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ACTIVATION OF WATER COOLING THE SYNCHROCYCLOTRAN (SELECTED PAGES)

by

M. M. Komochkov, Yu. G. Teterev





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# EDITED TRANSLATION

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\*ye initially, after vowels, and after ъ, ь; e elsewhere. When written as ё in Russian, transliterate as yё or ё. The use of diacritical marks is preferred, but such marks may be omitted when expediency dictates.

# GREEK ALPHABET

А	α	a		Nu	N	ν	
В	β			Xi	Ξ	ξ	
Г	Y			Omicron	0	0	
Δ	δ			Pi	п	π	
E	ε	e		Rho	P	ρ	
Z	ζ			Sigma	Σ	σ	ç
H	η			Tau	Т	τ	
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	A B C A E Z H O I K A M	Α       α         Β       β         Γ       Υ         Δ       δ         Ε       ε         Ζ       ζ         Η       η         Θ       Θ         Ι       ι         Κ       μ	A     α       B     β       Γ     γ       Δ     δ       E     ε       Z     ζ       H     η       Θ     θ       I     ι       K     %       Λ     λ       Μ     μ	Α       α       α         Β       β       Γ         Γ       γ       .         Δ       δ       .         Ε       ε       ε         Ζ       ζ       .         Η       η       .         Θ       θ       \$         Ι       ι       .         Κ       μ       κ       *         Λ       λ       .         Μ       μ       .	A $\alpha$ $\boldsymbol{\alpha}$ NuB $\beta$ Xi $\Gamma$ $\gamma$ Omicron $\Delta$ $\delta$ PiE $\varepsilon$ $\epsilon$ Rho $\zeta$ SigmaH $\eta$ Tau $\Theta$ $\boldsymbol{\vartheta}$ UpsilonI $\iota$ PhiK $\mathcal{H}$ $\kappa$ $\boldsymbol{x}$ A $\lambda$ PsiM $\mu$ Omega	A $\alpha$ $\alpha$ NuNB $\beta$ Xi $\Xi$ $\Gamma$ $\gamma$ OmicronO $\Delta$ $\delta$ Pi $\Pi$ E $\epsilon$ $\epsilon$ RhoPZ $\zeta$ Sigma $\Sigma$ H $\eta$ TauT $\Theta$ $\theta$ UpsilonTI $\iota$ Phi $\phi$ K $\mathcal{H}$ $\kappa$ $\kappa$ A $\lambda$ Psi $\Psi$ M $\mu$ Omega $\Omega$	A $\alpha$ $\alpha$ NuN $\vee$ B $\beta$ Xi $\Xi$ $\xi$ $\Gamma$ $\gamma$ OmicronOo $\Delta$ $\delta$ Pi $\Pi$ $\pi$ E $\epsilon$ $\epsilon$ RhoP $\rho$ Z $\zeta$ Sigma $\Sigma$ $\sigma$ H $\eta$ Tau $T$ $\tau$ O $\theta$ $\$$ Upsilon $T$ $\upsilon$ I1Phi $\Phi$ $\phi$ K $\varkappa$ $\kappa$ Chi $X$ $\chi$ $\Lambda$ $\lambda$ Psi $\Psi$ $\psi$ M $\mu$ Omega $\Omega$ $\omega$

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RUSSI	AN A	ND ENGLISH T	RIGONOMETRIC	FUNCTIONS
Edsnedl forent.		Russian	English	
		sin	sin	
		cos	cos	
		tg	tan	
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		cosec	csc	
		sh	sinh	
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		arc sin	sin <sup>-1</sup>	
		arc cos	cos <sup>-1</sup>	
		arc tg	tan <sup>-1</sup>	
		arc ctg	cot -1	
		arc sec	sec_1	
		arc cosec	csc -	
		arc sh	sinh <sup>-1</sup>	
		arc ch	cosh <sup>-1</sup>	
		arc th	tanh <sup>-1</sup>	
		arc cth	coth <sup>-1</sup>	
		arc sch	sech <sup>-1</sup>	
		arc csch	csch <sup>-1</sup>	
		orifi		
		rot	curl	
		lg	log	

# GRAPHICS DISCLAIMER

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Russian page 5.

The results of measurements of dose strength along the pipes of the cooling system are shown in Fig. 1.

Fig. 1. Distribution of dose strength along pipes of the cooling circuit.  $\blacktriangle$  - against the pipe; • - at a distance of 80 cm from the first pipe. 1 - with working accelerator; 2 - with inoperative accelerator (time after shutdown - 3.5 hours).

**KEY:** 1 - dose strength; 2 - distance from entrance to tunnel from housing I of synchrocyclotron, m.



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Russian pages 7-14

Table 1. Isotopes detected in cooling circuit of the JNRI [Joint Nuclear Research Institute] synchrocyclotron, and their activity (measured and calculated)

KEY: 1 - Isotope; 2 - Isotope activity; 3 - in circuit water, decays/s•t; 4 - measured; 5 - calculated; 6 - in residue, decays/s of sample; 7 - \*) the figures in parentheses are the deviations from the average (over all samples) value of activity in the circuit water; 8 - \*\*) activity of carbon at the outlet from the irradiation zone.

		1000 (A)		
···		E. ROC/COLLES		(6)
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(7,8)

The presence in the water samples cf isotopes other than 'Be and <sup>11</sup>C which, it is natural to assume, form from the oxygen in the water, stimulates explanation of the possible ways of contaminating the water with cobalt, manganese, chromium, and other isotopes. For this purpose we established the composition of the elements in the samples by the activation method (Table 2).

4. Discussion of the measurement results

The presence in the cooling system circuit of isotopes of cobalt, manganese, chromium, and vanadium, and also the relationships among the activities of the cobalt isotopes, give grounds for assuming that these isotopes formed as a result of the activation of the copper used for the pipes of the cooling system.

The formation of the indicated zinc isotopes in the established quantity is excluded because of the absence in the samples of radioactive zinc-65 with a half-life of 245 days. The upper limit of activity of zinc-65, established in the measurements, is 30 decays/(sol), whereas from the ratio of the formation cross sections of various isotopes of zinc [3] we should have observed zinc-65 with an activity of 300 decays/(sol). This fact, considering that there is just as much copper as zinc in the circuit water (Table 2), while the PAGE \$4

formation cross sections of the observed isotopes of copper and zinc are commensurate, also indicates that radioactive isotopes formed not from the copper in the cooling system water, but from the copper used to make the pipe of that section of the system being irradiated. The observable radioactive isotopes of cobalt, manganese, chromium, vanadium, and other radioactive and stable isotopes, not observed in the experiment, convert to water as a result of corrosion of the inner surface of the activated copper pipes. The corrosion of copper occurs more intensely under the action of radiation, and has been described in detail in [4].

The measured dependence of the values of the ratio of manganese and cobalt isotope activities on irradiation time showed that the accumulation of activity after replacement of the water in the circuit virtually ceases after several days and does not depend on the half-life for the identical element. This attests to the presence not only of the process of the conversion of radioactive isotopes into water but also the inverse process, that of the sorption of radioactive isotopes on the walls of the pipes and containers of the cooling system. Thus, with replacement of water in the system, the water can be contaminated with radioactive isotopes not only due to the radioactivity induced in part of the the pipes but also because of the desorption process. DOC = 1599

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An important argument in favor of this method of contamination of the water with radioactive isotopes that form from the copper is the fact that the activity of the water, 47 days after its complete replacement, with an inoperative accelerator was commensurate with the activity of samples taken after prolonged operation of the accelerator. On the basis of the measurement results we can describe the accumulation of radioactive isotopes in the water of the cooling circuit in analytical form.

4.1. Formation of radioactive isotopes from water oxygen

We can show that the specific activity of the i-th isotope of the j-th element  $A_{i,i}$  in the water of the cooling circuit at the output from the irradiation zone during  $\nu$  periods of water circulation in the circuit will be

$$A_{i,j} = i\sigma_{i} H \frac{\lambda_{i}}{\lambda_{i} + a_{j}} \frac{[1 - e^{-i\lambda_{i} + a_{j}}] \cdot e^{-i\lambda_{i} + a_{j}} T \cdot i]}{[1 - e^{-i\lambda_{i} + a_{j}}] T}$$
(1)

where I is the mean density of particle flow in the activation zone; •, is the formation cross section of the i-th isotope; N is the number of nuclei of the element entering into the nuclear reaction, in 1 liter (only one oxygen isotope - 160 - is used in the calculation); to is the time for the water to traverse the irradiation zone: A, is the decay constant of the i-th isotope; \*, is the extraction constant of the i-th element that forms, due to sorption, precipitation, evaporation, etc., from the water; T is the time of the water circulation period (in our case, T = 90 min).

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Formula (1) is valid for the condition  $(\lambda_1 + \alpha_1) + \alpha_2 < 1$ . For long-lived isotopes, for which  $\lambda_1 < \alpha_1$  and  $(\lambda_1 + \alpha_1) + \alpha_2 < 1$ , formula (1) acquires the following form:

$$A_{ij} = I_{i} \sigma_{j} N \frac{\lambda_{j}}{(\lambda_{j} + \alpha_{j})T}$$
 (2)

We define the value of  $\text{It}_0$ , knowing the activity of carbon <sup>11</sup>C. Assuming the coefficient  $\alpha$  small compared with the decay constant, and bearing in mind that the irradiation time t<sub>0</sub>, according to estimates, is short compared with the half-life, we get an expression for the equilibrium activity of carbon-11:

(3)

where 0,95 - 11 - - "A. " a.) T ] .

Measurement of the activity of carbon-11 in our case gave a value of  $3.48 \cdot 10^5 \text{ decays/(s \cdot l)}$ , whence It<sub>0</sub> =  $1.8 \cdot 10^9 \text{ protons/cm}^2$ .

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Knowing the activity of 'Be in the water circuit, it is easy to find, from formula (2), the beryllium extraction coefficient  $\alpha$ , which was equal to 3.4.10<sup>-5</sup> s<sup>-1</sup>, while the extraction half-life  $T_1/_2 = 6$ hours. Thus, the equilibrium concentration of 'Be is established in the circuit water in less than a day.

Using formula (2) we can determine the activity of tritium in the circuit water. The tritium extraction coefficient  $\alpha$  was calculated based on the fact that each month from the system with a volume of 36 m<sup>3</sup> there evaporates 12 m<sup>3</sup> of water; together with the water, tritium also evaporates in H<sub>2</sub>O or H<sub>2</sub>. From the calculation we have  $\alpha = 0.9 \cdot 10^{-7}$  s<sup>-1</sup>, or an extraction half-life T<sub>1/2</sub> = 63 days. The equilibrium value of the tritium activity was  $4.8 \cdot 10^{-7}$  curies/liter.

Using formula (2) we can calculate the directed activity caused by substances contained in the water and shown in Table 2. Estimates show that even if we disregard sorption of the formed isotopes, the calculated value of activity will be less than that observed by a factor of 500.

4.2. Contamination of the water by radioactive isotopes due to corrosion

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During corrosion, water is formed not only from the copper but also from the isotopes that form in the nuclear reactions, caused primarily by protons. The differential equation that describes the accumulation of radioactive nuclei in water in this case can be written in the following form:

$$\frac{da_{i,i}}{dt} = e_i \beta + e_i^{\prime} \beta_i^{\prime} - (\lambda_i + a_j^{\prime}) a_{i,j} , \qquad (4)$$

where  $\bullet_{i,i}$  is the number of nuclei of the i-th isotope of the j-th element in the circuit water; t is the time from the moment the system is filled with water;  $\bullet_i$  is the concentration of the i-th isotope in the pipes that undergo corrosion;  $\beta$  is the rate of conversion to water of the wall material of the pipes being irradiated, due to corrosion;  $\bullet_i$  is the concentration of the i-th isotope in the material sorbed on the surface of the system pipes;  $\rho_i$ is the conversion rate of the material of the j-th element from the surface of the system pipes as a result of desorption.

The solution to equation (4) is written in the form

" ... = - th, + a, to I fle, B + e' B' ) + th, + a, to' do' + B ] .

(5)

with an operating accelerator

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 $e_i = e_{0_i} e^{-\lambda_i \cdot i_j} + \frac{i\sigma_i}{\lambda_i} (1 - e^{-\lambda_i \cdot i_j});$ 

with a nonoperating accelerator  $\bullet_{1} \bullet \bullet_{2} \bullet^{-\bullet_{1} \cdot 2}$ , where I is the average proton density:  $\bullet_{1}$  is the microscopic formation cross section of the i-th isotope of copper;  $\bullet_{2}$  is the initial concentration of the i-th isotope of copper;  $t_{1}$  is the time from the moment the accelerator is turned on;  $t_{2}$  is the from the moment the accelerator is turned off. The value of coefficient  $\beta$  can be obtained using the data in [4]. The authors of this work determined experimentally the change in concentration of copper ions in a circuit mockup during irradiation with protons. The change in concentration of coppr ions in the water with time can be examined as a consequence of two processes: corrosion and sorption. The increase in copper concentration q at the first moment after water was poured into the mockup was caused only by corrosion; therefore,  $\frac{d \bullet}{d t} |_{t=0} \cdot \beta$ , for the mockup described in [4].



Fig. 2. Gamma-spectrum of dry residue of water from the cooling circuit of the JNRI synchrocyclotron, measured with a Ge(Li) detector.

KEY: 1 - Number of pulses in the channel; 2 - Channel number

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Table 2. Content of chemical elements in samples taken in the synchrocyclotron cooling circuit.

KEY: 1 - Element; 2 - Water from circuit, g/l; 3 - Precipitate from circuit walls, g/sample; 4 - Distilled water poured into circuit, g/l; 5 - copper; 6 - zinc; 7 - iron; 8 - sodium.

8200007 (1)	:Вода не :Контура, : г/д :	Осадож со стеном контура, г/проба	С.) : Алствиная : рода, за-: : Вода, за-: : изаемая : в контур, : г/а
-	2,4.10-3	7,6.10-3	<1.10 <sup>-5</sup>
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	<5.10-5	I4,8.10 <sup>-3</sup>	<5.10-5
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