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DETECTION OF ^{41}Ar IN THE VICINITY OF PAKISTAN RESEARCH REACTOR – 1 DURING 2007-2009

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The study reports the determination of ^{41}Ar in the vicinity of Pakistan Research Reactor - 1 (PARR-1) during 2007-2009. ^{41}Ar was determined in the neutron activation analysis laboratory, Chemistry Division, PINSTECH during its normal spectra acquisitions. Initially, the peak at 1293.7 keV went un-identified but recently it has been associated with ^{41}Ar. Although, the source of ^{41}Ar is known, which is PARR-1 but its pathway into the laboratory is unknown. Moreover, the rate of production of ^{41}Ar and the dose produced by it is also not known. The presence of a strong peak at 1293.7 keV not only masks the 1292.6 keV γ-line of ^{59}Fe but it also increases the limit of detection for most of radionuclides. Its presence, therefore, should be taken seriously for neutron activation analysis work and for low-level countings.

Keywords: ^{41}Ar, PARR-1, Detector background

1. Introduction

Radioactivity in the reactor coolant originates in several ways; one among these is the activation of argon in teaching and research reactors of the swimming pool type. The concentration of argon in air at sea level is 0.934% and is the third most abundant gas in air. Argon has three natural isotopes ^{36}Ar (0.337%), ^{38}Ar (0.063%) and ^{40}Ar (99.600%). A certain amount of argon present in air is usually dissolved in the water and when this water passes through the reactor core, the argon becomes activated. Later this radioactive argon emerges from the pool surface and passes into the atmosphere [1]. The activated products of the three natural isotopes are given as;

$$^{36}\mathrm{Ar} \xrightarrow{(n,\gamma)} {}^{37}\mathrm{Ar}$$

$$^{38}\mathrm{Ar} \xrightarrow{(n,\gamma)} {}^{39}\mathrm{Ar}$$

$$^{40}\mathrm{Ar} \xrightarrow{(n,\gamma)} {}^{41}\mathrm{Ar}$$

Among the three activation products only ^{41}Ar can be detected by γ -ray spectrometry. The radiative capture cross section for $^{40}\mathrm{Ar}(n,\gamma)^{41}\mathrm{Ar}$ is 0.66 barn and resonance integral is 0.41 barn. ^{41}Ar is removed from the reactor building by releasing it well above the ground level using stack. Its monitoring is important from the dose management point of view. Its presence has been reported around the Canada India Reactor, India, a 40 MW reactor, at a level of 640 Ci/day (1.2 BqL^{-1}) [2] around Taiwan Research Reactor, a 40 MW reactor, at 3000 Ci/day and around Kyoto University Research Reactor, Japan, a 5MW reactor, at an amount of 20 BqL^{-1} [4]. Its emission has also been reported qualitatively by Belgium BR1 research reactor, Mol [5].

Pakistan Research Reactor-1 (PARR-1) is a 10 MW swimming pool type reactor [6] has 20% enriched uranium core (U_3Si_2-Al), light water as moderator and graphite as reflector. It has 8 channels namely RS1, RS2, RS3, Beam Tube 4, Beam Tube 5, Thermal column A, B and C with varying level of neutron flux. The present paper discusses the detection of ^{41}Ar at the Neutron Activation Analysis (NAA) Laboratory during its normal spectra acquisitions operations. The NAA laboratory is located at the top floor of Chemistry Division, which is near the reactor building. Since there is no direct linkage between the reactor hall and the Chemistry Division, therefore the most probable pathway of ^{41}Ar into the NAA laboratory is through windows, or small passages into the laboratory.

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Table 1. Possible radionuclides associated with 1293.7 keV peak.

Radionuclide	Parent of (n,γ) reaction	Half-life	Energy (keV)	Intensity	Main Line (keV)	Intensity	Peak in Spectrum
^{41}Ar	^{40}Ar	109.6 min	1293.6	99.2	1293.6	99.2	✓
^{83}Se	^{82}Se	22.3 min	1293.8	2.4	356.7	100	×
^{131}Te	^{130}Te	25 min	1294.3	0.5	149.7	68.8	×
^{194}Ir	^{193}Ir	19.3 hour	1293.7	0.4	328.5	100	×
^{151}Nd	^{150}Nd	12 min	1293.6	0.3	116.8	39	×
^{101}Mo	^{100}Mo	14.6 min	1293.3	0.2	590.1	19.2	×
^{152}Eu	^{151}Eu	13.5 year	1292.8	0.1	121.8	28.7	✓
^{149}Nd	^{148}Nd	1.7 hour	1293.4	0.02	211.4	25.9	×

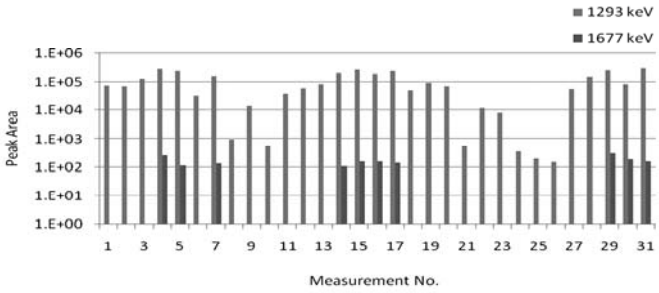

Figure 1. Peak areas found at 1293.7 keV and at 1677 keV peaks.

2. Experimental

All spectra were acquired by a p-type coaxial HPGe detector (Eurisys Mesures, France). The detector has 60% relative efficiency and 1.95 FWHM at 1332 keV and is connected to an Ortec-570 amplifier and Trump PCI 8k ADC/MCA card with GammaVision-32 ver. 6 software. The sensitive volume of the detector is 245 cm^3 with dead layer of 500 μGe and length 70mm, external diameter 67.5 mm having entrance window of Al with thickness ≤ 1 mm. The detector shield is made up of lead in the form of cylinder of length 40.5 cm, with bottom and top open to house detector crystal, the wall thickness of lead shield is 5.5 cm. There is no internal lining on the lead shields. All calculations, in this study, have been made in Microsoft Excel release 2007.

3. Results and Discussion

In our laboratory, it was known for a long time that a peak at 1293.7 keV appears only on non-contiguous weekends. The peak went unidentified till 2009. The main reason for not identifying this peak was its association with ^{41}Ar, which is usually not formed in solid samples analyzed by the laboratory, by neutron activation analysis. In 2007, the old Canberra 85 series multichannel analyzer (MCA) was replaced by the Ortec made MCA, which could save all the acquired spectra. Recently, the spectra acquired were analyzed by our in-house written program GammaLab. The 1293.7 keV peak was once again identified as ^{41}Ar. The peak at 1293.7 keV was present in 34 spectra starting from March 2007 to August 2009. Table 1 presents the candidate radionuclides associated with the 1293.7 keV peak with search window of ± 1 keV. The search provided 8 radionuclides produced by (n,γ) reaction, among all the candidates only ^{41}Ar is the gas. Since rest of the radionuclides exists in non-gaseous form, therefore ^{41}Ar was the most probable candidate associated with the peak. Moreover, other nuclides did not show their most abundant gamma-ray in the spectra except ^{152}Eu which is part of the NAA laboratory background. However, the intensity ratio of ^{152}Eu at 121.8 keV to 1293.7 keV is 28.7:0.1; the Spectrum ID 611 (acquired on 22/8/08) shows peak areas 2809 at 121.8 keV and 242013 at 1293.7 keV which indicates the presence of another radionuclide at 1293.7 keV. The presence of ^{41}Ar is further supported by its second most abundant peak found at 1677 keV with gamma abundance of 0.05. Figure 1 shows the presence of 1677 keV peak in the spectra, due to the small gamma abundance of this γ-line it appeared in fewer spectra.

Table 2. Record of the spectra recorded with reactor operation.

Spectrum ID	Spectrum acquisition start	Acquisition time (hr)	Peak 1293.7 keV	Reactor operated	Reason
168	02/03/2007 15:38	16	No	Yes	Unknown
160	16/03/2007 16:32	16	Yes	Yes	
193	11/05/2007 09:05	16	Yes	Yes	
Background	8/06/2007 15:55	65	Yes	Yes	
55	22/06/2007 16:18	16	No	Yes	Unknown
Background	6/07/2007 16:14	11	Yes	Yes	
47	20/07/2007 15:22	14	Yes	Yes	
193	17/08/2007 14:45	24	Yes	Yes	
Background	17/10/2007 14:24	19	Yes	Yes	
220	16/11/2007 14:56	24	Yes	Yes	
229	30/11/2007 09:29	24	No	Yes	Unknown
205	18/01/2008 08:21	24	Yes	Yes	
167	22/02/2008 09:42	24	Yes	Yes	
166	29/02/2008 15:26	24	Yes	Yes	
186	14/03/2008 13:27	24	Yes	Yes	
203	02/05/2008 15:27	24	Yes	Yes	
Background	23/05/2008 15:49	24	Yes	Yes	
Background	01/08/2008 13:15	24	Yes	Yes	
327	22/08/2008 14:04	16	Yes	Yes	
332	29/08/2008 08:04	16	Yes	Yes	
Background	10/10/2008 14:31	22	Yes	Yes	
358	24/10/2008 14:29	16	Yes	Yes	
735	21/11/2008 14:51	19	Yes	Yes	
Background	26/12/2008 14:07	24	Yes	Yes	
823	23/01/2009 15:07	26	Yes	Yes	
843	06/02/2009 11:25	14	Yes	Yes	
894	20/02/2009 15:12	12	Yes	Yes	
981	20/03/2009 08:29	25	Yes	Yes	
990	03/04/2009 15:30	13	Yes	Yes	
1062	15/05/2009 14:59	13	Yes	Yes	
1191	26/06/2009 15:07	13	Yes	Yes	
Background	10/7/2009 12:05	13	Yes	Yes	
1296	24/07/2009 13:33	13	Yes	Yes	
1356	07/08/2009 12:05	13	Yes	Yes	

In the normal practice PARR-1 operates on Friday, the 1293.7 keV peak was observed only in those days when PARR-1 was in operation, however, the peak was not observed on each and every operation of the reactor. Table 2 shows the record of all spectra which were acquired on the day of operation. Figure 2 shows 1293.7 keV peak area variation around the year, it shows that the recorded peak areas are higher in summer months. Figure 3 presents the peak area divided by the overlapping time of detector-reactor operation, which produces the similar information as depicted by Figure 2. The label on the top of each bar in Figure 3 represents the month of acquisition. This figure, however, indicates the presence of seasonal effect on peak areas. The peak areas increase during the summer months as compared to the winter. This indicates diffusion of more air into the NAA laboratory during summer months than winter.

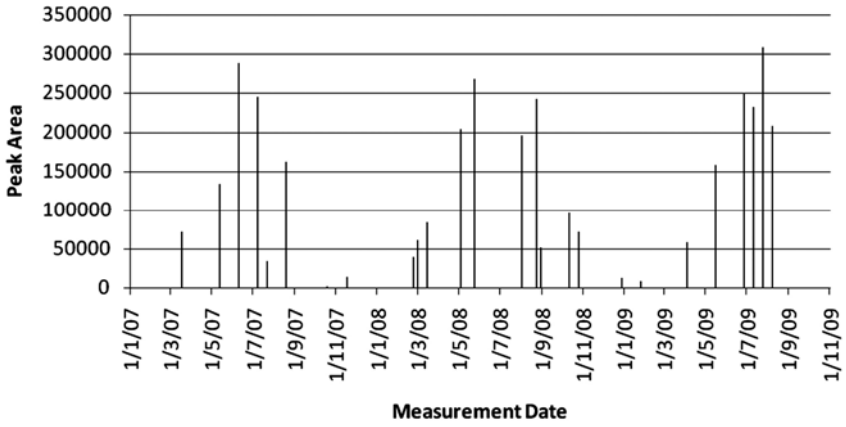

Figure 2. Variation of peak area of ^{41}Ar at 1293.7 keV peak with date.

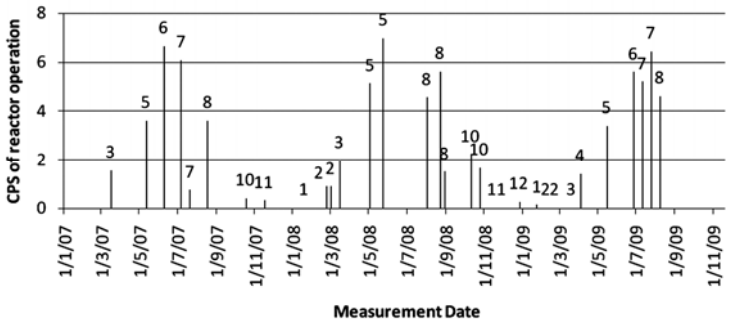

Figure 3. Peak area divided by the overlapped time of detector-reactor operation, number on each bar represents the month of acquisition.

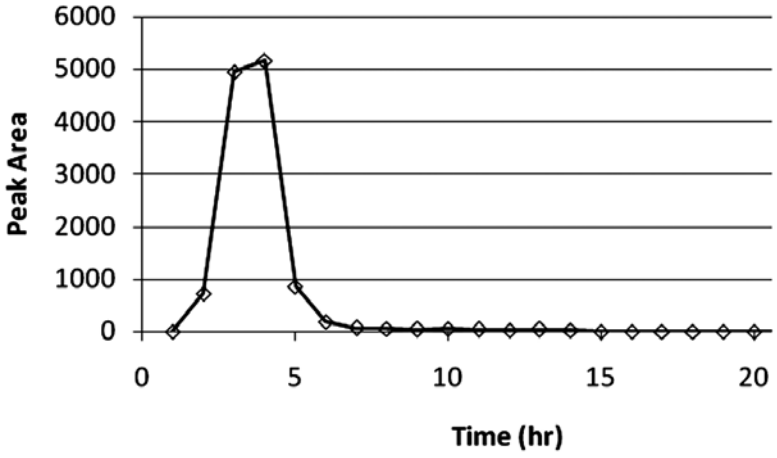

Figure 4. Variation of peak areas of ^{41}Ar at 1293 keV, collected for 1 hr in consecutive 24 spectra.

In order to study the peak area variation of ^{41}Ar during a single reactor run, a special experiment was conducted on 26/12/08. During this experiment 24 spectra were recorded consecutively each having acquisition time of 1 hour. The acquisition of the first spectrum was started at 1400 hr. The results are produced in Figure 4, which shows that ^{41}Ar was present in the laboratory in significant amount during the first 5

hours. The peak area at 1293.7 keV dropped to less than 100 counts in the last 7 hours of reactor operation. The reason for small peak areas in the last 7 hours is probably the drop in temperature during night hours, which caused smaller diffusion of ^{41}Ar into the laboratory.

The present data cannot be used to measure the amount of ^{41}Ar generated in the reactor pool because the peak area of ^{41}Ar is dependent on various parameters such as reactor operation timings, meteorological conditions, reactor power, the amount of air present in water, the water circulation pattern and other factors. The present paper, however, reports the presence of ^{41}Ar for the first time. The information in this paper is not only useful for health physics point of view but also from gamma-spectroscopy. The presence of ^{41}Ar main peak at 1293.7 keV masks strongly with the 1292.6 keV peak of ^{59}Fe, which is identified most of the time in NAA work. Moreover, the presence of ^{41}Ar in a gamma-ray spectrum increases the limit of detection for many radionuclides. It is, therefore, suggested that the gamma spectrum acquisition should be taken with more care during the time of reactor operation than in the other days.

4. Conclusions

This study shows that ^{41}Ar is the true radionuclide associated with the 1293.7 keV peak found in the spectra acquired during 2007-2009 in the NAA laboratory, Chemistry Division, PINSTECH. ^{41}Ar was observed in 31 spectra out of the 34 reactor operations, the absence of 1293.7 keV peak in the 3 acquisitions may be due to the different meteorological conditions. An estimate of the dose produced by the emission of ^{41}Ar cannot be made using the available data of peak areas. However, this information can be useful for NAA work, where ^{59}Fe is identified or low level counting is required. A further study, however, could be conducted with a suitable mathematical model or monte-carlo simulation to determine the rate of production of ^{41}Ar, which could be used for dose determination. This study, which started with the identification of an unknown peak at 1293.7 keV, concludes the release of ^{41}Ar from PARR-1, which was unknown before this writing.

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