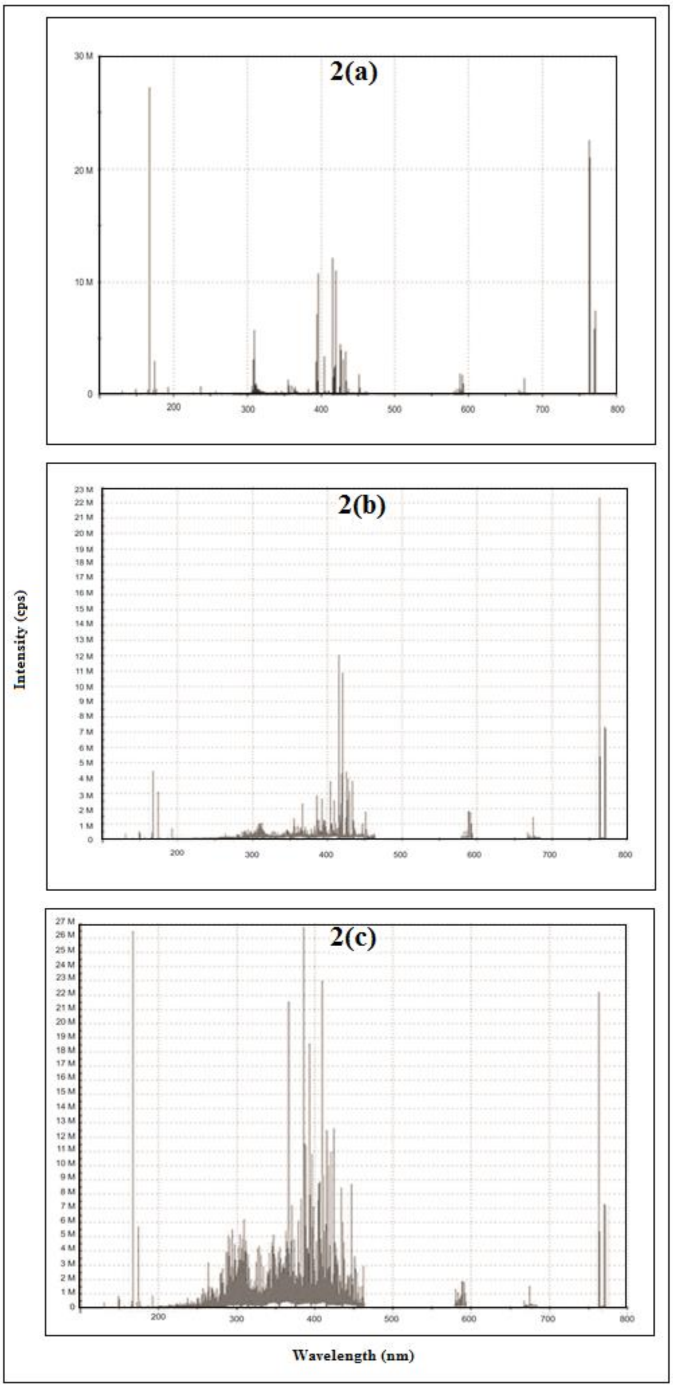

Figure 2: *Response of an Al-50 solution in the absence of U (a), in the presence of 100 mg* L^{-1} *U (b), and in the presence of 1000 mg* L^{-1}*U (c).*

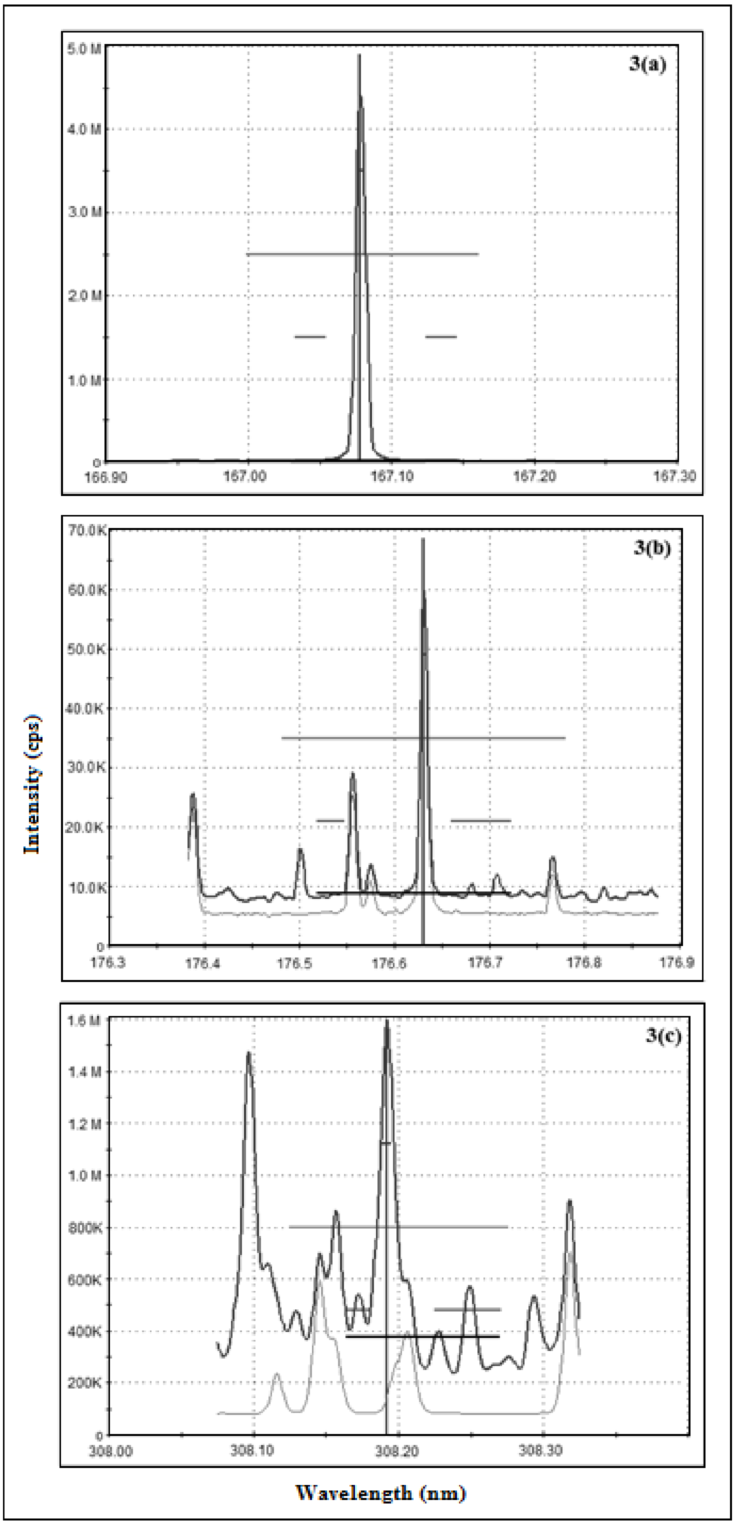

Figure 3: *Al emission spectra : effect of U 1000 mg L^{-1} on Al-50 considering the following lines: (a) 167.078 nm, (b) 176.641 nm, and (c) 308.215 nm.*

3.2 Figures of merit and Al determination

The proposed robustness of ICP is achieved when Mg II/Mg I ratio is higher than 10 [20,22]. A good response was observed using long residence time with auxiliary gas flow rate and nebulizer gas flow rate at 0.8 L min^{-1} and power supply of 1400 W. Under these conditions, the Mg II/Mg I ratio was 6.8. This ratio was multiplied by 1.8 to correct response intensities taking into account the use of an Echelle grating and a solid-state detector [23].

The calibration of the method was accomplished with standard solutions of Al, in concentrations ranging from 1.0 to 50 mg L^{-1} at 1.0% (v v^{-1}) HNO_3 solution (1, 5, 10, 20 and 50 mg L^{-1}). The characteristic parameters of the analytical calibration curve, such as correlation coefficient (R^2), limit of detection (LOD) and quantification (LOQ) are presented in Table 3.

Table 3: Analytical figures of merit calculated for all measured lines.

Al (λ, nm)	**167.078**	**396.152**	**394.401**	**176.641**	**308.215**	**309.271**
[a]R^2	0.9922	0.9999	0.9999	0.9999	0.9994	0.9984
[b]SBR	123.006	59.038	43.623	295.847	16.867	258.088
[c]BEC	0.244	0.847	1.146	0.169	2.964	0.194
[d]LOD	0.014	0.014	0.008	0.022	0.222	0.044
[e]LOQ	0.137	0.143	0.084	0.217	2.216	0.442

[a]R^2: correlation coefficient; [b]SBR: signal to background ratio; [c]BEC: background equivalent concentration; [d]LOD: limit of detection (mg L^{-1}); [e]LOQ: limit of quantification (mg L^{-1}).

The limit of quantification (LOQ) was calculated according to Eq. 4:

$$LOQ = 10\ x\ LOD \tag{4}$$

The quality of the obtained results was verified by doing the addition and recovery tests of Al in decomposed UAlx sample and using synthetic samples containing high levels of uranium. For this purpose, masses of approximately 100 mg of UAl_x sample (identified with the number “2671”), supplied by the Nuclear Fuel Center (CCN/IPEN), were weighed and fully decomposed using a mixture of Lefort aqua regia (inverted aqua regia, consisting of HNO_3 and HCl in a 3:1 ratio) on a digestion block, under a temperature of 50 °C for 30 minutes (final volume was set to 50 mL and achieved by adding ultrapure water).

Two approaches were tested in order to evaluate the proposed emission lines. First, the recovery was measured for each emmission line using synthetic samples containing 5 mg L^{-1} and 50 mg L^{-1} of Al, prepared in the presence of 400 mg L^{-1} of uranium (Table 4). This concentration of U was based on the UAl_x manufacturing process carried out by CCN/IPEN. After that, the recovery was similarly measured in the decomposed UAl_x sample (Table 5) in two conditions: in the absence of Al and in the presence of 5 mg L^{-1} of Al.

Table 4: Recovery of Al in samples containing 400 mg L^{-1} of U.

Al (λ, nm)	**Al (5 mg L^{-1})**	**Recovery (%)**	**Al (50 mg L^{-1})**	**Recovery (%)**
(167.078)	5.60	112	40.17	81
(396.152)	5.73	114	49.26	99
(394.401)	8.83	176	51.95	104
(176.641)	5.11	102	48.36	97
(308.215)	4.67	94	48.35	97
(309.271)	2.34	47	45.72	92

As seen in Table 4, the obtained recoveries ranged from 47 to 176% and from 81 to 104% considering the addition of 5 mg L^{-1} and 50 mg L^{-1} of Al, respectively, in the synthetic samples.

Table 5: Recovery of Al in decomposed UAl_x sample.

Al (λ, nm)	**Without addition (mg L^{-1})**	**With addition of Al (5 mg L^{-1})**	**Recovery (%)**
(167.078)	47.14	48.63	30
(396.152)	74.28	79.64	107
(394.401)	76.53	81.82	106
(176.641)	72.29	77.66	107
(308.215)	73.95	78.50	91
(309.271)	70.29	75.77	110

Regarding the results of the decomposed UAl_x samples, as presented in Table 5, the recoveries ranged from 30 to 110%. According to the guidelines of INMETRO [24], the only emission line that did not show good recovery values (between 80% and 110% for a concentration range of 10 to 100 mg L^{-1}) in the UAl_x samples was Al 167.078 nm (30%).

As already mentioned, starting from an aluminum concentration of 5 mg L^{-1} and multiplying by a factor of 6, the concentration expected for uranium is 30, and at that concentration the interference is negligible, as shown in Figure 1, being only significant in concentrations close to 100 mg L^{-1}. As the concentration of U in synthetic sample was 400 mg L^{-1}, which would give a ratio of nearly 80 times the aluminum concentration, the interference can no longer be considered negligible. Then, considering the real UAl_x sample, the use of the emission lines 176.341 nm and 308.215 nm are enabled, since they exhibited the best comparable recovery values (Table 4).

According to Table 6, the average aluminum concentration found in the provided UAl_x samples was of 17% (wt %), ranging between 15.8 and 19.96 %, being of 19.96% for the 176.341 nm emission line and of 19.27% for the 308.215 nm emission line. These results are in agreement with the expected values considering the manufacturing process (approximately 20% of Al and 80% of U) and were verified by the Student t-test at 95% confidence applied on the results obtained by addition and recovery tests, thus showing that the proposed method is selective and accurate.

Table 6: Al concentration in the UAl_x sample for all measuring lines.

Al (λ, nm)	**% w/w**
(167.078)	19.77
(396.152)	19.42
(394.401)	19.92
(176.641)	19.96
(308.215)	19.27
(309.271)	15.76

Direct Al determination is enabled using the 176.641 and 308.215 nm emission lines, which is also in accordance to Figure 1 (these lines suffer the least deflection to the expected values compared to the others). This also shows that it is possible to work with the 308.215 nm emission line by applying a background correction (Figure 3), thus proving that spectral interferences and high background signals cannot invalidate an emission line for itself without further analytical studies. It is important to note that some spectrometers may not offer the 176.641 nm line. Therefore, the 308.215 nm emission line would be the most suitable for valid measurements.

Because the emission line 167.078 nm is situated in the VUV region, a longer period is required to clean the optical system. This is likely the reason why this emission line, despite being free of spectral interferences, provided poor recovery values compared to the other studied lines, thus showing that a clear spectrum doesn't ensure good results. Therefore, the need to perform analytical tests before choosing or dismissing an emission line.

4. CONCLUSION

The study demonstrated that it is possible to select Al emission lines after correction for spectral interferences resulting from high concentrations of uranium, thus, enabling direct determination of aluminum in uranium compounds, without undergoing previous time-consuming separation or extraction steps. The emission lines 176.341 nm and 308.215 nm exhibited the best recovery values and, therefore, their use for direct determination of Al are enabled, as long as the background correction is applied for the 308.215 nm emission line. The calculated Al mass percentage obtained for the 176.341 nm emission line was 19.96% and for the 308.215 nm emission line was 19.27%, both in accordance to IPEN's declared content of 20% of Al in the produced UAl_x targets. The proposed method is fast and simple and the good results allow its recommendation as a simple and accurate procedure for routine determination of Al in UAl_x nuclear fuel matrices by inductively coupled plasma optical emission spectrometry (ICP OES).

ACKNOWLEDGMENT

We gratefully acknowledge the financial support of MCTI/CNEN, CAPES and CNPq. We would also like to express our eternal gratitude to our deceased colleague, Maurício H. Kakazu, for his significant contribution in this study.

REFERENCES

[1] MARQUES, F. L.N.; OKAMOTO, M. R. Y.; BUCHPIGUEL, C. A. Alguns aspectos sobre geradores e radiofármacos de tecnécio-99m e seus controles de qualidade. **Radiol Bras**, v. 34, p. 233-239, 2001.

[2] DOMINGOS, D. B.; SILVA, A. T.; JOÃO, T. G.; SILVA, J. E. R.; ANGELO, G.; FEDORENKO, G. G.; NISHIYAMA, P. J. B. O. Comparison of low enriched uranium (UAlx-Al and U-Ni) targets with different geometries for the production of molybdenum-99 in the IEA-R1 and RMB reactors, In: **INTERNATIONAL NUCLEAR ATLANTIC CONFERENCE**, 2011, Belo Horizonte, Annals... Belo Horizonte: Comissão Nacional de Energia Nuclear, 2011. p. 24-28.

[3] WIENCIEK, G. F.; VANDEGRIFT, A. B.; LEVYA, A. A.; HEBDEN, A. S. Status and progress of foil and target fabrication activities for the production of Mo-99 from LEU, In: **30TH INTERNATIONAL MEETING ON REDUCED ENRICHMENT FOR RESEARCH AND TEST**, 2008, Washington, Annals... Washington: IAEA, 2008.

[4] GOLDMAN, I. N.; RAMAMOORTHY, N.; ADELFANG, P. Progress and Status of the IAEA Coordinated Research Project: Production of Mo-99 Using LEU Fission or Neutron Activation, In : **29TH INTERNATIONAL MEETING ON REDUCED ENRICHMENT FOR RESEARCH AND TEST**, 2007, Prague, Annals... Prague: U.S. Department of Energy / National Nuclear Security Administration's Office of Global Threat Reduction, 2007.

[5] DONG, D. J.; VANDEGRIFT, G. F. Alkaline peroxide processing of low-enriched uranium targets for ^{99}Mo production -- Decomposition of hydrogen peroxide. **Nucl Sci Eng**, v.126, p. 213-223, 1997.

[6] IAEA - International Atomic Energy Agency. **Good Practices for Qualification of High Density Low Enriched Uranium Research Reactor Fuels, IAEA Nuclear Energy Series No. NF-T-5.2**, Vienna: IAEA, 2009. 74p.

[7] NRCC - National Research Council Committee. **Medical Isotope Production without Highly Enriched Uranium**, Washington, DC: National Academies Press, 2009.

[8] MARAGHEH, M. G.; DAVARPANAH, M. R.; FAZLALI, M.; BOUDANI, M. K.; NOSRATI, S. A. Industrial-scale production of 99mTc generators for clinical use based on zirconium molybdate gel. **Nucl Technol**, v. 169, p. 279-284, 2010.

[9] GOLDMAN, I.; RAMAMOORTHY, N.; ADELFANG, P. Progress in the IAEA Coordinated Research Project on Molybdenum-99 Production using LEU or neutron activation, In: **27TH INTERNATIONAL MEETING ON REDUCED ENRICHMENT FOR RESEARCH AND TEST**, 2005, Boston, Annals... Boston: Argonner National Laboratory, 2005.

[10] LEENAERS, A.; VAN DEN BERGHE, S.; KOONEN, E.; JAROUSSE, C.; HUET, F.; TROTABAS, M.; BOYARD, M.; GUILLOT, S.; SANNEN, L.; VERWERFT, M. Post-irradiation examination of uranium 7 wt% molybdenum atomized dispersion fuel. **J Nucl Mater**, v. 335, p. 39-47, 2004.

[11] MANI, R. S.; NARASIMHAN, D. V. S. Development of kits for short-lived generator-produced radioisotopes, In: INTERNATIONAL ATOMIC ENERGY AGENCY, editor. **Radiopharmaceuticals and Labelled Compounds**. Vienna: IAEA STI/PUB/344, 1973.

[12] PADMANABHAN, S.; RAMAMOORTHY N. Technetium-99m generators: Principles and features, In: RAMAMOORTHY, N.; SHIVARUDRAPPA, V.; BHELOSE, A. A., editors. **Radiopharmaceuticals and hospital radiopharmacy practices: course manual for accreditation/certification of hospital radiopharmacists**. Mumbai: Department of Atomic Energy, Government of India, 2000.

[13] VANDEGRIFT, G. F.; CONNER, C.; HOFMAN, G. L.; LEONARD, R. A.; MUTALIB, A. ; SEDLET, J.; WALKER, D. E.; WIENCEK, T.C.; SNELGROVE, J. L. Modification of targets and processes for conversion of ^{99}Mo production from high- to low-enriched uranium. **Ind Eng Chem Res**, v. 39, p. 3140-3145, 2000.

[14] HAYDT, H. M.; CINTRA, S. H. Nota preliminar sobre a fabricação de elementos combustíveis planos contendo núcleo de ligas alumínio-urânio. **Metalurgia**, v. 23, n. 121, p. 955-959, 1967.

[15] PONSARD, B. Mo-99 supply issues: status report and lessons learned. **EUROPEAN NUCLEAR SOCIETY**, ENS, RRFM 2010, Transactions, 2010.

[16] MALHOTRA, R. K.; SATYANARAYANA, K. Estimation of trace impurities in reactor-grade uranium using ICP-AES. **Talanta**, v. 50, n. 3, p. 601-608, 1999.

[17] REIS, E. L. T.; SCAPIN, M.; COTRIM, M. B. E.; SALVADOR, V. L.; PIRES, M. A. F. Impurities determination on nuclear fuel element components for the IEA-R1 Research Reactor by analytical methods based on ED-XRF and ICP-OES, In: **INTERNATIONAL NUCLEAR ATLANTIC CONFERENCE**, 2009, Rio de Janeiro, Annals... Rio de Janeiro: Comissão Nacional de Energia Nuclear, 2009.

[18] DUNG, N. T. K.; SON, D. T.; VAN TRUNG, H. Application of atomic absorption spectrometry for the quantitative determination of metallic impurities in pure uranium compounds. **Anal Sci**, v. 18, n. 11, p. 1263-1266, 2002.

[19] SOUZA, A. L.; LEMOS, S. G.; SHERLAN, G.; OLIVEIRA, P. V. A method for Ca, Fe, Ga, Na, Si and Zn determination in alumina by inductively coupled plasma optical emission spectrometry after aluminum precipitation. **Spectrochim Acta, Part B**, v. 66, p. 383-388, 2011.

[20] LAJUNEM, L. H. J.; PERÄMÄKI, P. **Spectrochemical analysis by atomic absorption and emission**, 2nd ed. Cambridge, UK: Royal Society of Chemistry, 2004.

[21] MERMET, J. M.; POUSSEL, E. ICP emission spectrometers: 1995 analytical figures of merit. **Appl Spectrosc**, v. 49, n. 10, p. A12-A18, 1995.

[22] Davies, W and Gray, W. A rapid and specific titrimetric method for the precise determination of uranium using iron (II) sulphate as reductant, **Talanta**, v. 11, n. 8, p. 1203-1211, 1964.

[23] MERMET, J. M. Use of magnesium as a test element for inductively coupled plasma atomic emission spectrometry diagnosis. Anal Chim Acta, v. 250, p. 85-94, 1991.

[24] INMETRO – Instituto Nacional de Metrologia, Normalização e Qualidade Industrial. **DOQ-CGCRE-008 – Orientações sobre Validação de Métodos de Ensaios Químicos**, Rev. 05, 2016. 31p.