EVALUATION OF ANALYTICAL PROCEDURES FOR THE DETERMINATION OF CADMIUM, BORON AND LITHIUM IN UAlx SAMPLES BY INDUCTIVELY COUPLED PLASMA OPTICAL EMISSION SPECTROMETRY (ICP OES)

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ABSTRACT

Used in over 80% of the worldwide diagnostic procedures, Technetium-99m (^{99m}Tc), which is obtained from the decay of molybdenum-99 (^{99}Mo), is the most important radioisotope in nuclear medicine. IPEN/CNEN-SP has been developing technologies in order to produce Mo-99 by the irradiation of low-enriched uranium (LEU < 20% of 235U) targets in its research reactor IEA-R1 (IPEN, São Paulo/Brazil). These targets consist of low-enriched uranium dispersed in a matrix of aluminum (UAlx-Al). Several impurities may be incorporated during the target's production process, such as boron, cadmium and lithium, which have a high capture cross section that may reduce the irradiation's efficiency. This study describes a simple and rapid inductively coupled plasma optical emission spectrometric method for the determination of cadmium, boron and lithium in uranium-aluminum (UAlx) dispersion targets. The method involves a previous separation step, in which uranium gets removed from the matrix by chromatographic extraction with the use of a divinylbenzene resin Amberlite XAD - 4 doped with tri-n-butyl phosphate (TBP). TBP selectively separates the uranium, leaving behind the impurities in an aqueous medium for a further quantification by ICP OES. Possible spectroscopic interferences are also discussed in this article, because of the high amount of aluminum in the remaining solution. Experimental and instrumental conditions, such as initial mass, acid solution ratio and amount, resin mass, emission lines and interfering concentrations are carefully established. This method is to be applied for the determination of several others impurities in Ualx in the future, providing means to verify the UAlx targets' compliance to the current established specifications through routine laboratory analysis.

INTRODUCTION

Technetium-99m is the most extensively radioisotope used in nuclear medicine worldwide, corresponding to approximately 80% of all procedures in this area [1]. It is obtained from the decay of its parent isotope Molybdenum-99 in a ^{99m}Tc generator ([99]Mo/^{99m}Tc). However, Brazil still depends on [99]Mo imports to make up for its internal demand of ^{99m}Tc generators. Since 2009, IPEN/CNEN-SP is engaged into developing domestic technologies aiming, in a near future, self-reliance of [99]Mo production.

[99]Mo can be produced from uranium-aluminum (UAlx) dispersion targets of low-enriched uranium (LEU < 20% of [235]U) dispersed in a matrix of aluminum (UAlx-Al). This powder mixture is pressed into very thin foils, which, when irradiated, produce [99]Mo. The uranium-aluminum (UAlx) alloy is obtained by smelting and casting operations. As a result of this process, the alloy may incorporate several impurities during the production process. Some of these impurities, such as the elements cadmium, boron and lithium, are likely to absorb thermal neutrons and, as a result of that, the target will exhibit a decreased efficiency when irradiated in a reactor [2-4].

Therefore, the impurities quantification in UAlx targets presents itself as a routine requirement for IPEN/CNEN-SP. For this, the development of analytical procedures that allow a reliable control of these targets' impurities is of absolute importance. Several techniques may be used to quantify trace elements in nuclear fuel: inductively coupled plasma mass spectrometry (ICP-MS) [5], inductively coupled plasma optical emission spectrometry (ICP OES) [6], flame atomic absorption spectrometry (FAAS) and graphite furnace atomic absorption spectrometry (GF AAS) [7-8], X-ray fluorescence spectrometry (XRF) [9], total reflection X-ray fluorescence spectrometry (TXRF) [10], carrier-spectrographic technique and recently high-performance liquid chromatography combined with inductively coupled plasma mass spectrometry (HPLC-ICP-MS) [11].

Among all these techniques, ICP OES stands out for its simplicity and simultaneous multi-elemental determination capability. However, this technique cannot be directly applied in the determination of trace impurities present in uranium compounds, such as UAlx targets, because of the spectroscopic interferences caused by the emission lines of uranium. Also, one should avoid using highly concentrated uranium solutions to prevent equipment contamination. This matrix issue requires a separation step prior to the analysis per se. The appropriate technique will allow a selective removal of uranium from the compound, isolating the impurities that can undergo further determinations [12].

The determination of impurities in UAlx samples plays a key-role in quality control and the achievement of regulatory requirements. Impurities above specified limits are indicators of contamination that may have occurred during the manufacturing process.

This study describes a procedure to successfully dissolve samples of UAlx powder previously to the separation step, which combines solvent extraction and solid phase adsorption [13-15]. The mounting consisted of a glass column packed with divinylbenzene resin Amberlite XAD-4. The resin had been doped with tri-n-butyl phosphate (TBP), a well-established solvent for liquid-liquid extraction [16-19].

This article also evaluates the feasibility of the determination of B, Cd and Li in the presence of aluminum, since the separation step only removes uranium from the UAlx sample, leaving

a final eluate consisting of a solution in which aluminum is present in approximately 20% of the sample's original mass (about 8,000 mg L^{-1}). This may or may not be an issue, depending on how emission lines of the three elements of interest behave in the presence of such amount of aluminum.

The subject of this study is to evaluate the spectral interferences caused by high amounts of aluminum over spectral emission lines of boron, cadmium and lithium and the extent of these interferences on the analytical results.

1.   MATERIALS AND METHODS

This study was conducted in the Chemical and Environmental Analysis Laboratory (LAQA) of IPEN's Center for Chemistry and Environment (CQMA). The UAlx material was provided by IPEN's Nuclear Fuel Center (CCN).

2.1 Chromatographic separation

Uranium was separated from UAlx samples by liquid-liquid extraction chromatography. This step was performed using columns of 1.8 cm diameter x 22 cm of height, containing a 55 mL resin bed volume consisting of a packed Amberlite XAD-4 resin doped with analytical grade tri-n-butyl phosphate (TBP) and acetone solvents at 150% w w^{-1} (Merck, Darmstadt, Germany).

Masses of approximately 2.0 g of UAlx sample, supplied by CCN (IPEN/CNEN-SP), were weighted, dissolved in high purity reverse aqua regia (Lefort solution) and heated on a heating plate at 50°C for 30 minutes for complete decomposition. The samples were thereafter evaporated to dryness on a heating plate and further reconstituted to a final volume of 10 mL of 5.5 M HNO$_3$.

The chromatographic columns were preconditioned at a flow of 2 mL min^{-1} with 5.5 M HNO$_3$. The reconstituted samples were then loaded onto separated columns and the columns' wash and load solutions were collected in clean beakers. Each of these solutions was brought up to a final volume of 50 mL with 5.5 M HNO$_3$ in volumetric flasks. The uranium retained in each column was later eluted with water and disposed as radiological waste.

2.2 Study of the aluminum spectroscopic interferences

Before the eluted solutions could be analyzed, it was necessary to select the most suitable emission lines for each element of interest. To do so, it was mandatory to isolate the lines which suffer minimum to zero interference caused by the highly concentrated aluminum matrix.

Since the corresponding aluminum mass percentage in UAlx, according to CCN (IPEN/CNEN-SP), is about 20%, the estimated amount of aluminum in a 2.0g of UAlx sample is 0.4 g, yielding a final concentration of 8,000 mg L^{-1} (for a final volume of 50 mL). That is, samples of UAlx that undergo uranium separation by extraction chromatography get rid of the uranium, the major component in its matrix, but continue to bear a fairly big amount of aluminum that can undermine the determination of some impurities, which, when present, correspond to approximately less than 0.01% of the sample's total mass.

To estimate the degree to which aluminum affects the intensity of the emission lines of B, Cd and Li, a stock aluminum solution was prepared through complete dissolution of 1.2 g of high purity aluminum metal powder (Alcoa, Pittsburgh, EUA) using Lefort solution. The diluted solution was evaporated near dryness on a hot plate and diluted to a final volume of 100 mL with HNO_3 5.5M, yielding a concentration of 12,000 mg L^{-1}. A series of aluminum secondary solutions with increasing concentrations were prepared from the stock solution to serve as matrix solutions for the impurities (Table 1).

To each solution, B, Cd and Li were added to yield a concentration of 0.5 mg L^{-1} (Table 1). This value was estimated to fit into the analytical curve and to be lower than the expected concentration of the UAlx sample. The medium in which these solutions were prepared consisted of HNO_3 5.5M, corresponding to that of the chromatographic separation step. The obtained emission intensities, as well as the profiles of the Al emission signals, provided indications of possible interferences caused by the aluminum in the solution.

Table 1: Test solutions prepared with increasing concentrations of Al

Al (mg L^{-1})	B, Cd, Li (mg L^{-1})
-	0.5
100	0.5
500	0.5
1,000	0.5
2,000	0.5
4,000	0.5
8,000	0.5

Notably, the solution containing Al at 8,000 mg L^{-1} mimics a sample of 2.0 g UAlx that undergoes U separation by extraction chromatography. This allows a reasonable approximation of the matrix effects over the impurities that might be present in the sample.

The impurities (B, Li and Cd) were added one by one in order to yield a concentration that approximates to some previous readings of UAlx samples. Also, the standards used to perform the addition, as well as to prepare the calibration solutions, were certified standard solutions of 1,000 mg L^{-1} by Merck (Darmstadt, Germany). The analytical curve for each element consists of three different concentrations: (i) 0.01 mg L^{-1}, (ii) 0.1 mg L^{-1}, and (iii) 1 mg L^{-1}.Three emission lines for each element were selected (Table 2) so that the best line could be chosen, based on the obtained data.

Table 2: Emission lines selected for B, Cd and Li

B (nm)	Cd (nm)	Li (nm)
249.773 (I)	228.802 (I)	670.780 (I)
249.677 (I)	214.438 (II)	670.776 (I)
182.641 (I)	226.502 (II)	670.791 (I)

(I) Atomic line, (II) Ionic line

2.3 Analysis of the impurities

The eluted solutions were analyzed with 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. All relevant ICP operating parameters are software-controlled, allowing easy selection of the optimum operating conditions.

The method's capability of retention for uranium was previously established in the laboratory by Kakazu et al. [15], resulting in nearly 100% retention of the uranium for the samples analyzed. The UAlx samples were then eluted through the column at a rate of one drop per second and the elements of interest were quantified in the collected fractions by ICP OES. The results were compared to the maximum allowed limits [20], shown in Table 3, in order to establish the accordance of the UAlx produced by CCN (IPEN/CNEN-SP) to the specified patterns.

Table 3: Impurity level in UAlx used in Mo-99 target plate

Element	Concentration (mg L^{-1})
B	< 1
Cd	< 1
Li	< 8

3. RESULTS AND DISCUSSION

3.1 Evaluation of aluminum's interference

Aluminum spectral interferences over the emission lines of boron, cadmium and lithium were investigated in aluminum concentrations ranging from 0 to 8,000 mg L^{-1} in 7 test-solutions, as described in Table 1. The results are expressed as normalized emission intensities, which are the ratio of the intensities of the analytes' signals in increasing aluminum concentrations and the intensities of the analytes' signals obtained in the absence of the matrix. Values near "1" are expected, if no spectral interference is observed.

The results for each analyte's emission lines are show in Figures 1, 2 and 3.
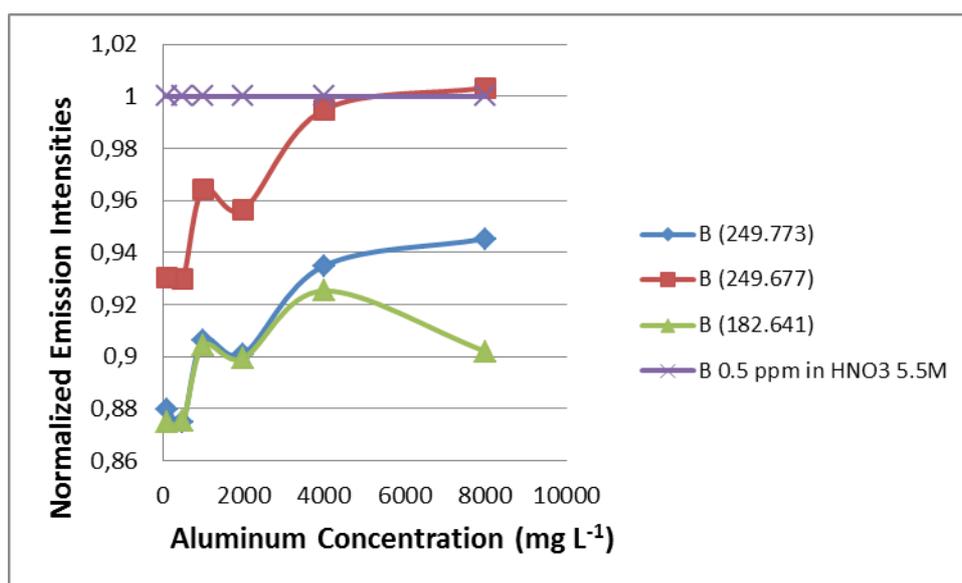

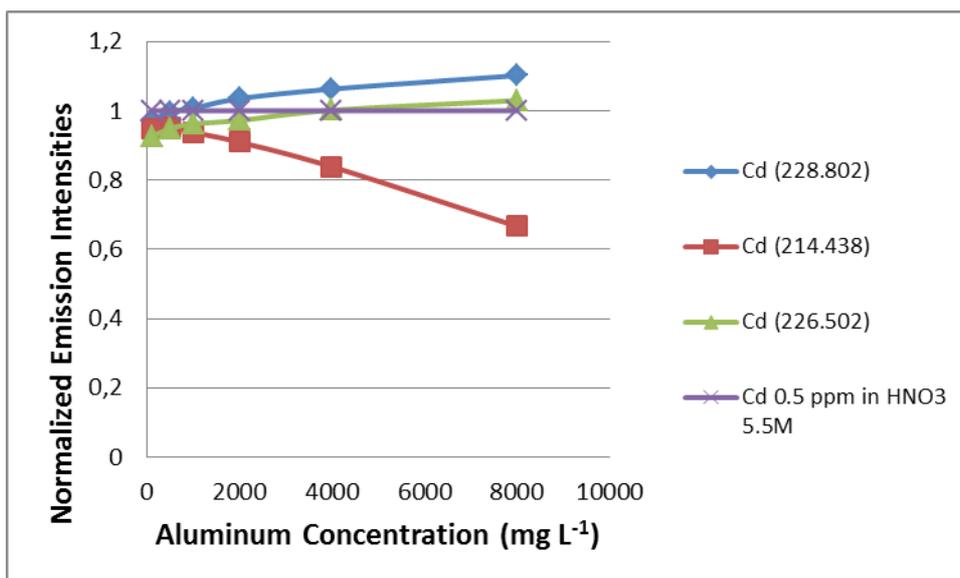**Figure 1. Normalized Emission Intensities for boron.**

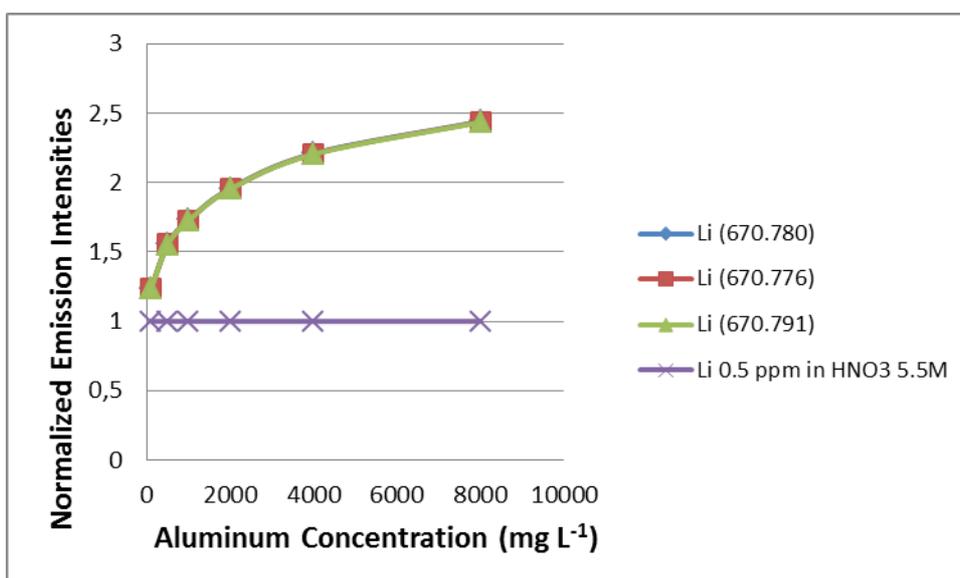**Figure 2. Normalized Emission Intensities for cadmium.**

Figure 3. Normalized Emission Intensities for lithium.

In Figure 1, it is possible to observe that increasing matrix concentrations approximate boron's intensities to the desirable value, when boron is present at a 0.5 mg L^{-1} concentration. All lines exhibit the same pattern, that is, a negative effect, showing that the matrix exerts a similar effect over all emission lines.

Figure 2 shows how the analysis of cadmium suffers the effects produced by matrix interference as its concentration increases. Similarly, Figure 3 shows the response for lithium.

Comparing the response obtained for cadmium and lithium, although both exhibit a positive effect as the matrix concentration increases, this effect is much less pronounced for cadmium than it is for lithium. This is an indication that it may still be possible to accept the deviations suffered for cadmium by choosing the emission line that exhibits the lowest deflection with increasing concentrations of aluminum in the solution.

Lithium, however, demands further studies. Increasing concentrations of aluminum exhibit greater influence over lithium at the studied concentration (0.5 mg L^{-1}). Moreover, all emission lines chosen exhibit the same behavior.

3.2 Selection of working lines

From Figure 1, it's clear that boron's emission line 182.641 nm is the most affected by the matrix. Therefore, this line shouldn't be used to perform determination of boron at 0.5 mg L^{-1} in UAlx samples, where aluminum is present at a concentration of 8,000 mg L^{-1}.
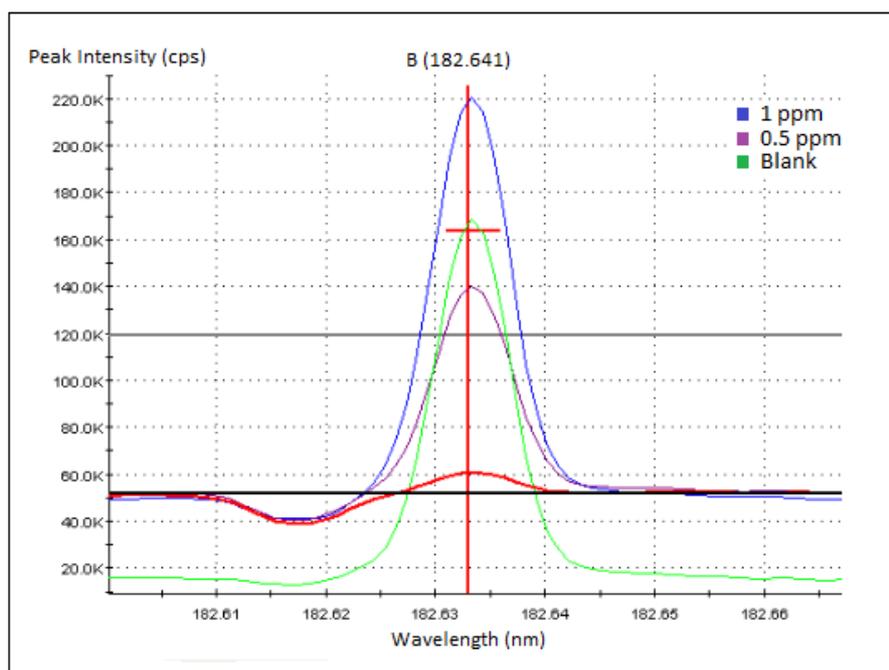
Figure 4. Analytical profile of boron's emission line 182.641 nm.

Figure 4 displays the analytical profile of boron's emission line 182.641 nm, where the peak intensities are obtained from three different solutions: (i) **Blank** (a solution consisting of aluminum at 8,000 mg L^{-1}); (ii) **0.5 ppm** (boron solution of 0.5 mg L^{-1} concentration in aluminum matrix of 8,000 mg L^{-1}); and **1 ppm** (boron solution of 1.0 mg L^{-1} concentration in aluminum matrix of 8,000 mg L^{-1}). This profile is a visual evidence of the spectral interference exerted by the matrix over the selected line's analytical signal.

Boron's emission lines 249.677 nm and 249.773 nm both exhibit a positive effect as the concentration of aluminum increases. The analytical profile of boron's emission line 249.677 nm in Figure 5 shows that this line is free of spectral interferences.

The same profile of the analytical signal was observed for boron's emission line 249.773 nm, thus evidencing that there's no spectral interference going on.
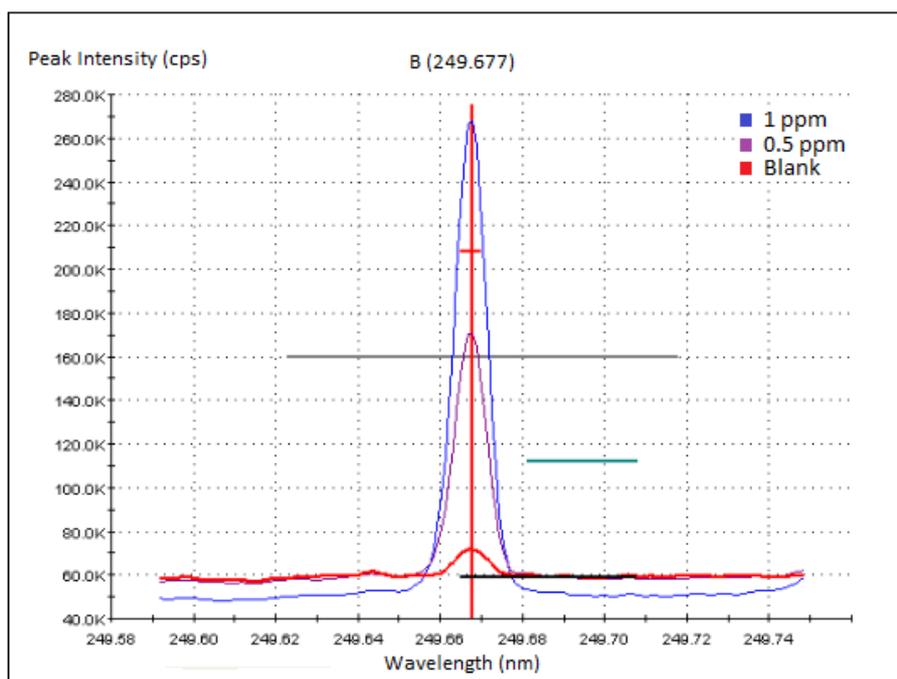
Figure 5. Analytical profile of boron's emission line 249.677 nm.

The lines, 249.773 nm and 249.677 nm, can be considered suitable for boron determinations in matrices containing high concentrations of aluminum. They are both free of spectral interferences and exhibit a similar behavior. Although the line 249.773 nm is more sensitive, the line 249.677 nm shows the smallest deviation from the desirable value and, thus, being chosen as the working line for further determinations in this matrix.
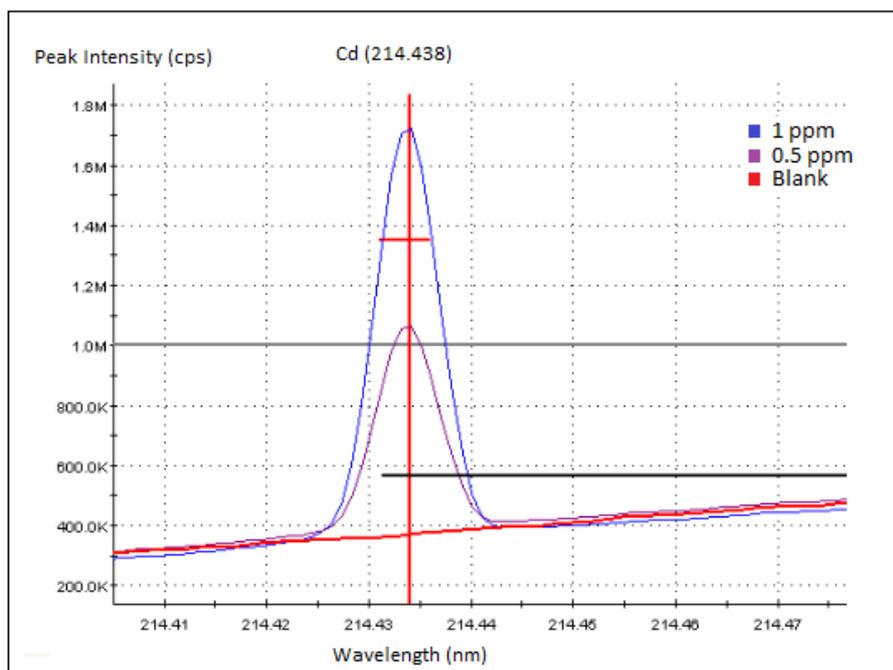
The same parameters have been investigated for cadmium.

Figure 6. Analytical profile of cadmium's emission line 214.438 nm.

The worst line is 214.438 nm (Figure 6), which, compared to the other lines, exhibits a greater deflection as the aluminum concentration in the matrix increases. However, according to its analytical profile, this behavior is not caused by spectral interference.

Cadmium's emission lines 226.502 nm and 228.802 nm both yielded low deviations to the expected result, presenting a slightly positive tendency as the concentration of aluminum increases, and both exhibited analytical profiles that are free of spectral interferences (Figures 7 and 8). This way, they could both be used to determine cadmium at 0.5 mg L^{-1} in the presence of high concentration of aluminum in the sample.
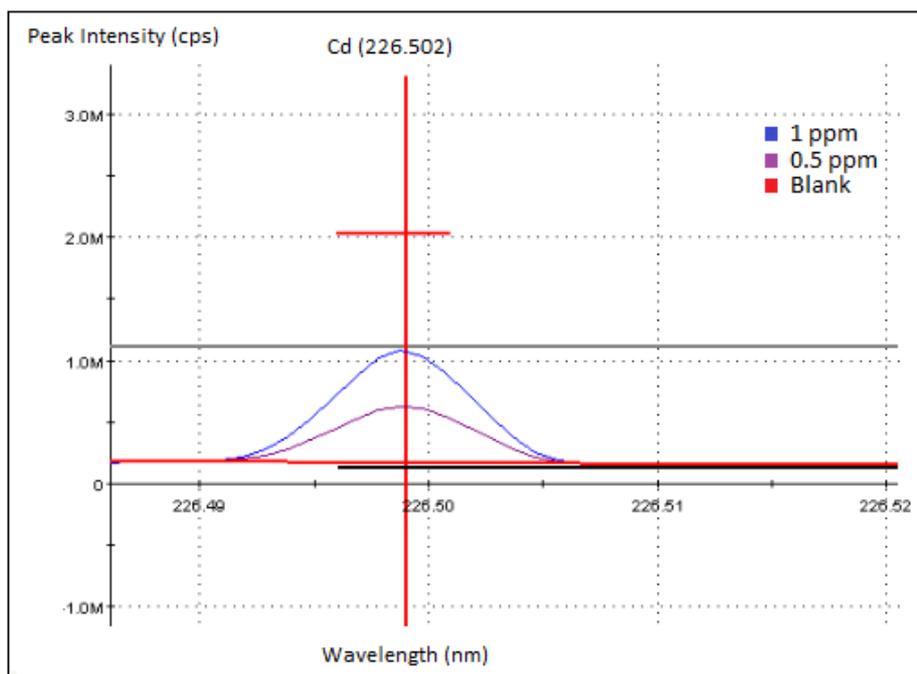

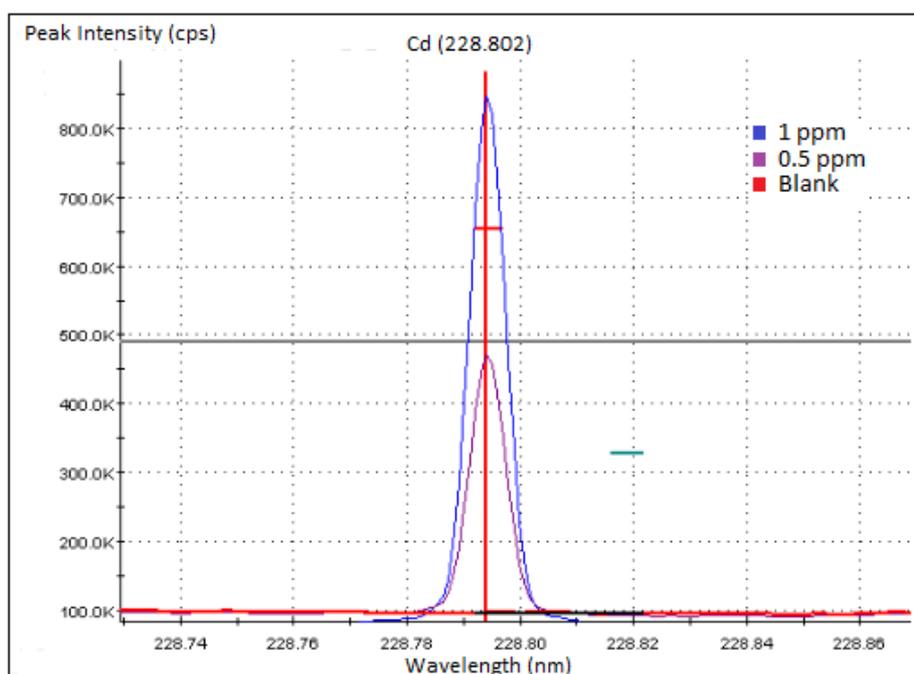**Figure 7. Analytical profile of cadmium's emission line 226.502 nm.**

Figure 8. Analytical profile of cadmium's emission line 228.802 nm.

Although the line 226.502 nm is the best one, according to Figure 2, considering the intensities obtained throughout the range of study, the line 228.802 nm was selected as the working line because it has a better-defined profile of the analytical signal. Moreover, line 228.802 nm is more stable under lower aluminum concentrations, which is an expected behavior.

Figure 9 displays lithium's emission line 670.780 nm analytical profile, also clear of aluminum spectral interferences. The other emission lines, 670.776 nm and 670.791 nm, also exhibit the same profile.
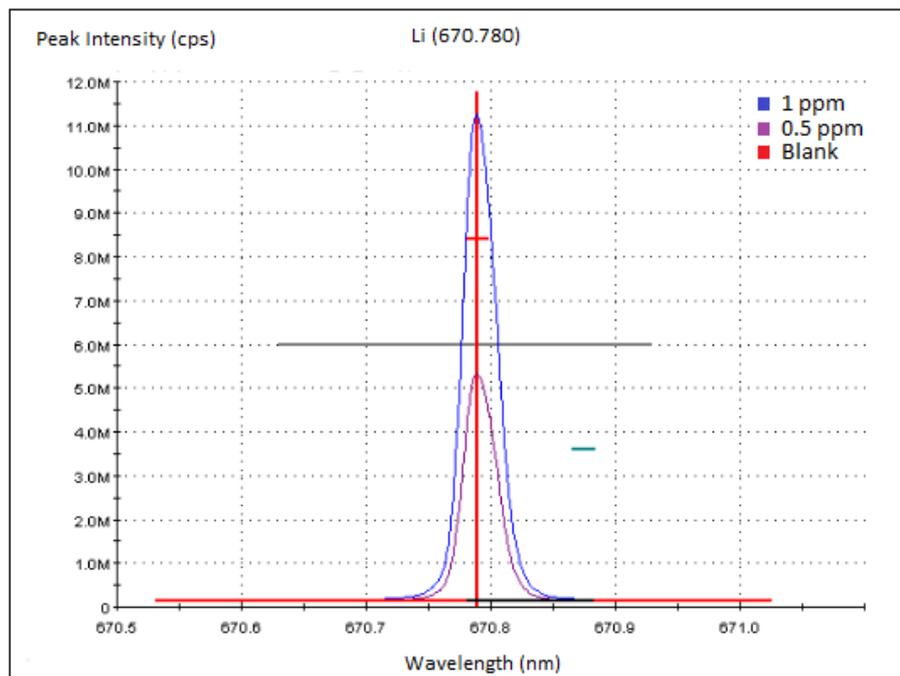

Figure 9. Analytical profile of lithium's emission line 670.780 nm.

None of lithium's emission lines show any apparent influence from the matrix and all lines exhibit the same response as the concentration of aluminum increases in the solution. The determination of lithium in such a low concentration of 0.5 mg L^{-1} in UAlx samples, where aluminum is present at a concentration of 8,000 mg L^{-1} is thus limited by other effects that will be further discussed (section 3.4).

Taking these results into consideration, it was possible to select emission lines for each element, as seen in Figure 10. For boron, the emission line that suffered the lowest deviation over the range of study is 249.677 nm. Cadmium's best line was 226.502 nm, but the line 228.802 nm, that also yielded a lower deviation to the expected result (0.5 mg L^{-1}), was selected because of its analytical profile. The latter also displayed a slightly better fit calibration compared to the first. The correlation coefficient for the line 228.802 nm is 1 while the one for the line 226.502 nm was 0.99998.
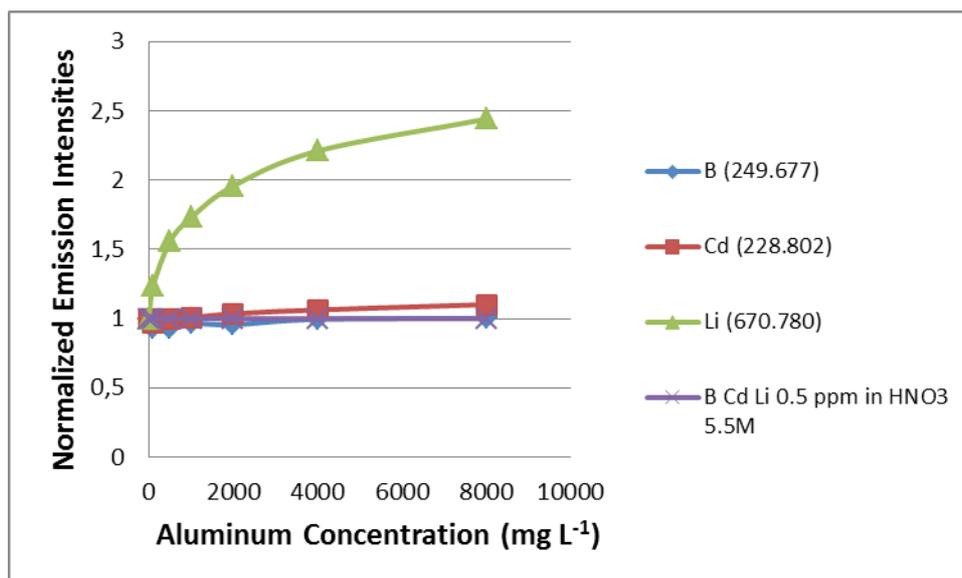
Figure 10. B, Cd and Li emission lines vs aluminum concentration

From Figure 10, it's clear that all elements suffer a positive effect as the concentration of aluminum increases. Lithium, however, suffers a much more pronounced effect. All three emission lines of lithium studied exhibit the same behavior, thus showing no distinguishable feature among them. All lines also show the same correlation in the analytical curve. This way, it was selected 670.780 nm as the emission line for purely comparison reasons. However, the deviations for this element cannot be neglected. It's shown that lithium cannot be determined at the studied concentration when aluminum is present in a concentration above 100 mg L^{-1}.

3.3 Simple analysis of a UAlx sample

Table 4 contains the results obtained for B, Cd and Li in an UAlx sample provided by CCN (IPEN/CNEN-SP) . The UAlx sample was prepared as described in section 2.1, previously removing the uranium by extraction chromatrography. The eluted solution was further analysed by ICP OES. The results were obtained in triplicate. The wavelengths selected for each element of interest were: Cd 228.802 nm, B 249.773 nm, and Li 670,784 nm

Table 4: Concentrations in UAlx sample

Element	Concentration (mg L^{-1})
B	5.29
Cd	< LOD (0.003)
Li	0.25

According to these results, it would be reasonable to state that the specifications required for the production of Mo-99, using UAlx targets fabricated by CCN, are being met for both cadmium and lithium, but not boron, which concentration is over 1 mg L^{-1}. However, as seen in section 3.1, some effects induced by the aluminum in the matrix can significantly influence the analytical results, especially for lithium. Therefore, it's not possible to make statements of this nature, until these effects are fully elucidated.

Further studies are then required to allow trustworthy results. Moreover, it was also demonstrated that the line 249.677 nm is the most suitable line for boron and, thus, should be adopted from now on.

3.4 Hypothesis

Boron's emission line 249.677 nm is an atomic line and its ionization potential is 8.29 eV, whereas aluminum's ionization potential is 5.98 eV. As aluminum is present in the solution in increasingly higher concentrations, several orders of magnitude bigger than boron, the electron displacement favors boron's atomic emission [21-23].

Cadmium's ionization potential is 5.41 eV, whereas aluminum's is 5.98 eV. However, since the concentration of aluminum is much greater than the concentration of cadmium in this matrix, the electronic vapor pressure exerted by the aluminum suppresses the line 214.438 nm of cadmium. Since the other two emission lines (228.802 nm and 226.502 nm) are both very close to each other and present a very similar excitation potential, the influence on these lines isn't very significant, even the line 226.502 nm being an ionic line.

Lithium has an ionization potential of 5.39 eV. Compared to aluminum, lithium is favored to remain in ionic form. As the difference in potential is low and aluminum is present in a much greater concentration, aluminum increases the electron population in the vapor phase and, thus, ends up favoring atomic lithium lines by displacing the electronic balance [21-23]. This explains the observed effects and why it is so difficult to quantify lithium in a concentration of 0.5 mg L^{-1} in UAlx samples when aluminum is present in a relatively much higher concentration (8,000 mg L^{-1}) and spectral interferences are not an issue.

4. CONCLUSIONS

Extraction chromatography is a suitable technique to separate uranium from UAlx samples. However, the determination of key elements, such as lithium, may be compromised by the presence of aluminum in the sample. ICP OES is a valuable technique in the determination of several elements in a wide range of matrices, but its physical-chemical and spectral interferences may affect the analytical results. It was demonstrated that boron and cadmium's emission lines 182.641 nm, 214.438 nm, respectively, and all studied lithium's emission lines (670.791 nm, 670.780 nm and 670.776 nm) undergo, in different degrees, some type of interference regarding the high concentrations of aluminum in the solution. Some hypotheses have been proposed and are to be investigated in future studies, as well as the dimension of these interferences on the analytical results. Also, methods to partially or completely remove the aluminum from the solution in order to avoid significant interferences will be further investigated.

ACKNOWLEDGMENTS

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