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Report 20

PRODUCTION OF RADIOACTIVE AEROSOLS IN THE RADIATION FIELD OF THE SYNCHRO-CYCLOTRON AT THE LENINGRAD NUCLEAR PHYSICS INSTITUTE

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A b s t r a c t

It is established that the radioactivity of aerosols during accelerator runs is caused by the nuclides ^{31}Si, ^{24}Na, ^{28}Mg, ^{32}P, ^{33}P and ^{7}Be. It is shown that the main process leading to the formation of active aerosols is the activation of argon in the free air. For an internal synchrocyclotron current of 0.4 μA, an extracted beam of 6.10^{11} protons/sec. and an air renewal rate in the accelerator hall of 2.5 hours^{-1}, the equilibrium concentrations of ^{31}Si, ^{24}Na, ^{28}Mg, ^{32}P, ^{33}P and ^{7}Be are (96 ± 30), (7 ± 2), (3 ± 1), (2 ± 1), $\lesssim 1$, $(17 \pm 4).10^{-14}$ curie/litre respectively. The results are compared with the data obtained at other accelerators.

Introduction

As was shown in paper [1], radiation from the synchrocyclotron, which accelerates protons up to 1 GeV, makes the air radioactive. The induced radioactivity may be not only gaseous but also like an aerosol [2,3].

Paper [2] describes the measurement of the concentration of radioactive aerosols close to the synchrocyclotron at a proton energy of 680 MeV. A value of 5.10^{-13} curie/litre was obtained, and it was assumed that the radioactivity of the aerosols was caused by ^{24}Na. Paper [3] investigated the radioactivity of aerosols at the CERN proton synchrotron. As the experiment was performed only a few hours after the accelerator had been shut down, only long-lived aerosols were detected: ^{54}Mn (50%), ^{7}Be (25%), ^{59}Fe (9%), ^{48}V (9%) and ^{51}Cr (7%) which were produced as a result of the erosion of one of the activated surfaces. After the accelerator had been shut down for an hour or more, the activation of the aerosols was less than 8.10^{-14} curie/litre.

Unlike paper [1] which investigated only the gaseous induced radioactivity in the air, this report studies the radioactive aerosols produced in the radiation field of the 1 GeV Leningrad synchrocyclotron [4].

Method

NEL type tapes [5] were used as a filter to collect the aerosols. The aerosols were sampled remotely as in paper [1]. The air pumping rate was 200 litres/min. The aerosol collection coefficient, which was measured by means of several filters, was 0.85 ± 0.05. The maximum time for the selection of one aerosol sample was 7 hours. The time lag until the radioactivity in the filter was measured varied from several tens of minutes up to 80 hours. Samples were taken from the accelerator hall, the experimental hall and in the open air outside the synchrocyclotron buildings.

The accelerator was run for not less than two hours before samples were taken. The extracted proton beam was monitored in the same way as in paper /1/.

A scintillation spectrometer with an NaJ(Te) crystal and a semi-conducting spectrometer with a Ge(Li) detector were used to determine the isotopic composition of the radioactivity in the filters. The nuclides ^{7}Be, ^{22}Na, ^{54}Mn, ^{60}Co and ^{24}Na, the radioactivity value of which is known to within 2.5%, were used for the absolute calibration of the spectrometers. A CBT-10 end-window counter was used to record pure beta emitters. The gauge of the counter's mica window was 1 mg/cm^2. The counter was calibrated in terms of nuclides ^{60}Co and ^{24}Na which were applied directly to the filter in the form of a solution with a known radioactivity (the accuracy of the radioactivity value was $\pm$ 3%). A correction for gamma radiation was taken into account. ^{204}Te and $^{90}Sr + ^{90}Y$ were also used to calibrate the counter. Sheets of aluminium foil, the thickness of which was selected in accordance with the data in paper /6/, were used as absorbers for the measurements with the end-window counter.

Results

Table 1 gives the main characteristics of the nuclides detected in the aerosol samples. The last column gives the equilibrium concentrations of these nuclides, measured in the accelerator building. The values correspond to an internal synchro-cyclotron current of 0.4 μA, an extracted beam of 6.10^{11} protons/sec. at a proton energy of 1 GeV and an air renewal rate in the accelerator building of η = 2.5 hours^{-1} /1/.

Table 1

Nuclides	Half-life	Type of decay Radiation energy, MeV	Concentration 10^{-14} curie/litre
^{31}Si	2.65 hours	β^- (1.47 ≈ 100%) γ (1.26 < 0,07%)	96 ± 30
^{24}Na	14.95 hours	β^- (1.39 ≈ 100%) γ (1.37 - 100%; 2.75 - 100%)	7 ± 2
^{28}Mg	21.3 hours	β^- (0.46 - 100%) γ (1.35 ≈ 73%; 0.95 ≈ 30%; 0.4 ≈ 32%; 0.032 - 100%)	3 ± 1
^{32}P	14.3 days	β^- (1.71 - 100%)	2 ± 1
^{33}P	24.6 days	β^- (0.25 - 100%)	≲ 1
^{7}Be	53.01 days	γ (0.48 ≈ 11%)	17 ± 4

The equilibrium concentration of radioactive aerosols in the experimental hall (at a proton beam intensity of $4.5 \cdot 10^{11}$ protons/sec., with a thick aluminium target and $\eta = 2$ hours^{-1}) is approximately 10 times lower. In particular, the concentration of ^{7}Be measured in the experimental hall is $(1.8 \pm 0,8) \cdot 10^{-14}$ curie/litre.

In order to explain the mechanism for the production of radioactive aerosols, a study was made of the aerosol content in "pure" argon when activated. A balloon filled with argon was irradiated in the accelerator building for control purposes. The results, consisting of the relative yield of certain nuclides, are shown in Table 2. For purposes of comparison, the relative yields of these nuclides in the accelerator building are also shown.

Table 2

Nuclide	Relative yield	
	accelerator building	argon-filled balloon $(\lambda i + \eta)\,\lambda i^{-1}$ *
^{24}Na	1	1
^{28}Mg	0.6	0.6
^{32}P	6.5	6.7
^{31}Si	3.3	3.8

*λi - decay constant of i-nuclide; η = 2.5 hours^{-1}.

The mass concentration of particles of various substances present in the air circulating in the accelerator building was measured separately. The value obtained was $\sim$ 10 $\mu g/m^3$. As the measurements show, the radioactivity of the natural aerosols is mainly caused by RaB (half-life 26.8 min.), the concentration of which does not exceed $(7 \pm 2) \cdot 10^{-14}$ curie/litre.

Discussion of results

Let us estimate the radioactivity of ^{7}Be which is produced in the accelerator building on the nuclei of oxygen and nitrogen and which is also caused by the activation of particles suspended in the air. As was shown in papers [1,7], it is sufficient to take into account activation caused by neutrons. By assuming that the production cross-section for ^{7}Be is, in the first case, 10 mbarn [8], and, in the second case, not more than 7 mbarn [8], and by using the expression [1]:

$$A = \bar{\Phi}_{eff.} \Sigma \lambda / (\lambda + \eta) \quad , \qquad (1)$$

where A is the radioactivity, $\bar{\Phi}_{eff.} \leq (7.4 \pm 1.5) \cdot 10^4$ neutron/(cm^2 · sec)[1] - the mean effective density of the neutron flux and Σ is the macroscopic cross-section, we obtain ~ $2.2 \cdot 10^{-13}$ curie/litre for O_2, N_2 and a negligible value for suspended particles. By comparing the result of the measurements from Table 1 with this estimate, it may be concluded that the production of ^{7}Be is caused by activation of oxygen and nitrogen in the air.

In the same way we shall estimate the production of ^{24}Na on the nuclei of argon and suspended particles. In this case the cross-section may be taken to be 7 mbarn [8]. We obtain $6 \cdot 10^{-4}$ curie/litre and 10^{-19} curie/litre. The value from Table 1 is in good agreement with the rated estimate for argon present in the air. Moreover, by comparing the data in Table 2, it may be seen that, allowing for errors in the measurements, both sets of values are in good agreement. Lastly, it must be pointed out that, of the nuclides included in Table 1, not one has a Z value greater than 18. Therefore, it may also be concluded that the production of radioactive aerosols (except for ^{7}Be) in the radiation field of the Leningrad synchrocyclotron is caused by the activation of argon present in the free air. In paper [9] an attempt was made to study the composition of the products from the activation of argon near a target which had been irradiated by 3 GeV protons. The authors obtained good evidence for the production of measured quantities of ^{24}Na, ^{27}Mg, ^{29}Al and ^{37}S (in gaseous form) under conditions similar to those for the activation of argon in the air near the accelerator.

Taking into account the above conclusion, let us examine which nuclides, apart from those mentioned in Table 1, may be found in the radioactive aerosols. Clearly it is sufficient to take into account ^{27}Mg, ^{28}Al, ^{29}Al and ^{30}P. The nuclides ^{11}C, ^{35,37,38}S and ^{22}Na may be ignored as the former are present mainly in a gaseous state [1], and the equilibrium concentration of ^{22}Na is negligible due to the long half-life. By assuming that $\bar{\lambda} \simeq 0.14$ min^{-1} and $\bar{\sigma} \simeq 10$ mbarn, we obtain from expression (1) the value $3.6 \cdot 10^{-12}$ curie/litre for the concentration of the above nuclides. Taking into

account the data from Table 1, the total concentration of aerosols in the accelerator building during runs does not exceed $5 \cdot 10^{-12}$ curie/litre. The drawing illustrates the dependence of the aerosol concentration on the time that has elapsed since beam switch-off. As can be seen in the drawing, the aerosol concentration in the accelerator building decreases rapidly. In the first 15-20 minutes, this decrease is mainly determined by the decay of short-lived nuclides, and from then on the major factors are emission from the accelerator building and refraction of the air coming in from outside. After about 80 minutes, the aerosol concentration drops to the natural radioactivity level.

By comparing the aerosol concentration from Table 1 with the data in paper /10/, it may be seen that the measured values are considerably below the average yearly permissible concentrations both for the accelerator building and for the shielding area, and the radiation hazard presented by aerosols is negligible in comparison with the external radiation from the induced activity of the synchrocyclotron /1/. A similar conclusion was reached in paper /3/ in the case of the CERN proton synchrotron. Moreover, assuming that the composition and concentration of radioactive aerosols during a long synchrocyclotron shut-down are the same as in paper /3/, it may be stated that a 100-fold increase in proton intensity with the same particle losses will not call for any extra precautions against radioactive aerosols.

It is interesting to estimate the effective density of the radiation fluxes contributing to the formation of radioactive aerosols. As has already been shown, the main process in this case is activation caused by neutrons. The effective mean density of the neutron flux in the accelerator hall was obtained earlier /1/. By assuming that the cross-section for the production of ^{7}Be is 10 mbarn and using the equilibrium concentration of ^{7}Be, we obtain the following value from expression (1) for the experimental hall:

$$\bar{\Phi}_{eff.} \simeq (2.5 \pm 1.2) \cdot 10^{3} \text{ neutrons}/(\text{cm}^2 \cdot \text{sec})$$

Conclusions

1. Radioactive aerosols around the Leningrad synchrocyclotron are mainly produced by the activation of argon present in the free air.

2. The concentration of aerosols in the synchrocyclotron building does not exceed $\sim 5.10^{-12}$ curie/litre during runs. The comparatively long-lived component ($T_{1/2} > 1$ hour) has a concentration of $\sim 1.3 \cdot 10^{-12}$ curie/litre and consists not of one nuclide, as was stated earlier /2/, but of several: ^{31}Si, ^{24}Na, ^{28}Mg, ^{32}P, ^{33}P and ^{7}Be.

3. The radiation hazard caused by the aerosols is negligible in comparison with the external radiation from the synchrocyclotron induced activity, and it may be ignored even at proton intensities 100 times greater.

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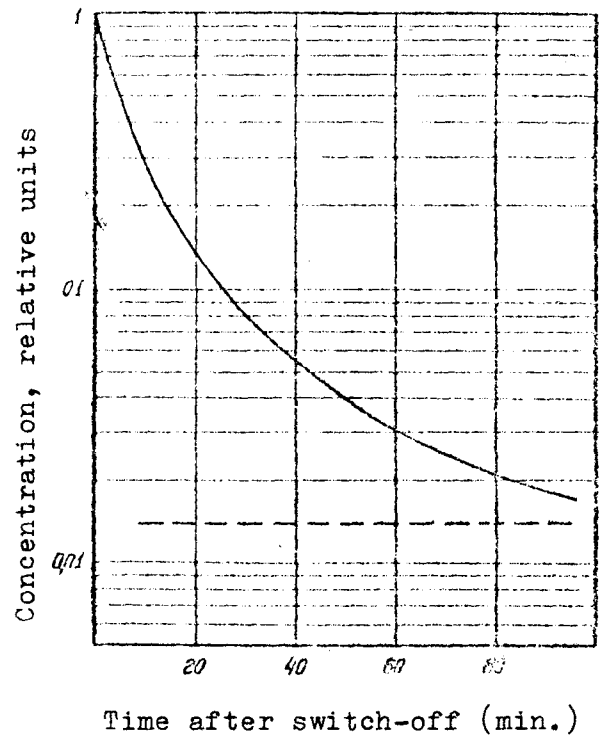

Variation in aerosol concentration after accelerator shut-down:

(———) - rated curve for accelerator building;

(----) - aerosols' natural radiation level.