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Advances in Applied Science Research, 2013, 4(1):158-162

ISSN: 0976-8610
CODEN (USA): AASRFC

Preliminary studies on the performance of Ghana's MNSR neutron fluxes in reactor neutron activation analysis after the addition of a beryllium shim

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ABSTRACT

An accurate knowledge of the neutron fluxes in MNSRs core after any reconfiguration is of paramount importance to achieving reliable results in RNAA regardless of the methodology used for determining the elemental concentrations. A plethora of reliable analytical results can therefore be exclusively linked to a fruitful assessment of the stability, reproducibility and the extent of thermalization of the neutron fluxes for RNAA. Hence, two different matrix standards NIST SRM 1571, Orchard leaves representing a biological standard and NIST SRM 1632, Coal representing an environmental standard, were analyzed in order to test the performance of GHARR – 1 neutron fluxes after configurational changes to the core in the single comparator mode. The precision and trueness of the results were generally found to be within 6% at 99% confidence interval of certified values - confirming the suitability of GHARR – 1 neutron fluxes in RNAA after core changes.

Keywords: Single comparator method; nuclear research reactors; flux parameters; GHARR – 1; Activation Analysis.

INTRODUCTION

It is well documented that reactor neutron activation analysis (RNAA) which occupies a prominent position among the various analytical methods, is a nondestructive, exceedingly selective, flexible and simultaneous multielement analytical technique for precise elemental analysis [1, 2, 3]. The analytical advantages of this powerful technique are also well discussed elsewhere [4, 5, 6]. Although there are several types of neutron sources (Ra – Be mixtures, accelerators, radioisotopic neutron emitters etc.) that one can use for this analysis, nuclear research reactors with their high fluxes of neutrons from uranium fission offer the highest available sensitivities for most elements [7].

One of such neutron research reactors is the Miniature Neutron Source Reactor (MNSR) that is extensively designed as an excellent source of neutrons for RNAA. MNSRs are classified as low power reactors [8] even though, they have a very high power density in the core primarily due to their very low thermal power output of about 30 kW at maximum nominal operations compared to nuclear power plants (power reactors). The guiding principles for the design of these reactors are to produce long-term reproducible, highly stable, and well thermalized neutron flux with a large flux to power ratio for analytical and other purposes. Thermalization in the extremely compacted "tank – in – pool" MNSR core is effectively attained by surrounding it with annulus beryllium reflectors. These reactors also have bottom beryllium plates and sets of top beryllium shims [9]. The design considerations, technical specifications and inherent safety features of Ghana's MNSR known as Ghana research reactor – 1 (GHARR – 1) is also available in literature [10, 11, 12].

Even though MNSRs are noted to possess long – term stable and reproducible neutron fluxes, nevertheless, due to the small size of the core which aids neutron leaking, loss of reactivity from fuel burn up, accumulation of fission products and various poisons buildup, the neutron fluxes in the core reduces over time (aging lifetime of the core) [11]. For instance the excess reactivity of the Pakistan research reactor -2 (PARR -2) was reduced from 4 *mk* to approximately 2.96 *mk* after 10 years of operation [13]. Similarly, GHARR -1 core was reconfigured after over 10 years of operation [14]. Therefore, regardless of the methodology (absolute, relative or single comparator method) used for determining the elemental concentrations in RNAA, the reduction of neutron fluxes samples receive directly affect the accuracy of the results [15]. Again, with the current international interest in converting Highly Enriched Uranium (HEU) fuels of MNSRs to Low Enriched Uranium (LEU) fuels, efforts aimed at increasing the thermal neutron fluxes of LEU fuels are vigorously being pursued. Hence monitoring of neutron fluxes in MNSRs is of utmost importance in neutron dosimetry, fast neutron physics, radiation damage, production of radioisotopes and achieving accurate results in RNAA.

For these reasons, preliminary studies on the performance of Ghana's MNSR neutron fluxes in reactor neutron activation analysis after the addition of a beryllium shim to the top aluminium tray of the core is justified. The main aim of this work therefore, was to experimentally assess the performance of Ghana's MNSR neutron fluxes in RNAA after configurational changes in the core. The neutron fluxes and spectrum shaping factors of this reactor have previously been done [16]. Hence, another aim was to validate these parameters for routine RNAA in the single comparator mode. Also, the precision and accuracy of the analytical method after the addition of the beryllium shim was reassessed.

MATERIALS AND METHODS

About 200 *mg* of two accurately weighed matrix multielement standard reference materials (NIST SRM 1571, Orchard Leaves and NIST SRM 1632, coal) both obtained from the National Institute of Standards and Technology (NIST), USA, were wrapped in clean polyethylene films and encapsulated in pre – clean 7 mL polyethylene vials obtained from Olympic Plastic Company, USA. The ultimate comparator, gold solution, was transferred with calibrated eppendorf tip ejector pipette obtained from Brinkmann Instruments, Inc. Westbury, New York, into a pre – clean and pre- weighed 1.5 mL polyethylene vials to obtain a mass of about 20 µg. All the polyethylene vials were pre – cleaned by thoroughly washing with distilled water before soaking them in 1:4 reagent grade HCl for a day. The vials were then rinsed with distilled de-ionized water and then air – dried in fumehood. Quadruplicates of each standard were similarly prepared for irradiation. All vials were stack with cotton and heat-sealed for irradiation.

All standards were irradiated in the inner pneumatic irradiation site 2 of GHARR – 1 facility with the reactor operating at half thermal power, 15 kW. Irradiation schemes were chosen depending on the half – lives of the elements of interest present. The induced activities of the standards after appropriate decay periods were ascertained using a PC – based gamma – ray spectroscopy system. A detailed description of this system has previously been presented by Osae et al. [17] and Adotey et al. [18]. The activities of the standards were ascertained at a constant detector – to – standard distance of 7.2 cm by mounting a plexiglass support system at the top of the detector. As a way of improving the counting statistics, care was taken to reduce the pile-ups and random coincidence losses by keeping the dead time around 10%. To ensure quick and accurate spectral analysis without personal errors, the spectral data were collected and then loaded into a Hypermet – PC version 5.12 program for automated analysis [19]. The detection efficiency for the γ– ray spectrometer was calibrated with an IAEA mixed standard radionuclide solution containing ^{60}Co, ^{241}Am, ^{109}Cd, ^{54}Mn, ^{65}Zn, ^{85}Sr, ^{203}Hg and ^{57}Co. The measured detection efficiencies were fitted by a polynomial function [16, 20].

RESULTS AND DISCUSSION

The results of mean elemental concentrations of two types of standard reference materials (NIST SRM 1571, Orchard Leaf - a biological standard and NIST SRM 1632, Coal – an environmental standard) analyzed in the single comparator mode based on Hogdahl convention [21] are presented in Table 1 and Table 2 respectively. The neutron flux parameters (thermal to epithermal neutron flux ratio and the neutron shaping factor) were previously determined by Sogbadji et al [16]. Other parameters used in this quantification method are presented elsewhere [5, 7, 22, 23, 24]. The uncertainties associated with the mean measured concentrations were ascertained as expanded uncertainties ($k = 2$).

The addition of beryllium shims to the top aluminium tray of MNSRs core is grounded in reactor design understanding that, these shims have very good scattering but poor absorption cross section abilities for thermal neutrons – an outstanding feature of a reflector material. In the realm of neutron physics, this addition largely affects the degree of neutron leakage, the reactivity worth of the core for sustaining a chain reaction, the extent of

thermalization of fast neutron produced from fission and the overall neutron flux levels of the reactor. Therefore, the performance assessment of the suitability of GHARR – 1 neutron flux spectra in reactor neutron activation analysis after configuration changes [25] to the core for routine analysis cannot be over – emphasized.

Table 1. Analysis of NIST 1571, Orchard Leaf in mg/kg unless stated otherwise by RNAA.

Element	This work				Reported Values
	Measured [a]	RSD[b](%)	Error[c] (%)	Z – Scores	
As	10.53 ± 0.63	5.98	-1.59	-2.84	10.7 ± 2.5
Br	9.91 ± 0.56	5.65	1.12	1.95	9.8 ± 0.9
Ca (%)	1.78 ± 0.09	5.06	-6.32	-2.37	1.9 ± 0.02
Cu	12.99 ± 0.74	5.70	-0.84	-1.93	13.1 ± 2.2
K (%)	1.53 ± 0.08	5.23	2.00	0.57	1.5 ± 0.02
Mg (%)	0.53 ± 0.02	3.77	1.92	0.27	0.52 ± 0.02
Mn	90.38 ± 5.85	6.47	-0.13	-1.85	90.5 ± 3.5
Na	83.11 ± 4.98	5.99	0.13	1.84	83 ± 5
Rb	11.61 ± 0.81	6.98	0.96	1.58	11.5 ± 0.9
Sb	3.29 ± 0.03	0.91	-0.30	-1.10	3.3 ± 0.2
Sc	36.00 ± 2.43	6.75	0.56	2.96	35.8 ± 1.5
Zn	26.05 ± 1.41	5.41	0.19	0.92	26 ± 3.2

[a] Mean measured elemental concentrations, [b] Relative standard deviation, [c] Relative error

Table 2. Analysis of NIST 1632, Coal in mg/kg unless stated otherwise by RNAA.

Element	This work				Certified Values
	Measured [a]	RSD[b](%)	Error[c] (%)	Z – Scores	
Al (%)	0.954 ± 0.0172	1.80	4.26	2.16	0.915 ± 0.0137
As	6.07 ± 0.34	5.60	-1.78	-1.96	6.18 ± 0.27
Br	18.63 ± 0.61	3.27	-0.37	-2.14	18.7 ± 0.4
Ca (%)	0.139 ± 0.004	2.88	-4.14	-0.21	0.145 ± 0.03
Cd	0.076 ± 0.005	6.58	5.56	0.06	0.072 ± 0.007
Cl (%)	0.11 ± 0.0054	4.91	-3.42	-0.08	0.1139 ± 0.0041
Co	3.51 ± 0.21	5.98	0.86	0.50	3.48 ± 0.20
Cu	6.13 ± 0.43	5.55	2.00	2.16	6.01 ± 0.25
Fe (%)	0.765 ± 0.019	2.48	4.08	1.21	0.735 ± 0.011
K	0.105 ± 0.0041	3.92	-4.55	-0.13	0.11 ± 0.0033
Mg (%)	0.0406 ± 0.002	4.93	5.73	0.04	0.0384 ± 0.0032
Mn	12.98 ± 0.41	3.16	-0.46	-1.90	13.04 ± 0.53
Na	298.75 ± 5.2	1.74	-0.02	-2.87	298.8 ± 4.8
Ni	9.51 ± 0.61	6.41	2.04	2.96	9.32 ± 0.51
S (%)	1.53 ± 0.078	2.7	7.4	1.33	1.462 ± 0.051
Si	1.582 ± 0.04	2.53	-4.35	-2.85	1.654 ± 0.034
V	23.79 ± 0.60	2.52	0.30	2.78	23.72 ± 0.51
Zn	12.25 ± 0.76	6.20	1.24	2.42	12.1 ± 1.3

[a] Mean measured elemental concentrations, [b] Relative standard deviation, [c] Relative error

For these reasons, the stability and reproducibility of the neutron fluxes of the GHARR – 1 and the accuracy of the single comparator analytical method were re – evaluated for internal quality control. The stability and reproducibility of the neutron fluxes were calculated as the percentage relative standard deviations (%RSD) for both standards. A least value of 0.91% in Sb to 6.98% in Rb was recorded in NIST SRM 1571, Orchard leaf. Similarly, these values were 1.74% in Na to 6.58% in Cd for NIST SRM 1632, Coal. Thus it is evident that, most of the values were within 6% suggesting high order of precision of our data. Thus, confirming one of the hallmarks of MNSRs neutron fluxes for activation analysis [26]. Also, the accuracy of the analytical method was also calculated as the relative errors for both standards. A critical examination of Table 1 and Table 2 shows that except for Ca (- 6.32) in NIST SRM 1571, Orchard leaf and S (7.4) in NIST SRM 1632, Coal, all elements were within ± 6% confirming the accuracy of the analytical method used in the calculations.

However, Serfor – Armah et al. [27] found the precision and accuracy of GHARR – 1 in RNAA using NIST SRM 1571, Orchard leaf in relative comparator mode to be within ± 10%. But these values were assessed by Appenteng et al. [28] using the same standard and method to be within ± 5%. This discrepancy can largely be explained by the configurational changes in the core of GHARR – 1 that probably led to better thermalization of neutrons for much more precise analysis. Thus, our results were in good agreement with the latter.

Notwithstanding, in order to elucidate the confidence levels of the mean measured elemental concentrations, Z – scores which was previously developed for comparison of interlaboratory results were calculated as shown in Table 1 and Table 2.

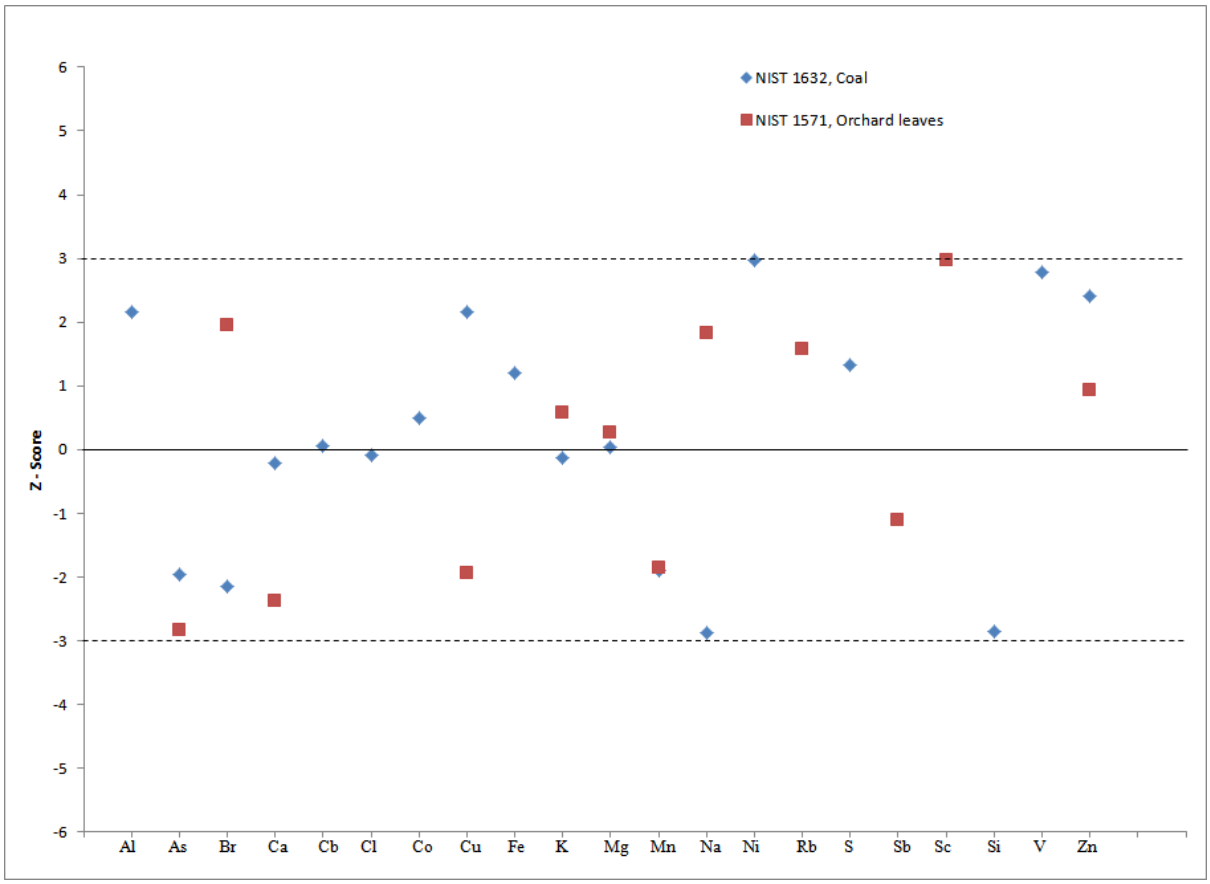

Fig. 1. Z – Score values obtained for the elements determined in NIST reference materials

Pictorially, most of the elements determined were dotted away from the zero mark of the Z score axis as shown in Fig. 1. This phenomenon can partially be attributed to the neglect of correction factors such as neutron self-shielding factors, gamma – ray attenuation in the standards and coincidence summing effects, that might have cumulatively led to questionable ($2 \leq |Z| < 3$) results for some of the elements. Nevertheless, all the elements analyzed had Z – scores values within $|Z| < 3$, which means that the results obtained are in the 99% confidence interval of the certified or reported values [29].

CONCLUSION

The performance assessment of Ghana's miniature neutron source reactor neutron fluxes in reactor neutron activation analysis was found to be excellent with a general slight deviation of about 6% at a 99% confidence interval of certified or reported values. We therefore wish to categorically state that, even though the core of Ghana's MNSR had been critical for nearly two decades, it is still an excellent source of neutrons for reactor neutron activation analysis if regular configuration changes in the core are made. The renaissance of different neutron flux parameters in the core after the addition of the beryllium shim led to a much more precise determination of elemental concentrations in the reference materials suggesting better performance of GHARR – 1 neutron fluxes. It is therefore expected that, these fluxes will greatly ameliorate the sensitivities of elements in samples.

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