

Estimation of trace element concentration and neutron induced radioactivity in rock samples of different geological compositions for neutron shielding

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Rock samples of different geological compositions intended for use as coarse aggregates (CA) in self-compacting concrete (SCC) have been analysed for concentrations of the elements Sc, Co, Fe, Cs, Ta, Eu. These elements give rise to long-lived radioactivity in concrete used as neutron shields, consequent to long-term neutron irradiation. This study has been performed on all the eleven samples of different geological compositions to compare their low-activation characteristics and determine their suitability for developing SCC based neutron shielding. The collected samples quartzite rock exhibits lowest induced activity on neutron irradiation. But it failed the fresh property tests for SCC as observed in an earlier study (Pai B H V, *et al.*) [communicated to Journal of The Institution of Engineers (India): Series A]. Among the CA samples suitable for SCC composition, dolomite rock produces lowest activity for all the isotopes studied. Concretes having low radioactivation properties will facilitate mitigating the prevailing radiological waste management problems during decommissioning of nuclear facilities.

Keywords: Elemental concentration, Coarse aggregate, Induced activity, Neutron activation analysis, Gamma spectrometry

1 Introduction

Compact, efficient and cost-effective shield design for nuclear reactors and accelerators has gained importance in the last few decades. In reactors and in positive ion accelerators prompt radiation field is dominated by neutrons. For the entire life-time of the facility, the bulk shield, mostly concrete, will be subjected to long term irradiation by neutrons. An estimate of induced radioactivity per unit mass of the bulk shield is important to plan decommissioning of the facility¹.

At the end of the year 2011, about 124 nuclear power reactors were shut down throughout the world². The structural decommissioning of these facilities would produce huge amount of low level radioactive waste, mostly concrete. Depending on the type of the reactor (e.g., water cooled/ gas cooled/research/power reactor) operating at hundreds of MWe, few hundred to thousands of tons of radioactive concrete wastes are produced³⁻⁵. These wastes often contain long-lived radioactivity. Masumoto *et al.*¹ showed that in an accelerator environment ¹⁵²Eu, ⁶⁰Co, ¹³⁴Cs, ²²Na, ⁵⁴Mn are some of the important isotopes produced in

concrete through neutron activation. For the accelerator facility studied in their work, an activity of 0.02 Bq/g of ⁶⁰Co resulted in a dose rate of 0.1 μ Sv/h. Calandrino *et al.*⁶ found that the measured dose from external exposure (from activated concrete shielding and other components) to the decommissioning staff of a compact self-shielded 11 MeV medical cyclotron was between 10-20 μ Sv.

Presently, in Europe the amount of recycled radioactive concrete waste² is between 1200-2000 tons/year and this is expected to rise to about 7,000 tons/year by the year 2030. The disposal of these materials would pose a major problem because the facilities and the final disposal technologies are not only limited but also expensive. For a medium energy, high current accelerator, the cost of decommissioning will far exceed the cost of the accelerator. Thus, the amount of radioactive waste that arises from operating and decommissioning activities is a serious impediment for the proliferation of nuclear technology. Waste recycling, volume and activity reduction are very essential in decreasing the costs of decommissioning nuclear facilities.

Radiological as well as physical and chemical characterisation of these waste materials are important, in order to segregate, process, and dispose them off. Mehta and Monterio⁷ have shown that aggregates are the major constituents of concrete (approx. 70%), which determines its strength and durability. Hence, generation of radioactivity in aggregates plays an important role in determining the activation potential of concrete. Self-compacting concrete (SCC) containing some chemical admixtures in addition to normal coarse and fine aggregates can preferably be used to construct radiation shields in nuclear installations because of the following advantages. SCC is able to flow under its own weight, completely filling formwork and achieving full compaction, even in the presence of congested reinforcement. The fluidity and segregation resistance concrete strength, providing the potential for a superior level of finish and durability to the structure. However, all the CA is not suitable for the preparation of a SCC mix.

Identification and selection of the appropriate CA that will be suitable for making SCC and at the same time will produce lesser amount of long lived neutron activation products can be achieved by quantifying trace elements present in the aggregates that are capable of generating long lived activation products. This can be done using neutron activation analysis (NAA) which can be used for trace element quantification down to ppb-ppm range of multiple components in different types of materials, including rock samples.

The primary objective of this work is to identify amongst different geological formations the appropriate coarse aggregates (CA) (stone chips used in preparation of concrete) that may be employed in the preparation of low radio-activation SCC for radiation shielding. In a previous work by the authors attempts have been made to identify CA suitable for preparation⁸ of SCC from the rock samples collected from eleven different geological compositions in India. In the present work, we estimate radio-activation of these eleven CA samples due to neutron irradiation and determine the relative concentration of six different elements which give rise to long-lived radioactivity. Relative concentration of an element in different CA samples will help to identify the CA samples which will generate low amount of induced radioactivity. These data will be useful for selecting appropriate CA for neutron shielding amongst different types of geological composition found globally.

2 Experimental Details

2.1 Materials and Methods

Coarse aggregate found in different areas vary in elemental composition, water content, porosity, etc. For optimization of shield design and minimizing residual radioactivity in different upcoming nuclear facilities and also to make them cost-effective, it is necessary to determine the concentration and activation of the various constituents of concrete. In the present work, NAA was carried out on coarse aggregate samples procured from eleven different geological formations in the state of Karnataka, India. These CA (rock) samples were collected to prepare SCC mixes which provide one of the most cost-effective solutions for shielding construction of reactor and accelerator facilities. A description of these samples is given in Table 1. The data of geological formations was obtained from the Geological Map, provided by the Department of Mines & Geology, Government of Karnataka⁹.

2.2 Neutron Activation Analysis of the CA samples

Neutron activation analysis (NAA) was carried out on the eleven CA samples collected. Nearly 50 mg of each of the eleven rock samples was wrapped separately with aluminium foil in a packet of dimension 1 cm × 1 cm × 0.1 cm. Two standard reference materials, IAEA-433 and IAEA-158, procured from International Atomic Energy Agency (IAEA), Vienna, Austria, were also wrapped in separate aluminium foils. These two packets of samples and reference materials were irradiated simultaneously (co-irradiated) for three and a half days in Dhruva research reactor at the Bhabha Atomic Research Centre, Trombay, India. The samples were kept in the sample holder cans inside the reactor calandria/ core assembly. The sample holder can is cylindrical in shape, having internal diameter 1.8 cm, height 4 cm standing upright during irradiation.

Table 1 — Rock samples collected from geological formations in Karnataka

Sample Code	Rock Type
KA-1	Gneiss
KA-2	Granite
KA-3	Trap
KA-4	Basalt
KA-5	Peninsular Gneiss
KA-6	Dolomite Rock
KA-7	Deccan Trap
KA-8	Sandstone
KA-9	Quartzite
KA-10	Limestone
KA-11	Laterite

The neutron fluence used for irradiation of the samples is given in Table 2. Since it was intended to estimate the trace elements that give rise to long lived radionuclides, the samples were allowed to cool for three months so that all the short lived activities decayed down to insignificant levels. After three months, the presence of ^{60}Co was detected from the aluminium foil used for wrapping the rock samples. So the aluminium foils were removed and samples were re-wrapped in paper. These samples were then counted for 5000 seconds each on a low background gamma counting set-up using a *p*-type high purity germanium (HPGe) gamma spectrometer. The gamma spectrometric system has 30% relative efficiency and a resolution of 1.95 keV at 1332.5 keV. Energy calibration of the detector was carried out using reference standards ^{60}Co , ^{137}Cs and $^{152+154}\text{Eu}$. The room background spectrum where the counting set-up is installed, was also measured for the same counting time. The net spectrum for each of the samples was obtained by subtracting the background spectrum from the recorded sample spectrum.

Some of the important long lived gamma emitting radionuclides, generally, produced in the concrete shields of nuclear reactors and accelerators along with the thermal neutron cross-section for the relevant reactions¹⁰ are listed in Table 3. It may be noted that total cross-section for the reaction $^{151}\text{Eu}(n,\gamma)^{152}\text{Eu}$ is the sum of the cross-sections ($\sigma_g = 5900$ barns) populating the ground state of $^{152}\text{Eu}_g$ and that ($\sigma_{m2} = 3300$ barns) for the second metastable state $^{152}\text{Eu}_{m2}$ which decays by isomeric transition with a half life of 9.2 min. From Table 3, we find that, in concrete,

long-lived activity (half lives in years) due to ^{60}Co , ^{134}Cs , $^{152,154}\text{Eu}$, are formed due to (n,γ) reactions on ^{59}Co , ^{133}Cs , and $^{151,153}\text{Eu}$, respectively. Relatively short lived activity (half lives in days) ^{46}Sc , ^{59}Fe , ^{182}Ta are formed due to (n,γ) reactions on ^{45}Sc , ^{58}Fe , ^{181}Ta . The long-lived radionuclides are important from the point view of decommissioning after the end of life time of the facility, whereas the short-lived radionuclides are responsible for collective dose to personnel during maintenance and other activities during the operational life of the facility. So, in order to identify the CA samples suitable for low radioactivation, it is necessary to determine the amount of the respective parent nuclide in different samples. The eleven CA collected from eleven different geological compositions were analysed for the production of the seven radioisotopes mentioned in Table 3 due to neutron activation. Quantitative analysis of the respective parent elements were carried out by comparing the gamma spectrometric data of the samples with those for two reference materials IAEA-158 and IAEA-433.

2.3 Quality Control Analysis

Quality control analysis of the experimental set-up to test the accuracy of the measured results were realized by using two certified reference materials, treating one as a standard and the other as a sample. Of the two standard samples, IAEA-158 (S_1) was treated as known standard, and IAEA-433 (S_2) was treated as an unknown sample. Elemental composition of both S_1 and S_2 are provided by IAEA. The counts have been recorded under each of the gamma peaks mentioned in Table 3 for both the samples. Elemental composition of S_2 is determined from the measured counts, measured weight of the two samples S_1 and S_2 and known elemental composition of S_1 as detailed below. Since both the samples S_1 and S_2 were co-irradiated and counted in identical geometry, the concentration (N_2) of an element in the unknown standard S_2 is given by:

Table 2 — Neutron fluence values at the position of irradiation of the samples

Neutron Energy Range	Fluence
< 0.625 eV	1.03×10^{18} n/cm ²
0.625 eV - 0.821 MeV	2.26×10^{17} n/cm ²
0.821 MeV-10MeV	1.64×10^{16} n/cm ²

Table 3 — Long lived radionuclides produced in the concrete shields of nuclear reactors and accelerators through (n, γ) reaction

Radioisotope	Half-life	γ energy (keV)	Parent nuclide	Abundance (%)	Thermal neutron cross-section (barns) [13]
^{46}Sc	83.8 d	889.3	^{45}Sc	100	27.2
^{59}Fe	45 d	1099.2	^{58}Fe	0.28	1.32
^{60}Co	5.26 y	1332.5	^{59}Co	100	37.2
^{134}Cs	2.05 y	795.8	^{133}Cs	100	30.3
^{152}Eu	13.5 y	1408	^{151}Eu	47.8	9200
^{154}Eu	8.5 y	1274.4	^{153}Eu	52.2	312.0
^{182}Ta	115 d	1121.3	^{181}Ta	100	20.5

$$N_1/N_2 = (C_1/W_1)/(C_2/W_2) \quad \dots(1)$$

$$\text{or, } N_2 = \left(\frac{C_2}{C_1}\right)\left(\frac{W_1}{W_2}\right)N_1 \quad \dots(2)$$

where C_1 is counts/sec for sample S_1 (IAEA-158); C_2 the counts/sec for sample S_2 (IAEA-433); W_1 the weight of the sample S_1 ; W_2 the weight of the sample S_2 ; N_1 the elemental concentration of a particular element in sample S_1 as quoted by IAEA; N_2 the calculated elemental concentration of a particular element in sample S_2 and N_2 is the elemental concentration of a particular element in sample S_2 as quoted by IAEA.

From Eq. (2), if the weights W_1 and W_2 for samples S_1 and S_2 , respectively and concentration N_1 of an element in sample S_1 are known, concentration of the element in sample S_2 can be determined from the measured counts C_1 and C_2 under the relevant gamma peak for the two samples. The concentration N_2 of an element is calculated in sample S_2 using the IAEA quoted value for N_1 . This calculated N_2 is compared with the IAEA quoted value for N_2 in Table 4.

The concentrations of Sc, Fe, Co, Cs, Eu and Ta in the eleven different CA samples have been determined following the same method described

above for the quality control analysis. For this purpose, the peak counts for the seven different radioisotopes produced from the samples are compared with those from the two IAEA standards. The result of this analysis is given in Table 5.

2.4 Calculation of Induced Activity

Activity of the seven radioisotopes mentioned in Table 2 and induced in the CA samples due to neutron irradiation has been determined from the measured concentration of the parent elements. The activity A of a particular radio-nuclide in the sample after the cooling period is given by:

$$A = N \int \sigma(E)\phi(E)dE [1 - \exp(-\lambda t)] \exp(-\lambda \tau) \quad \dots (3)$$

where N is the number of atoms of the target parent nuclide available in the sample for interaction; $\sigma(E)$ the production cross-section of the radioisotope through neutron activation at energy E ; $\phi(E)$ the neutron fluence at energy E ; λ the decay constant of the radionuclide formed; t the irradiation time and τ is the cooling time of the irradiated sample.

To estimate the potential activity build-up in the rock samples due to thermal neutrons, the number of parent atoms in a given sample was determined from

Table 4 — Quality control analysis Weight of $S_1 = 50.3$ mg, weight of $S_2 = 50.6$ mg

Element	γ energy (keV)	CPS for S_2 (C_2)	CPS for S_1 (C_1)	N of S_1 (N_1)	N of S_2 (calculated) (N_2')	N_2 (IAEA quoted)	95% confidence interval (quoted by IAEA)	Ratio (N_2'/N_2)
Cs	795.8	1.69	0.952	3.73	6.58	6.4	6.03-6.77	1.027
Sc	889.3	22.81	12.60	8.32	14.97	14.6	13.4-15.8	1.025
Fe	1099.2	8.43	5.46	26.3	40.36	40.8	40.3-41.3	0.989
Ta	1121.3	0.28	0.24	0.97	1.12	1.03	0.94-1.12	1.087
Co	1332.5	4.70	3.26	9.2	13.18	12.9	12.6-13.2	1.021
Eu	1408.0	0.97	0.87	1.08	1.20	1.18	1.11-1.25	1.017

Table 5 — Trace element concentrations of rock samples by NAA

Sample Code	Trace Element Concentration (mg/kg for all others and g/kg for Fe) ^a					
	Cs	Sc	Fe	Ta	Co	Eu
Gneiss (KA-1)	1.46 ± 0.04	6.55 ± 0.04	31.42 ± 0.26	0.87 ± 0.04	7.62 ± 0.09	1.76 ± 0.03
Granite (KA-2)	2.09 ± 0.08	5.61 ± 0.04	35.37 ± 0.17	0.64 ± 0.03	10.31 ± 0.11	1.28 ± 0.02
Trap (KA-3)	3.72 ± 0.08	3.04 ± 0.08	15.42 ± 0.16	0.95 ± 0.03	3.44 ± 0.06	1.04 ± 0.02
Basalt (KA-4)	3.87 ± 0.08	3.40 ± 0.03	21.27 ± 0.20	0.88 ± 0.04	5.01 ± 0.07	1.07 ± 0.03
Peninsular Gneiss (KA-5)	0.5 ± 0.02	26.19 ± 0.12	58.93 ± 0.44	0.14 ± 0.01	37.67 ± 0.33	0.79 v 0.02
Dolomite Rock (KA-6)	0.09 ± 0.01	1.64 ± 0.02	7.39 ± 0.10	0.05 ± 0.005	5.31 ± 0.07	0.18 ± 0.01
Deccan Trap (KA-7)	0.64 ± 0.02	32.88 ± 0.15	101.11 ± 0.69	0.63 ± 0.03	51.84 ± 0.43	1.94 ± 0.04
Sandstone (KA-8)	0.47 ± 0.02	4.24 ± 0.03	7.95 ± 0.10	0.41 ± 0.02	7.04 ± 0.08	0.79 ± 0.02
Quartzite (KA-9)	0.09 ± 0.01	0.65 ± 0.01	3.07 ± 0.06	0.08 ± 0.01	0.79 ± 0.02	0.12 ± 0.01
Limestone (KA-10)	0.55 ± 0.02	4.82 ± 0.03	15.77 ± 0.16	0.13 ± 0.01	6.14 ± 0.08	0.35 ± 0.01
Laterite (KA-11)	0.28 ± 0.02	37.97 ± 0.17	282.81 ± 1.85	0.66 ± 0.03	6.58 ± 0.08	0.37 ± 0.01

the measured elemental concentration. It was then folded with the neutron flux and production cross-section at neutron energy E . The total activity induced for a radioisotope was thus calculated using Eq. (3) for a given irradiation time t and a decay time τ by integrating over the entire energy range of the neutron fluence. In order to obtain the detailed neutron fluence distribution $\phi(E)$ we have used a normalized typical reactor core spectrum for a ^{235}U based thermal reactor¹¹. This normalized spectrum is folded with the total neutron fluence used in the present work to obtain the energy distribution of neutron fluence. Thermal neutron induced excitation function $\sigma(E)$ for different radioisotopes are taken from published data¹⁰. The results for the calculated activity are given in Table 6.

2.5 Sources of Uncertainty

Uncertainty in determining the concentration of different elements in the unknown sample comes in from different sources. These are mainly uncertainties in measurement of the mass and the total counts of standard and unknown samples. In the present work, uncertainty in mass determination amounts to 0.2-1%. Both the standard and unknown samples were counted for 5000 s. Uncertainty in the sample counts amounts to a maximum of 5% which contributes to the total uncertainty in the measured concentration of an element in the sample. Another source of uncertainty in estimating the elemental concentration is the uncertainty in the counting geometry. This comes from manual variation in the relative position of the standard and the unknown samples in the counting set-up. The elemental concentration in the unknown sample is determined through comparison with that of the standard sample and both the standard

and the unknown are irradiated simultaneously in the reactor. So contribution from other sources like irradiation geometry, sample isotopic abundance, decay half-life, gamma decay branching ratio, fluctuation of reactor power and resulting neutron flux, neutron scattering in the reactor core, gamma detection efficiency of the HPGe detector, need not be considered.

However, in a situation where simultaneous irradiation with standard samples is not possible, NAA becomes complicated. To analyze such situations, we have calculated the activities of the seven isotopes studied for all the eleven samples from the measured concentration, measured neutron flux and reaction cross-sections published in the literature. In this activity calculation, uncertainty from all the above-mentioned sources has contributions of varying degree. Uncertainty in sample isotopic abundance amounts to a maximum of 3%. Decay half-life of the isotope and the gamma branching ratio are taken from published literature and the uncertainties involved in these quantities are negligible. Uncertainty in the detection efficiency of the HPGe detector varies by 0.3-3% over the energy range studied. The other source of uncertainty in the measurements comes from irradiation geometry and estimation of the neutron fluence. The neutron fluence for the reactor Dhruva is given in three different energy bins. But as the production cross-section varies rapidly with energy, we have estimated the energy distribution of neutron spectrum using a typical thermal reactor core neutron energy spectrum from the work of Bitelli *et al*¹¹. A variation of 10% in the low energy part of this spectrum would result in a similar variation in the total activity, as the production cross-section as well as the neutron fluence dominate in this energy region over that at higher energies.

Table 6 — Activity build-up in the rock samples (half-lives given in parentheses)

Sample Code	Activity build-up (Bq) in the rock samples (mass: 50 mg, irradiation time: 3.5 days, cooling time: 94 days)						
	Cs-134 (2.05y)	Sc-46 (83.8d)	Fe-59 (45.0d)	Ta-182 (115d)	Co-60 (5.26y)	Eu-152 (13.5y)	Eu-154 (8.5y)
Gneiss (KA-1)	101.2±2.9	5325.1±34.1	2986.5±25.9	119.7±5.8	601.4±7.5	2582.4±46.2	148.7±2.7
Granite (KA-2)	144.9±5.8	4560.8±34.1	3361.9±17.0	88.1±4.3	813.8±9.1	1857.0±30.5	107.0±1.8
Trap (KA-3)	257.9±5.8	2471.5±68.3	1465.7±16.0	130.7±4.3	271.5±5.0	1508.8±30.4	86.9±1.7
Basalt (KA-4)	278.7±6.0	2764.2±25.6	2021.7±20.0	121.1±5.8	395.4±5.8	1552.3±47.5	89.4±2.7
Peninsular Gneiss (KA-5)	34.7±1.5	21292.1±102.4	5601.3±43.9	19.3±1.4	2973.3±27.3	1146.1±32.5	66.0±1.9
Dolomite (KA-6)	6.6±0.8	1333.3±17.0	702.4±10.0	6.9±0.7	419.1±5.8	275.6±11.2	15.9±0.6
Deccan Trap (KA-7)	44.4±1.5	26731.0±128.0	9610.5±68.9	86.7±4.5	4091.7±35.6	2887.1±62.5	166.3±3.4
Sandstone (KA-8)	32.6±1.5	3447.1±17.1	755.6±10.0	56.4±2.9	555.7±6.6	1146.1±32.5	66.0±1.9
Quartzite (KA-9)	6.2±0.7	528.4±8.5	291.8±6.0	11.0±1.4	62.4±1.7	174.1±15.2	10.0±0.9
Limestone (KA-10)	38.8±1.5	3918.6±25.6	1498.9±16.0	17.9±1.5	484.6±6.6	507.8±15.2	29.2±0.9
Laterite (KA-11)	20.8±1.6	30869.1±145.1	26881.0±184.6	93.6±5.0	519.4±6.7	580.3±16.5	33.4±0.9

3 Results and Discussion

The results of the quality control analysis are given in Table 4. From Table 4, it is seen that the measured concentration of unknown standard S_2 is well within the 95% confidence interval quoted by IAEA. The measured concentration of all the elements, except Ta in S_2 agrees within $\pm 2.7\%$ of the IAEA quoted values. The counting set-up can be used for a reliable estimate of the elemental concentration in unknown samples by this method of comparison. After carrying out the quality assurance test for our counting set-up, the trace elemental concentration has been determined in the eleven CA samples through gamma spectrometric analysis of the neutron irradiated samples. Figure 1 shows the gamma spectra for sample Trap (KA-3) in the room background

For each of the eleven CA samples under study, concentrations of the six elements Sc, Fe, Co, Cs, Eu and Ta are determined from counts under the respective photo peak in the gamma spectrum. This is done using Eq. (2) by comparing the sample counts with that for the IAEA reference materials. The results of determination of trace element concentrations in CA samples collected are given in Table 5.

From a comparative study of the trace element compositions determined from the neutron activation analysis of the rock samples, the following observations may be made and conclusions may be drawn:

- (1) Caesium: The minimum concentration of caesium is found in samples of Dolomite Rock (KA-6),

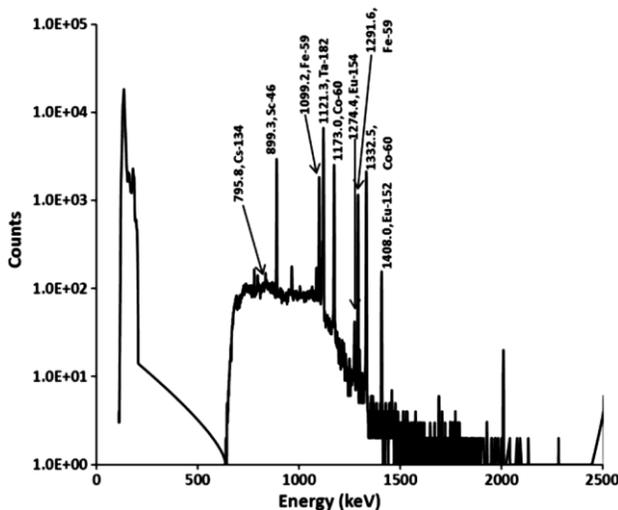


Fig. 1 — Gamma spectrum recorded for CA sample Trap (KA-3) + background

and Quartzite (KA-9) (0.09 mg/kg), and maximum concentration of caesium is found in Basalt (KA-4) (3.87 mg/kg). The average value of caesium for this set of samples is 1.25 mg/kg.

- (2) Scandium: The minimum and maximum concentration of scandium is found in the samples of Quartzite (KA-9) (0.65 mg/kg) and Laterite (KA-11) (37.97 mg/kg), respectively. The average value of scandium for this set of samples is 11.54 mg/kg.
- (3) Iron: The minimum and maximum concentration of iron is found in the samples of Quartzite (KA-9) (3.07 g/kg) and Laterite (KA-11) (282.81 g/kg), respectively. The average value of iron for this set of samples is 52.77 g/kg.
- (4) Tantalum: The minimum and maximum concentrations of tantalum is found in the samples of Dolomite Rock (KA-6) (0.05 mg/kg) and Trap (KA-3) (0.95 mg/kg), respectively. The average value of tantalum for this set of samples is 0.50 mg/kg.
- (5) Cobalt: The minimum and maximum concentrations of cobalt is found in the samples of Quartzite (KA-9) (0.79 mg/kg) and Deccan Trap (KA-7) (51.84 mg/kg), respectively. The average value of cobalt for this set of samples is 12.89 mg/kg.
- (6) Europium: The minimum and maximum concentration of europium is found in the samples of Quartzite (KA-9) (0.12 mg/kg) and Decan Trap (KA-7) (1.94 mg/kg), respectively. The average value of europium for this set of samples is 0.88 mg/kg. This is the most important of the elements considered here from the point of decommissioning since it has very high thermal neutron capture cross-section to produce long lived radionuclides that emits hard gamma rays.

The activity of the seven isotopes induced in the rock samples have been calculated from the measured elemental concentration using Eq. (3). The neutron spectrum used for the purpose is shown in Fig. 2. It is estimated by folding the normalised reactor core spectrum from Ref. (11) with the energy distribution of neutron fluence given in Table 2. Production cross-sections for the radioisotopes are taken from the measured data of other researchers¹⁰. The induced activity of the isotopes after 3.5 days of irradiation followed by 94 days of cooling is given in Table 6. For all the samples, activity induced for the isotopes other than those for ^{46}Sc and ^{59}Fe vary from few Bq to

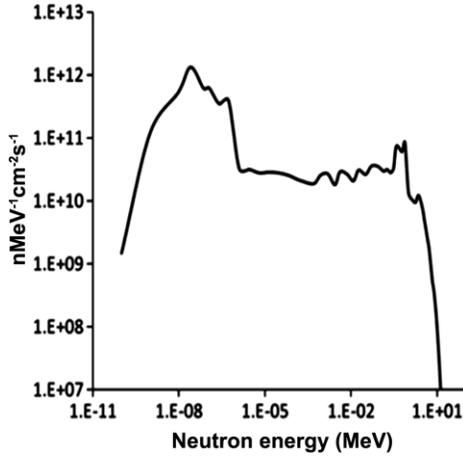


Fig. 2 — Estimated energy distribution of neutron fluence used for this work

several hundreds of Bq (Table 6). Activity of ^{46}Sc and ^{59}Fe is of the order of several kBq in the rock samples. Of the eleven samples studied quartzite shows the least induced activity. But in a previous study, sandstone and quartzite failed the fresh properties tests while laterite failed the fresh and hardened properties tests for preparation⁸ of SCC. Of the other CA samples, Dolomite Rock shows minimum induced activity. Since for 3.5 days of irradiation, long-lived activity induced in this CA sample is at most of the order of 420 Bq, due to ^{60}Co , (^{59}Fe and ^{46}Sc have half-lives of several days), Dolomite Rock can be considered suitable for developing SCC for reactor and accelerator shielding.

The induced activities are determined from the measured elemental concentration using estimated neutron fluence rate and production cross-section of the radioisotopes. Induced activity of a given isotope at the end of 94 days of cooling can also be calculated from the measured counts C under the characteristic gamma peaks using the relation:

$$C = (A \varepsilon_i b_i [1 - \exp(-\lambda t_m) c_i]) / \lambda \quad \dots(4)$$

where ε_i is the detection efficiency of the detector at the gamma energy, b_i is the gamma branching ratio, t_m is the counting time, c_i contains other correction factors due to primary and secondary neutron scattering, other symbols have the same meaning as described in Eq. (3). In Eq. (4), $\varepsilon_i c_i$ constitutes the total correction factor. Using the calculated activity of different isotopes from Table 3 and Eq. (4), this correction factor has been evaluated. The correction

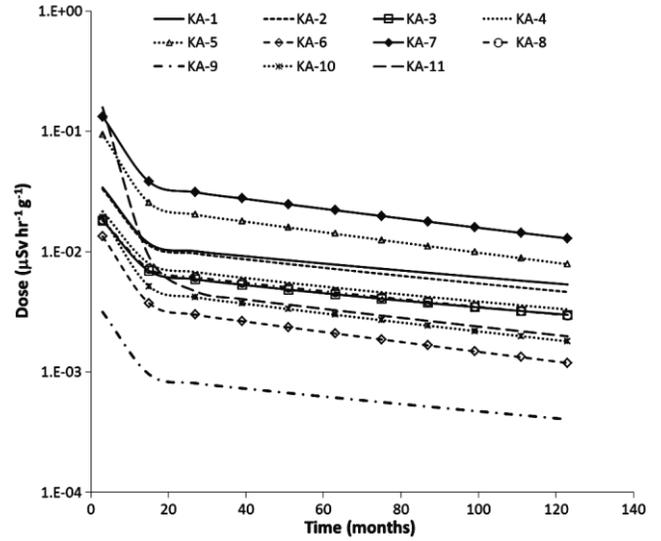


Fig. 3 — Dose decay profile for the eleven CA samples

factor varies between $9.0\text{E-}3$ and $1.58\text{E-}2$, the average value being $1.02\text{E-}2$.

The γ dose has also been estimated from the calculated activity of each of the seven radioisotopes in all the eleven CA samples using the relation:

$$D = 6AE/d^2 \quad \dots(5)$$

where D is the dose in mR/h, A the activity of the γ emitting radioisotope in mCi, E the total energy of emitted γ radiation in MeV, d is the distance in ft. of point of observation from the source. The total dose from a CA sample is obtained from the sum of the doses due to all the seven isotopes. Time variation of the dose has been followed starting from a cooling time of 94 days to 123 months. Figure 3 shows the time profile of γ dose from all the CA samples. From Fig. 3, it is observed that among the SCC compatible CA samples Dolomite Rock (KA-6) produces the lowest dose of the order of $1.32 \times 10^{-2} \mu\text{Sv h}^{-1} \text{g}^{-1}$. Highest dose comes from Deccan Trap (KA-7) and is of the order of $1.33 \times 10^{-1} \mu\text{Sv h}^{-1} \text{g}^{-1}$. It has also been observed from Fig. 3 that at the end of 94 days of cooling the CA sample Laterite (KA-11) produces the highest radiation dose of all the samples, but the dose falls off quickly with time.

This is attributed to the fact that Laterite contains a high concentration of ^{46}Sc and ^{59}Fe with half lives of 45 days and 83.8 days, respectively. It should be noted that the dose values reported here are for one g of sample and equation 5 is valid for a point source. But in the case of any real facility, the total dose

would be much higher depending on the dimension of the shield walls. Moreover, the point source approximation needs to be modified for an extended source. Nevertheless, the present analysis gives the relative trend that holds well despite very approximate dose rate values.

4 Summary and Conclusions

- (1) Trace element concentrations having the potential of generating long lived gamma emitting radionuclides in the eleven rock samples have been determined using the neutron activation analysis. From these results, one can select the aggregates that would produce low amounts of induced activities in the self-compacting concrete shields of nuclear reactors and particle accelerators, which in turn would reduce the amount of low level radioactive waste to be handled at the time of decommissioning of the facility.
- (2) Of the eleven coarse aggregate samples collected and analysed, it is seen that the samples of Quartzite show the least activity build-up (Table 6). But, this sample failed the fresh property test for preparing self-compacting concrete⁸. Among the CA samples compatible for SCC, Dolomite rock developed least induced activity. In this sample, the activity induced for ⁴⁶Sc is about 1.2 kBq, for the other six isotopes, the activity varies between few Bq to a few hundreds Bq. Our estimation of the time variation of radiation dose from this sample shows that the total dose from one g of the sample falls below $1.2 \times 10^{-3} \mu\text{Sv h}^{-1} \text{g}^{-1}$ 10 years after the reactor is shut down. It may, therefore, be concluded that the coarse aggregate containing Dolomite rock (KA-6) is best suited for use in building the SCC shields with low potential for neutron induced radioactivation.
- (3) A judicious choice of the coarse aggregate (CA) can reduce the residual radioactivity in concrete shield (SCC or normal) by a factor of about ten.

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