ARGON ACTIVATION IN AIR AT MEDICAL CYCLOTRON RDS ECLIPSE DURING PRODUCTION OF ^{18}F

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Medical cyclotrons are widely used to produce positron emitting isotopes for positron emission tomography. High level of air activation is possible due to high neutron flux as a result of $^{18}O(p,n)^{18}F$ reaction. Reaction $^{40}Ar(n,\gamma)^{41}Ar$ with radioactive nuclide argon-41 gives main contribution in total activity of air. Value of this activity depends from neutron spectrum and geometry of cyclotron equipment. We considered various approaches for estimation of ^{41}Ar production and calculated concentration of argon-41 in vault air for cyclotron facility with inner shield containing boron carbide. FLUKA code was used for these calculations.

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1. INTRODUCTION

Positron emission tomography (PET) is one of the most effective and informative diagnostic methods in nuclear medicine. PET method is very useful for oncology, cardiology and brain studies. Production of positron emitting isotopes with short half-life times (^{18}F, ^{11}C, ^{13}N, ^{15}O and others) is needed for PET technology. Medical cyclotrons are the most appropriate for this purpose. Fluorine-18 is currently widely used and being obtained often due to reaction $^{18}O(p,n)^{18}F$. Neutrons emitted in such reaction have high energy and neutron fluence may be large enough. Therefore it is important to take into account both in design and operation periods of medical cyclotron facilities the activation, induced not only for constructive parts of cyclotrons, but also activation of air components. Nuclide ^{41}Ar gives major contribution to the total activity of air for cyclotrons with not very high energy of protons (within 10...20 MeV). Estimation of ^{41}Ar concentration in the cyclotron vault air is an important part of full scope radiation surveillance system.

2. UPPER-BOUND ESTIMATES OF ^{41}Ar YIELD

Argon-41 is the product of reaction $^{40}Ar(n,\gamma)^{41}Ar$, induced by secondary neutrons emitted from reaction $^{18}O(p,n)^{18}F$. The natural concentration of Argon-40 in the air is 0.93% (volume part). Argon-41 mainly emits β-particles with maximum energy 1198 keV and gamma rays of 1294 keV with 109.3 minutes half-life. The cross-section of neutron capture by ^{40}Ar is equal 0,66 barn for thermal neutrons and quickly falls down with increase of neutron energy up to dozens of microbarns. In the energy range between 10 keV and 1.5 MeV this cross-section has complex resonance structure. For cyclotrons without own shield some part of neutrons can be efficiently thermalized by concrete walls of cyclotron vault. These neutrons can produce in large air volume of vault the dangerous concentrations of argon-41. As a rule, simple upper-bound estimates are used for calculation of ^{41}Ar yield, where all neutrons are considered as thermal and cross section of neutron capture is large [1]. According to this approach specific activation (in $Bq \cdot cm^{-3}$) due to neutrons emitting is given by:

$$S = \phi \cdot \frac{N_A}{A} \cdot f \cdot \rho \cdot \sigma \,, \qquad (1)$$

where ρ is the density of air, f is the weight fraction composition of air of parent nuclide, ϕ is thermal neutron fluence rate, N_A - Avogadro's number, A - atomic weight of element, σ is cross section of neutron capture by ^{40}Ar. ϕ is defined as thermal neutron fluence rate inside a concrete vault:

$$\phi = \frac{1.25 \cdot Q_F}{S_\nu} \,,$$

where Q_F is yield of fast neutrons and S_ν is an internal surface area of vault. Other form of equation (1) is used also widely:

$$S = Q_F \cdot r \cdot \frac{N_A}{V \cdot V_A} \cdot f_V \cdot \sigma \,, \qquad (2)$$

where r is radius of the sphere (approximation of vault volume by sphere), V is volume of vault, V_A - molar volume, f_V - is the volume argon concentration in air. Calculations with using formulae (1) and (2) give values of ^{41}Ar mean concentrations approximately $10^6 \; Bq \cdot m^{-3}$ for medical cyclotrons with proton beam current (40...100) μA and for radius of

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vault volume nearly $2\,m$. Mean concentration of ^{41}Ar amount can reach $(10^7...10^8)\,Bq \cdot m^{-3}$ for small volumes (with radius $0.3...0.5\,m$) close to $H_2^{18}O$ water target . However these calculated values do exceed experimental estimations more than tenfold. This discrepancy results from the fact that only small part of all neutrons is thermalized by vault and cyclotron materials.

3. CONTRIBUTION OF NONTHERMAL NEUTRONS IN PRODUCING RADIOACTIVE ARGON

Some modern constructions of medical cyclotrons use built-in complex shield consisting of lead, concrete and polyethylene with boron admixtures. As an example this shield is used by Siemens cyclotrons of RDS ECLIPSE series [2]. Boron-10 has large cross-section values for thermal neutrons capture. It leads to significant suppression of thermal neutrons fluence in the air volume and using formulae (1) or (2) is incorrect in this case. Cyclotron vendor supposes level of argon activation as negligibly small, but resonance increase of cross-section in the region within $10\,keV...1.5\,MeV$ (see Fig.1) [3] and cross section of fast neutrons can give essential contribution to total argon activation.

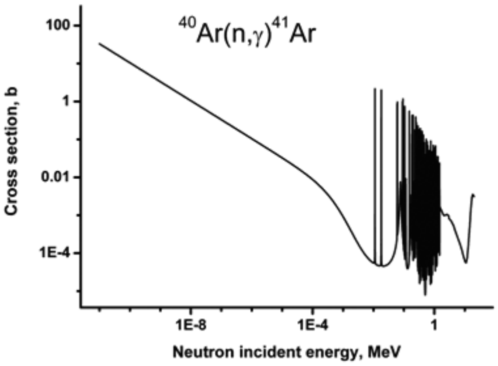

Fig.1. *Estimated neutron capture cross section for ^{41}Ar from database ENDF/B-VII.0 [3]*

This addition can dramatically depend from cyclotron and vault geometry and material composition. Monte Carlo simulations are needed for correct modeling of these processes. We estimated the contribution to argon activation of neutrons in various energy regions for cyclotron configurations with built-in shield using FLUKA code for ionizing particles transport [4]. Estimated cross section of $^{18}O(p,n)^{18}F$ reaction from IAEA database for medical applications [5] was used (see Fig.2). Values of dE/dx for accelerated protons are shown in Fig.3. We used simple model of cyclotron built-in complex shield (see Fig.4). Volume of $H_2^{18}O$ water target is $2.5\,ml$. Air volume around target is 50 liters. Next layer consists of thick high-density core cast fabricated of a mixture of lead, epoxy with admixture of boron carbide. The thickness of this layer is approximately $25\,cm$. Next layer of borated polyethylene has width $16\,cm$ and

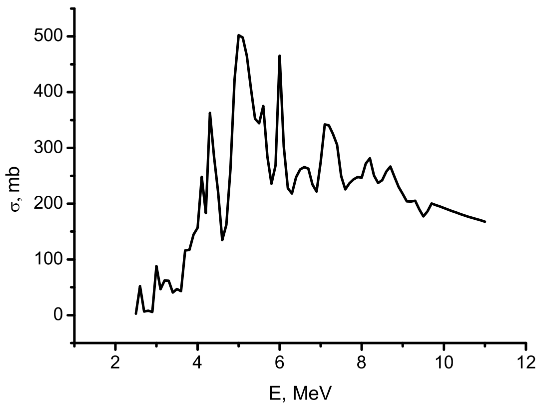

Fig.2. *Estimated cross section of $^{18}O(p,n)^{18}F$ reaction from IAEA database for medical applications*

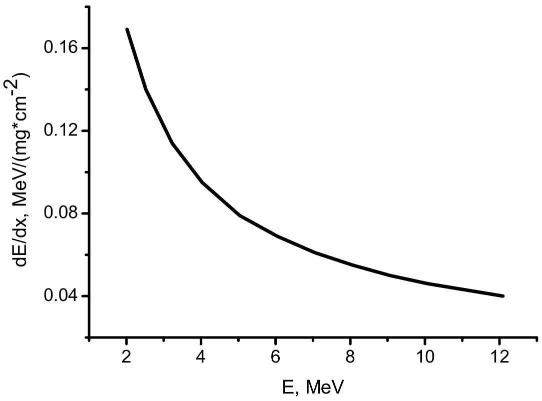

Fig.3. *Values of dE/dx in water for accelerated protons*

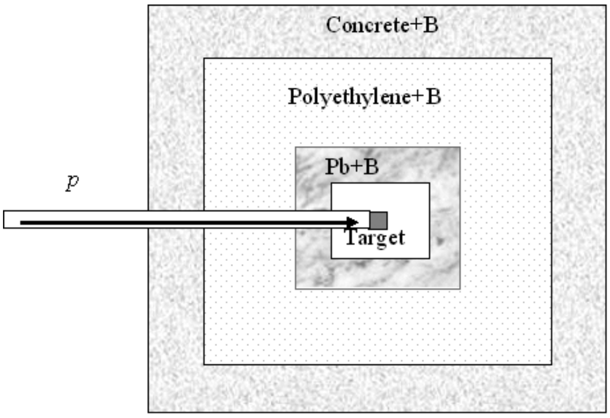

Fig.4. *Simple model of cyclotron built-in complex shield*

thickness of last layer with boron carbide loaded concrete is $60\,cm$. H_2O water target was 93 % enriched by ^{18}O. Proton current was $60\,\mu A$ with energy of protons $11\,MeV$. Argon concentration (volume part) in air was supposed as 0.00934. As a result the calculated concentration of ^{41}Ar in the inner shield volume has value not exceeding $6 \cdot 10^4\,Bq \cdot m^{-3}$. Estimation of ^{41}Ar concentration for same volume of air near the target using formula (2) comes up with much higher value - nearly $10^7\ Bq \cdot m^{-3}$.

4. CONCLUSIONS

In this paper we considered various approaches for estimation of ^{41}Ar production and calculated concentration of argon-41 in the vault air of medical cyclotron facility for ^{18}F production with inner shield containing boron carbide. FLUKA code was used for these calculations. Result shows that concentration of ^{41}Ar in air near cyclotron target for such case is not negligible and consists of nearly 0.5 % (saturation activity $6 \cdot 10^4\ Bq \cdot m^{-3}$) of yield (saturation activity $10^7\ Bq \cdot m^{-3}$) for very conservative approximation of thermal neutron fluence produced by proton beam with current 100 μA due to reaction $^{18}O(p,n)^{18}F$.

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АКТИВАЦИЯ АРГОНА В ВОЗДУХЕ ПРИ ПОЛУЧЕНИИ ^{18}F НА МЕДИЦИНСКОМ ЦИКЛОТРОНЕ RDS ECLIPSE

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Медицинские циклотроны широко используются для производства изотопов для позитронно эмиссионной томографии. При наработке ^{18}F возможен высокий уровень активации воздуха в бункере циклотрона нейтронами, которые образуются в результате реакции ^{18}O(p,n)^{18}F. Основной вклад при этом дает реакция ^{40}Ar(n,γ)^{41}Ar, в результате которой образуется радиоактивный аргон-41. Величина этого вклада зависит от спектра нейтронов и геометрических факторов оборудования циклотрона. Мы рассмотрели различные подходы для оценки количества ^{41}Ar и рассчитали концентрацию аргона-41 в воздухе внутренних полостей циклотрона, который имеет специальную защиту с содержанием бора. Для расчетов использовался программный пакет FLUKA.

АКТИВАЦІЯ АРГОНА В ПОВІТРІ ПРИ ВИРОБНИЦТВІ ^{18}F НА МЕДИЧНОМУ ЦИКЛОТРОНІ RDS ECLIPSE

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Медичні циклотрони широко використовуються для виробництва ізотопів для позитронно емісійної томографії. При напрацюванні ^{18}F можливий високий рівень активації повітря бункеру циклотрону нейтронами, які утворюються в результаті реакції ^{18}O(p,n)^{18}F. Основний внесок при цьому дає реакція ^{40}Ar(n,γ)^{41}Ar, в результаті якої утворюється радіоактивний аргон-41. Величина цього внеску залежить від спектру нейтронів та геометричних факторів обладнання циклотрону. Ми розглянули різні підходи для оцінки кількості ^{41}Ar і розрахували концентрацію аргону-41 в повітрі внутрішніх порожнин циклотрону, який має спеціальний захист із вмістом бору. Для розрахунків використовувався програмний пакет FLUKA.