Neutron activation analysis of archaeological artefacts using the conventional relative method: A realistic approach for analysis of large samples

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Resumen

Se describe un nuevo método para el análisis de cerámicos enteros de interés arqueológico, mediante INAA, empleando el método relativo convencional. La propuesta se basa en la preparación de réplicas del cerámico arqueológico original, con una composición química bien conocida. Esta réplica es usada como estándar de comparación y se irradia simultáneamente con la muestra (cerámico arqueológico original), en un haz de neutrones térmicos del reactor nuclear RP-10. Las concentraciones determinadas por la irradiación de las réplicas, en comparación con los obtenidos en el análisis de las alícuotas, muestran valores razonables; en particular, el porcentaje de recuperación de lantano y escandio, con diferencias ≤ 10% con respecto a la referencia. La principal ventaja del método propuesto es evitar la necesidad de efectuar complicadas e inciertas correcciones matemáticas, debido a efectos no deseados, tales como auto-blindaje neutrónico, auto-termalización y atenuación de rayos gamma. Adicionalmente y en contraste con los otros métodos, la propuesta ofrece las ventajas de la factibilidad de evaluación de la incertidumbre de los resultados y fundamentalmente, validación completa del método.

Abstract

A new approach for analysis of entire potsherds of archaeological interest by INAA, using the conventional relative method, is described. The analytical method proposed involves, primarily, the preparation of replicates of the original archaeological pottery, with well known chemical composition (standard), destined to be irradiated simultaneously, in a well thermalized external neutron beam of the RP-10 reactor, with the original object (sample). The concentrations determined by irradiation of the entire replicates, in comparison with those obtained from the analysis of the aliquots, show a reasonable agreement; in particular, the percentage recoveries for lanthanum and scandium, with differences ≤ 10 % with respect to the reference. The basic advantage of this proposal is to avoid the need of performing complicated effect corrections when dealing with large samples, due to neutron self shielding, neutron self-thermalization and gamma ray attenuation. In addition, and in contrast with the other methods, the main advantages are the possibility of evaluating the uncertainty of the results and, fundamentally, validating the overall methodology.

1. Introduction

Neutron activation analysis (NAA) has proven to be a powerful quantitative analytical technique in many branches of the sciences [1-4]. One of the major applications that specially fit to the technique is in the field of archaeology. Since 1957 [5] NAA is currently used, because of its particular advantages, such as the possibility to analyze small quantities of sample and to perform with good accuracy multielemental determinations, including rare earths elements, by non-destructive methods. The results obtained can be used by archaeologists in provenance research and

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other connected aspects: e.g. to understand socio-economic aspects of the cultures studied, to verify authenticity of trade objects and to identify frauds or objects of illicit trafficking. In the world of museums this knowledge is also necessary to prepare and carry out appropriate restoration of damaged art objects [6].

The traditional way to perform the analysis has been limited to extract small portions of the ceramic pottery or fragment to be analyzed, to obtain quantitative elemental information. However, this sub-sampling could affect the integrity of the archeological pottery, which in many cases must be preserved intact because of its cultural heritage value. This premise demands the analysis of the entire object, the mass of which could be in the range of a few grams to kilograms.

X-ray fluorescence has been applied in non-destructive way to large archaeological pottery samples [7]; the information that can be collected is restricted to small portions of the surface layers of the artifacts.

Because of all the factors involved in the irradiation and measurement of large samples (in addition, having irregular shape) the accomplishment of instrumental neutron activation analysis of such objects is not trivial, taking into account the basic implicit requirements of reliability, as well as adequate accuracy and precision. The internal mono-standard NAA [8] has been employed to analyze pieces of pottery in the range of 10 – 50 g. A more general approach, concerning large samples, is the use of several specific mathematical corrections, which consider not only the characteristics of the samples, but also those of the reactor neutron spectrum and the detector. This possibility is readily applicable with the nowadays available computational facilities. However, the validation of the overall analytical method becomes uncertain, if not impossible.

The routine INAA technique applied to pottery analysis and carried out in the authors' laboratory considers irradiation of samples where no more than 300 mg that are conditioned in irradiation vials to be irradiated in a pneumatic rabbit system and then analyzed using the k_0-based INAA method [9]. Nevertheless, other irradiation facilities such as five radial conducts, which provide lower neutron flux, are available at the 10 MW research reactor (RP-10 reactor) of Instituto Peruano de Energía Nuclear (IPEN). This fact opened the consideration of a new approach: to analyze the entire potsherd by INAA (ES-INAA) using the conventional relative method. The analytical method proposed involves, primarily, the preparation of replicates of the original archaeological pottery, with well known chemical composition (standards), destined to be irradiated simultaneously (using a well thermalized external neutron beam of the reactor) with the original object (samples). The basic advantage of this proposal is to avoid the need of performing complicated effect corrections when dealing with large samples, due to neutron self-shielding, neutron self-thermalization and gamma ray attenuation [10-13]. In addition, and in contrast with the other methods, the main advantages are the possibility of evaluating the uncertainty of the results and, fundamentally, validating the overall methodology.

Peru has a vast cultural heritage around its more than one million km^2 of surface. Among the cultures developed in the valley of the north coast of the country, Mochica is one of the most relevant. Gods, men, animals and many scenes of its life [14] are represents in the works of Mochica ceramists, which are considered among the best in ancient Peru. The characteristics of the proposed method, as well as the results obtained in the analysis of replicates of a Mochica pottery, are presented in this work.

2. Experimental

Sample preparation: Twelve replicates of Mochica pottery (Figure 1, A-C) were prepared by a local artisan using a fine homogeneous paste. The average mass was 733,6 g, the minimum and maximum being 696,1 g and 804.1 g. After weighing, all samples were superficially cleaned with a soft brush and then put it in labelled polyethylene bags. The replicates were ordered in pairs, according to their masses; three pairs with very similar masses (difference 0.1%) and other three with mass difference up to 15.5%, (Table 1) were irradiated, one of the replicates of each pair acting as standard and the other one as comparator.

Multielemental determination in aliquots of the replicates: The methodology employed for the determinations, based on the k_0 method, has been described in [9].

Powder ceramic was extracted with a high purity carbide drill from different parts of each replicate, and mixed to form composites with masses of about 1 g (Figure 1, B - D) which were placed in clean glass containers, homogenized, dried at 110 ºC for 18 h in a calibrated oven, and stored, (Figure 1E).

Masses of about 250 mg were weighed in polyethylene vials and irradiated for 20 min., together with sodium comparators, using the pneumatic transfer system of the 10 RP-10 reactor, at a thermal flux of 1.9×10^{13} $n.cm^{-2}s^{-1}$ and epithermal flux per lnE of $3,2 \times 10^{11}$ $n.cm^{-2}s^{-1}$.

After a decay time of 5 days, a first measurement of the samples was performed for 5000 – 6000 s, using a Canberra GC 1518 HPGe detector (relative efficiency: 15%; FWHM: 1.8 keV at 1332.5 keV ^{60}Co). The comparators were measured for 1000 seconds after six days of decay in an identical geometry. Under these conditions, ^{76}As, ^{42}K, ^{140}La, ^{24}Na, ^{239}Np, ^{122}Sb, ^{153}Sm, and ^{175}Yb were determined. A second measurement was carried out after 15 – 17 d decay for 10000 seconds with another HPGe detector (Canberra GC 7019; relative efficiency: 70%, FWHM: 1.9 keV at 1332.5 keV ^{60}Co). The nuclides determined were ^{131}Ba, ^{141}Ce, ^{60}Co, ^{51}Cr, ^{134}Cs, ^{152}Eu, ^{59}Fe, ^{181}Hf, ^{177}Lu, ^{147}Nd, ^{86}Rb, ^{46}Sc, ^{85}Sr, ^{182}Ta, ^{160}Tb, and ^{233}Th. A certified standard reference material, NIST 1648, was used for quality control of the procedure.

Analysis of the entire objects: The pairs of the prepared replicates were placed onto an aluminum double turntable device of 46x23x71 cm and irradiated at the head of the radial tube number 3 of the RP-10 reactor (Figure 2). This radial tube is specially set up for the application of neutron radiography technique. It is formed by an internal aluminum collimator, covered with a cadmium sheet and located in the reactor pool. In front of the reactor core a lead shielding of 3 cm thickness, reduces the gamma radiation exposition to the exterior. The external collimator consists of two stainless steel tubes of 18 cm and 23 cm, respectively. The neutron flux dispersed by the stainless steel tubes is decreased by inner cadmium tubes covered with aluminum [15]. For each pair of samples, the replicate of higher mass was used as the comparator; it was mounted on the right side of the turntable device, in front of the head of the radial tube. Small disks of scandium and gold were stuck in different parts of both the samples and the comparators, as flux monitors (Table 1).

Each pair was irradiated by 7 - 13 hours, and measured for 100 000 – 200 000 s, after 3 and 10 d decay, using a 70% relative efficiency HPGe detector. The scandium and gold monitors were counted for about 20000 s and 5000 s, after about 10 d and 5 d, respectively from the end of irradiation.

Eventual differences in size between the replicates were assessed using a special, computer-controlled turntable-scanner device, which allows taking calibrated pictures of each replicate at different angles (Figure 3A-B). The projected areas, perimeters and Feret diameters were used as quantitative parameters of size (Figure 3C-D). Image processing was performed using the software ImageJ [16]. For every pair of replicates, the ratio of averaged size parameters was used as an approximated correction factor for the observed activities.

3. Results and Discussion

The average neutron flux of the collimated thermal neutron beam was 3.9×10^{10} $n.m.^{-2}s^{-1}$ at RP-10 reactor. The theoretical calculations demonstrated that various elements could be determined with reasonable precision.

The results of the analysis of the composites obtained from the potsherds are shown in Table 3. Although concentrations of around 25 elements were originally measured, only those determined in the entire pottery were kept. Low coefficients of variation for arsenic, europium, iron, potassium, lanthanum, scandium and samarium, and slightly higher for cobalt and antimony, are registered, thus showing the high degree of homogeneity in the batch of clay used to manufacture the objects. On the other hand, sodium and caesium, recognized as elements of high mobility, show a great dispersion in concentration; thus, their usefulness is questionable.

The results obtained for eleven elements in large sample replicates (LSR) are shown in Table 4. As it was already described, they correspond to paired irradiations, where one of the samples acted as comparator. In order to evaluate the trueness of this procedure, the concentrations obtained were compared with those resulted from the analysis of the composites. The relative percent difference of both results was defined as the recovery percentage of each element in the samples, and is also shown in Table 4 and graphically represented in Figure 4.

If a recovery percentage within ≤ 20 % difference with respect to the reference (100) is considered acceptable to evaluate the results, Co (78%) in LSR-2 and Fe (70%) in LSR-16 show poor recovery, while over estimated recovery is registered for Sb (128%) in LSR-16, Na (121%) in LSR-5, Cs (124%) in LSR-9 and Sm (124%) in LSR-3. The remaining elements present recoveries within the acceptance limits in all samples, the best ones corresponding to La and Sc (recovery percentage within ≤ 10 %). As it is well known, the relationships between these elements are a powerful tool for archaeological studies.

The low recovery results for Co and Fe can be explained as a consequence of the poor counting statistics for these elements, which are near the quantification limit. The inhomogeneous distribution of sodium and cesium in the ceramic artifact can be the cause of its anomalous recovery in some of the samples. Finally, two radionuclides used for the determination of the corresponding elements with overestimated recovery present possible spectral interferences: these are the cases of ^{122}Sb and ^{153}Sm, interfered by, respectively, ^{76}As and ^{239}Np. The option of determining antimony by measuring of the 1691 keV from ^{124}Sb, can not be applied, because of the poor statistics of the counting, at the conditions used.

4. Conclusions

As it is demonstrated by the experimental results, the proposed method opens a very promising field for the use of non-destructive neutron activation analysis of potsherd of archaeological interest, no matter the size of the samples and the irregularities in their shape, thus adding a new advantage to a technique unique in many aspects.

The replicates prepared by the local artisans join the conditions required to successfully accomplish the method.

The concentrations determined by irradiation of the entire replicates, in comparison with those obtained from the analysis of the aliquots, show a reasonable agreement. In particular, the percentage recoveries for lanthanum and scandium, with differences ≤ 10 % with respect to the reference, are the most notable cases and reinforce the conclusions about the possibilities of the method.

Depending on the possibilities of each institution, some significant improvements are the use of beams with higher fluxes and, in connection with the equipment, of Compton suppressors, as well as low background detectors of at least 150% relative efficiency.

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6. References

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Table 1. Experimental information about samples, monitors and irradiations.

Number of pair	Sample code	Irradiation time (H)	Mass of samples (g)	Monitor / Mass (mg)
1	LSR-2	8	725.89	Sc / 12.5
	LCR-14		727.08	Sc / 12.3
2	LSR-16	13.42	724.28	Sc / 24.31
	LCR-8		723.17	Sc / 25.10
3	LSR-5	7	722.19	Sc / 44.82 y Au / 8.86
	LCR-12		723.40	Sc / 47.04 y Au / 8.83
4	LSR-11	7.5	738.51	Au / 8.84
	LCR-4		760.24	Au / 8.67
5	LSR-9	6.83	712.23	Au / 8.79
	LCR-13		764.98	Au / 8.82
6	LSR-3	8.67	696.05	Au / 8.86
	LCR-6		804.09	Au / 8.74

LSR: Large sample replicate; LCR: Large comparator replicate.

Table 2. Illustrative values for a pair of replicates, imaged at an azimuth tilting angle of 0°. All values are indicated in pixels.

ID	Area	Perimeter	Feret D	Feret X	Feret Y
M1	194297	254	72	337	169
M2	194712	257	72	331	170

Table 3. Concentrations (mg /kg) found in aliquots (A) of the composites. Uncertainties expressed at 95% confidence interval.

Nº	**As**	**Co**	**Cs**	**Eu**	**Fe**	**K**	**La**	**Na**	**Sb**	**Sc**	**Sm**
A-2	60.5 ± 5.5	18.8 ± 1.3	48.8 ± 4.8	1.00 ± 0.08	36335 ± 2470	25234 ± 3150	30.8 ± 2.2	17240 ± 1200	6.3 ± 1.0	13.6 ± 0.90	4.80 ± 0.30
A-14	55.8 ± 5.1	16.90 ± 0.94	52.0 ± 5.1	1.00 ± 0.08	37322 ± 2540	28838 ± 3600	31.0 ± 2.2	14892 ± 1010	6.1 ± 0.90	14.1 ± 1.0	4.84 ± 0.30
A-8	53.7 ± 4.8	17.5 ± 1.2	58.9 ± 5.8	1.10 ± 0.07	37627 ± 2560	28257 ± 3500	32.0 ± 2.3	12789 ± 890	5.9 ± 1.4	14.2 ± 1.0	5.20 ± 0.40
A-16	55.2 ± 5.0	17.3 ± 1.2	60.4 ± 5.9	1.10 ± 0.07	37579 ± 2555	28719 ± 3560	32.4 ± 2.3	13310 ± 920	5.3 ± 1.6	14.2 ± 1.0	5.21 ± 0.36
A-5	58.7 ± 5.3	19.6 ± 1.3	46.7 ± 4.6	1.00 ± 0.07	37785 ± 2570	25290 ± 3140	31.9 ± 2.2	13310 ± 920	5.9 ± 1.4	14.0 ± 1.0	5.10 ± 0.35
A-12	53.2 ± 4.8	17.3 ± 1.2	39.7 ± 5.2	1.10 ± 0.07	37695 ± 2560	28562 ± 3540	32.0 ± 2.2	14343 ± 990	5.9 ± 1.4	14.2 ± 1.0	5.10 ± 0.35
A-11	50.6 ± 4.6	17.1 ± 1.2	45.4 ± 4.4	1.10 ± 0.08	36232 ± 2460	28430 ± 3525	29.9 ± 2.1	14958 ± 1040	5.2 ± 1.2	13.9 ± 1.0	5.00 ± 0.30
A-4	51.7 ± 4.7	19.8 ± 1.3	55.0 ± 5.4	1.10 ± 0.08	37337 ± 2540	27936 ± 3460	30.6 ± 2.1	12731 ± 880	5.2 ± 1.2	14.2 ± 1.0	5.13 ± 0.30
A-9	53.8 ± 4.8	16.3 ± 1.1	67.4 ± 6.6	1.20 ± 0.08	38274 ± 2600	26916 ± 33340	31.9 ± 2.2	11204 ± 780	5.8 ± 1.4	14.7 ± 1.0	5.28 ± 0.40
A-13	52.0 ± 4.7	16.6 ± 1.1	69.1 ± 6.8	1.10 ± 0.08	37689 ± 2560	27027 ± 3350	31.4 ± 2.2	10744 ± 750	5.7 ± 1.4	14.4 ± 1.0	5.20 ± 0.35
A-3	53.0 ± 4.8	17.0 ± 1.2	50.1 ± 4.9	1.10 ± 0.08	37756 ± 2570	28033 ± 33480	30.7 ± 2.1	14148 ± 980	5.3 ± 1.3	14.5 ± 1.0	5.20 ± 0.35
A-6	49.9 ± 4.5	15.7 ± 1.1	69.4 ± 6.8	1.10 ± 0.08	36889 ± 2510	26595 ± 3300	31.3 ± 2.2	9627 ± 670	5.2 ± 1.3	14.2 ± 1.0	5.10 ± 0.35
Aver.	*54.2*	*17.5*	*56.5*	*1.09*	*37276*	*27902*	*31.3*	*13312*	*5.7*	*14.1*	*5.1*
CV, %	*5.4*	*7.4*	*14.2*	*6.0*	*1.7*	*4.8*	*2.4*	*14.4*	*7.2*	*1.9*	*2.7*

Table 4. Concentrations results (mg/kg) obtained for large sample replicates (LSR) paired irradiations, where one of the samples acted as comparator. Uncertainties expressed at 95 % confidence interval.

Element	**As**	**Co**	**Cs**	**Eu**	**Fe**	**K**	**La**	**Na**	**Sb**	**Sc**	**Sm**
LSR-2	54.0	14.6	49.7	1.0	36470	28600	28.0	14570	6.0	13.3	4.6
Uncertainty	2.4	4.0	2.6	0.05	5130	3130	2.0	11	1.0	1.0	1.0
% Rec. LSR-2	*89*	*78*	*102*	*100*	*100*	*113*	*91*	*84*	*95*	*99*	*96*
LSR-16	56.2	20.0	72.0	1.2	26490	26725	32.9	12580	6.9	15.5	4.6
Uncertainty	8.0	2.5	2.8	0.20	13900	4230	2.6	120	1.1	0.50	1.0
% Rec. LSR-16	*102*	*116*	*119*	*114*	*70*	*93*	*101*	*94*	*128*	*110*	*88*
LSR-5	60.3	18.6	42.0	1.0	31500	24900	32.0	16100	6.8	13.0	4.9
Uncertainty	5.0	3.6	3.0	0.05	2580	10780	4.3	240	1.0	0.40	1.2
% Rec. LSR-5	*103*	*95*	*90*	*100*	*83*	*98*	*100*	*121*	*115*	*93*	*83*
LSR-11	58.8	17.8	56.0	1.0	29830	32290	32.0	14580	6.1	14.0	5.9
Uncertainty	5.5	6.3	3.6	0.1	6550	2210	1.5	73	0.60	0.70	0.50
% Rec. LSR-11	*116*	*104*	*120*	*91*	*82*	*113*	*107*	*97*	*115*	*101*	*118*
LSR-9	52.7	16.6	71.0	1.0	33060	26110	30.2	10554	6.0	14.0	4.8
Uncertainty	4.3	4.1	4.0	0.20	6110	1700	2.0	60	0.6	0.5	0.3
% Rec. LSR-9	*98*	*102*	*105*	*83*	*83*	*97*	*94*	*94*	*102*	*95*	*90*
LSR-3	56.0	15.0	62.0	1.2	37700	28195	34.0	10800	6.0	14.6	6.4
Uncertainty	2.5	2.0	4.4	0.25	5030	580	1.0	27	0.40	0.60	0.30
% Rec. LSR-3	*106*	*99.9*	*124*	*109*	*99.8*	*100*	*110*	*76*	*113*	*101*	*124*

% Rec.: percentage recovery (see explanation in the text).

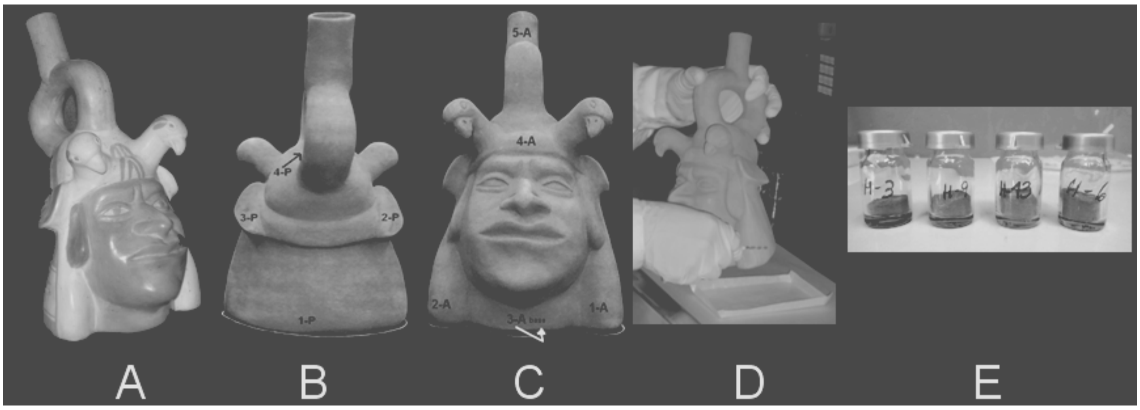

Figure 1. (A) Original Mochica pottery. (B), (C) Sections of powder pottery extraction, (D) Process of powder extraction, (E) Small bottles with sample, ready to analysis.

Figure 2. The irradiation facility, loaded with two replicates.

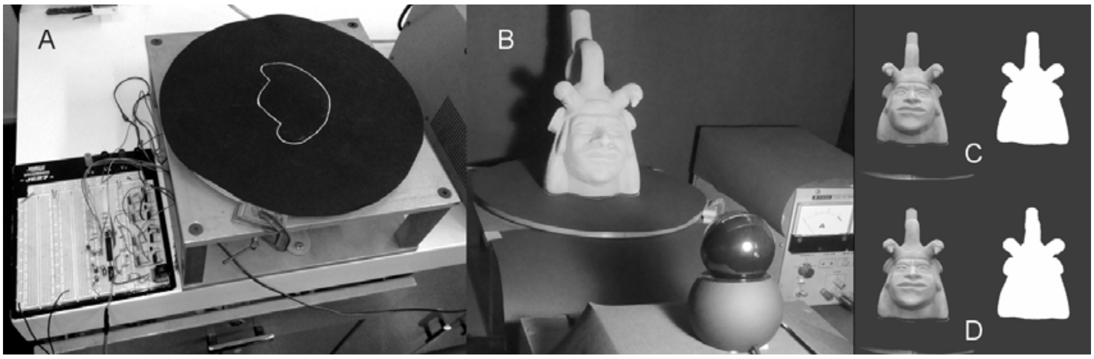

Figure 3. Partial views of the computer controlled turntable scanner device, empty (A) and loaded (B), calibrated images and corresponding projected sizes, at an azimuth tilting angle of 0°, for a pair of studied replicates (C-D).

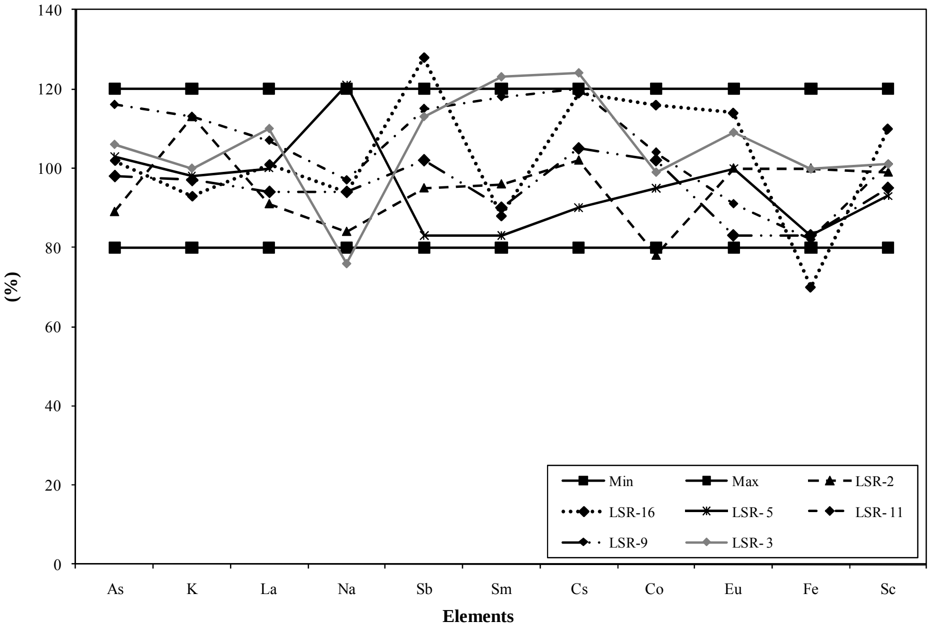

Figure 4. Percentage recoveries of the elements in the samples.