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RADIOACTIVE HALOGEN YIELDS FROM THE TUNGSTEN TARGET IRRADIATED BY HIGH-ENERGY PROTONS

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ABSTRACT

In this work we present the calculation's results for radioactive halogen's formation rates and yields from the spallation and fission reactions of solid tungsten target irradiated by protons with E_p = 800 MeV and with the target current I_p= 16 mA. The influence of the secondary neutrons on accumulation these nuclides is not analyzed.

1. Introduction

A considerable amount of the radioactive nuclides are accumulated in the target due to influence of the primary-order accelerator's protons and the second neutrons arisen from the target and outside blanket as well. For the proton energy $E_p \approx 1$ GeV the tungsten target will contain the most radioactive isotopes with $Z \leq 75$ as a result of spallation and fission reactions. To forecast the composition of that radioactivity and it's dynamics is rather complicated problem. From the radiation damage effects point of view it is very important to analyze the radioactive volatile gaseous nuclides, which are able comparatively easy penetrate to the coolant, ion guide and atmosphere and thus form a gas-aerosol radioactivity and contaminate the environment. On the basis of nuclear reactor experience among these volatile compounds first of all one can find so-called RPG - radioactive precious (noble) gases (argon, krypton, xenon) and tritium, bromine, iodine, fluorine, chlorine, cesium and strontium isotopes as well. With the exception of the RPG, these isotopes are very chemically-active and able to form different kinds of chemical compounds. Hence, taking into account their chemical formations from the solid and liquid targets, the evaluation of the volatile compound's yields is a high priority and difficult problem.
On one hand, our analyses have shown that the chemical radioactive halogens (bromine, iodine, fluorine, chlorine) can form different compounds with tungsten and it would seem that this process must prevent their release from the target. However, it was rather unexpectedly to find that volatility of the tungsten's halogenides is high. So, taking into account the radiological dangerous such halogens as radioactive iodine this problem was considered in the first approach.

Keywords: Yields, Radioactive Halogens, Target, Tungsten, High-Energy Protons

2. The evaluation of accumulation of the radioactive halogens from the tungsten target under proton action.

The considered nucleuses with Z= 9, 17, 35 and 53 are mainly formed as result of fission process in tungsten target irradiated by high-energy protons. In a thick target the yield of a given nucleus per one bombarding proton can be evaluated under the formula:

$$Y_i = \rho \int_0^{E_p} \left[\sigma_i(E') \Big/ \frac{dE}{dx}(E') \right] dE' \quad , \qquad (1.1)$$

where

$\sigma_i(E')$ - reaction cross-section, which forms the i-th nucleus, depending on energy, sm^2;

$\rho = 6.32 \times 10^{22}$ sm^3 - the concentration of target nucleuses;

$\frac{dE'}{dx}(E')$ - linear density of the ionizing losses of the proton energy, MeV / sm.

In the assumption, that $\sigma_i(E')$ does not depends on energy and is equal to the $\sigma_i(E_p)$ formula (1.1) maybe written as follows:

$$Y_i = \rho \sigma_i(E_p) R \ , \qquad (1.2)$$

where

R= 30 sm - the proton path in the target.

The specific activity of the i-th nuclide a_i from the target volume (V=SR, where S - the cross-section area of the proton beam with the current I), irradiated by protons is equal:

$$a_i = \frac{6.24 \times 10^{18} I \times Y_i \left(1 - e^{-\lambda_i T_{obl}}\right)}{V} =$$

$$= 6.24 \times 10^{18} \rho \sigma_i(E_p) \frac{I}{S} \left(1 - e^{-\lambda_i T_{obl}}\right) \quad , \text{Bq/sm}^2 \qquad (1.3)$$

where

λ_i - decay constant of the i-th nuclide, s^{-1} ;

T_{obl} - the time of the target irradiation, 1 year .

The correct evaluations of cross-sections $\sigma_i(E_p)$ or the formations of the i-th nuclides are the main difficulties here. The semiempirical Tsao-Silberberg algorithm for calculation of cross-section for the formation of the nucleus with given (A, Z) was used in the case, when proton or neutron of considered energy E_p interacts with nucleus (A_0 , Z_0) of the target. The appropriate computer program SILBER was realized as PC-code. The formations of the radioactive halogens with a half-lives more than a few hours were considered. The list of that halogens; their half-lives $T_{1/2}$ and decay constants λ_i ; the values of the cross-sections σ evaluated; the specific activities a_i, calculated for the proton beam diameter equal 10 sm (S=78.5 sm); and the values of an equilibrium activity in the ion guide A^u (see paragraph 3) as well are presented in Table 1.

Table 1. The formation's cross-sections, specific activities in the target and equilibrium activities in the ion guide for some radioactive halogens

Nuclide	$T_{1/2}$	λ, c^{-1}	σ, mBn	a_i, Bq/sm^3	A^u, Ci
I-123	13.2 h. [1]	1.45 -05 [2]	0.04	3.2 +09	86
I-124	4.18 d.	1.9 -06	0.0196	1.6 +09	43
I-125	60.14 d.	1.3 -07	0.0148	1.0 +09	27
I-126	13.02 d.	0.2 -07	0.0075	6.0 +08	16
I-131	8.04 d.	1.0 -06	0.0011	8.8 +07	2.4
I-133	20.8 h.	9.2 -06	0.00055	4.4 +07	1.2
I-135	6.6 h.	2.9 -05	0.00028	2.2 +07	0.6
Br-77	57.04 h.	3.3 -06	0.095	7.6 +09	200
Br-82	35.3 h.	5.4 -06	0.0025	2.0 +08	54
Cl-36	$3.01*10^5$ y.	7.2 -13	0.085	1.54 +05	0.004

[1] h.- hours: d- days; y.-years; [2] should be read as $1.45 * 10^{-05}$

3. Evaluation of the radioactive halogen yield's rate from tungsten target into ion guide. The electronic configuration of halogen atoms brings their ability to attach easily the electrons and, thus, determines their high ability to react with design substances. This ability depends on an aggregate conditions, the pulverization degree, temperature, properties of the formed compounds and other conditions the reaction goes under. For example, such fine-fractionated metals as aluminium Al, antimony Sb, and a copper Cu are inflamed in the atmosphere of bromine vapour. Such active metal, as sodium does not interact with liquid bromine, that depends on tight surface metal film of NaBr. Without water the majority of metals does not interact with iodine and bromine [2]. As is pointed in [3] tungsten does not react with iodine and bromine vapour, to all appearance, due to very small stability of the appropriate

halogen compounds. Based on the data from [4] it appears that bromine and iodine interact with the majority of metals and the result of this interaction is the formations of the bromides and iodides. However, the tungsten and I_2 interact with those metals at T=1073°K. As for fluorine and chlorine tungsten reacts with the first even at 20 °C and with the second one only at 573 °K . In the last case the WCl_6 is formed.

The identified compounds WBr_2, WBr_5, WBr_6, WI_2, WI_4, WCl_5, WCl_6 and other are considered in [5]. All of them are the sufficiently volatile ones. However, it should be noted, that the above view on reactive properties of the halogens is valid only for their macroscopic concentrations. Being interacted with tungsten target the halogens are formed in a very small amount, their mole fractions are $\sim 10^{-8} \div 10^{-11}$. Therefore it is possible to expect that the halogen's formation will be mainly due to low-atomic compounds, for example, such compounds as WBr_2, WI_2 and other one, when the formation rate is considerably higher, than in the reactions with macro-concentrations. The analysis of a temperature data [6], [7] shows, that the pressure of the halogen's saturated vapour very strongly depends on the temperature. Some characteristics of the halogens and their compounds , including the values for pressure saturated vapour are presented in Table 2 under the corresponding temperatures.

Table 2. Some characteristics of the halogens and their compounds with tungsten

Substance	The boiling or the sublimation temperature(^{0}C) for pressure saturated vapour in mm Mercury column					The melting point temperature, ^{0}C	The dissociation temperature, ^{0}C
	0.1 mm	1 mm	10 mm	100 mm	760 mm		
F_2	-226.6(S)	-221.0(S)	-213.7	-202.6	-188.1	-219.6	
Cl_2	-133.3(S)	-118.2(S)	-101.5(S)	-71.9	-34.1	-101.3	
Br_2	-71 (S)	-51 (S)	-26 (S)	8.6	57.9	-7.3	
I_2	12.1 (S)	39.4 (S)	73.2 (S)	115.8	182.8	113.7	
WBr_2							400
WBr_5	120	169	227	300	383	276	
WBr_6							
WI_2							
WI_4							dissociated
WF_6	-89.4 (S)	-71.7 (S)	-49.2 (S)	-21.1(S)	17.7	-0.5	
WCl_2							
WCl_4							dissociated
WCl_5		114 (S)	160 (S)	217 (S)	286	230	
WCl_6	117.4 (S)	153.7(S)	197.6 (S)	255.7 (S)	336.4	284	

(S) - means, that substance is in solid state.

In a general case the maximum evaporation rate of substance in vacuum can be described by a Lengmur equation [8] :

$$G_u = 4.38*10^{-3} P_s \sqrt{\frac{M}{T}} \qquad (2.1)$$

where

P_s - the substance's suturated vapour pressure, Pa;

M - the molecular mass, kg / kilomole;

G_u - the evaporation rate, kg / m^2 s;

T - the solution temperature , ° K.

Taking into account the Raul law for an ideal solution of substance considered (in our case for the tungsten halogens solutions) the formula (2.1) maybe transformed to another expression (2.2):

$$G_u = 4.38\times10^{-3} P_s m \sqrt{\frac{M}{T}} \qquad (2.2)$$

where

m - the mole fraction of the dissolved substance.

For the substance, which is evaporated in the form of the molecules or the single atoms of radioactive samples the expression (2.2) can be immediately written as follows:

$$Q_{ij} = \frac{4.38 \times N_A P_s^j m_j x_{ij} \lambda_i}{\sqrt{M_j T}} \qquad 2.3)$$

where,

N_A - the Avogadro constant, N_A= 6,023 * 10^{23};

x_{ij} - the number of atoms of i-th radioactive isotope from the j-th compound;

λ_i - the decay constant for the i-th radioactive isotopes, s^{-1};

Q_{ij} - the radioactivity quantity of the i-th isotope for j-th compound, evaporated into vacuum from the target's surface unit during time unit, Bq/m^2 s.

Otherwise

$$Q_{ij} = \frac{6.86\times10^{13} P_s^j m_j x_{ij} \lambda_i}{\sqrt{M_j T}} \qquad (2.4)$$

where,

Q_{ij} is expressed in the units of Ci/m^2 s.

Using the expression (2.4) and the data from Table 1 on activity of the Br-82, that is for a_{82}^{Br} = 2.0*10^8 Bq / sm^3, and at target temperature t=200° C (4 73 ° K) the rate of the evaporation from the target's butt-end into ion guide of the Br-82 as a component of the molecule of WBr_5 was calculated for beam diameter d=100 mm. It was assumed, that the target is not separated from ion guide by a membrane, i.e. it is in vacuum. It was assumed also, that all bromine atoms of the WBr_5 are indeed the radioactive isotopes of Br-82 ($T_{1/2}$ = 35.3 hours). The pressure of saturated vapour for WBr_5 at 200 °C was ~ 5 * 10^2 Pa. In these conditions the evaporation rate of the Br-82 was equal ~ 2.1 * 10^{-1} Ci/m^2 s, that corresponds to equilibrium activity of the Br-82 on the ion guide surfaces due to condensed phase of the WBr_5, and $A_{82}^{u}=300Ci$.

Really, the activity changes released into ion guide is described by the equation:

$$\frac{dA_i^u}{dt} = SQ_{ij} - \lambda_i A_i^u \qquad (2.5)$$

For the equilibrium case $\frac{dA_i^u}{dt} = 0$ è $A_i = \frac{SQ_{ij}}{\lambda_i}$ (2.6)

Thus, our evaluation shows, that the evaporation rate of the WBr_5 at 200 °C is so high, that one can observe the non-conformity of the calculational results on equilibrium activity of the Br-82 on the target itself and on the surface of ion guide. Obviously, the total activity of the Br-82 on the ion guide walls should be less than total equilibrium activity of the Br-82 in a target $A_{82}^{M} = \int_V a_m dV$.

This fact permits to propose the assumption that at the target temperatures higher than 150 - 200° C the halogen's evaporation rate, including radioactive halogens, maybe equal to their formation's rate in tungsten target due to spallation and fission reactions under action of the proton beam in some effective volume of the target. We have also supposed that the halogens diffusion time through the evaporation's surface is much less than their half-lives. From the physical point of view and taking into account the proton energy loss per unit of the target length, , $\frac{\partial E_p}{\partial x}$, that effective target volume is found to be equal ~ 1 / 3 of the target volume V_R, which is correspondent to the proton path R with initial energy E_{Po}. In our evaluations the effective target volume is accepted equal V_{eff} ~ 103 sm^3. Hence, the equilibrium radioactivity of the i-th halogen will be:

$$A_i^u = a_i V_{eff} \qquad (2.7)$$

In case for the Br-82 it's A^{u}_{82} under accepted preconditions will be equal only 5.4 Cm instead of 30 Ci. The calculated values of the other radioactive halogens are presented in Table 1.

One can see, that the total halogen's equilibrium activity in the ion guide reaches 430 Ci. If to assume, that the main part of this activity is deposited on the length ~ 5 m of the ion guide by a diameter ~ 15 sm, than the total surface contamination of this part will be equal ~180 Ci /m^3.

It is necessary to note, that in case the target is separated from the ion guide by a partition and places into the atmosphere of a inert gas (or even air), the halogen evaporation's rate will be reduced approximately in 10^3 times

The similar effect was observed in the earliest works [9], [10], [11] ,[12] in the experiments on lead, lithium, lead-bismuth and polonium evaporation in the vacuum and in atmosphere of the inert gas (argon, helium) as well, conducted at temperatures ~ 250-800° C.

4.Conclusion

1. On the basis of nuclear reactor safety experience the analyses of volatile radionuclide's composition has been carried out. Such nuclides can be formed with proton impact and comparatively easy released from a solid tungsten target.
2. This analyses revealed that halogen containing radiological hazardous iodine and forming high volatile compounds with tungsten are important.
3. On the basis of Silberberg-Tsao systematic the cross-sections of the main long-lived radioactive halogens formation and their build-up were calculated after one year of the target operation. These halogens are produced mainly under target niclei fission by protons.
4. The model was developed for halogens from a tungsten target. It was demonstrated that volatility of both chemically pure halogens and their compounds with tungsten into vacuum determined by saturated vapour pressure is so high that its release rate under temperature 150-200 ° C can be assumed to be equal to the formation rate.
5. At proton energy of 800 MeV, current of 16 mA and the ion guide without diaphragm (window) the equilibrium activity of halogens in the guide reaches 400 Ci. This can result in the surface activity of the guide final part with the length of about 5 m to be equal to 100-200 Ci/m^2, that is rather sufficient value.
6. The data obtained indicate the fact that even in case of a solid target the proton beam insertion without "window" on the guide can results in its contamination with volatile radionuclides and aggravation of radiation safety issues.
7. If there is a gaseous medium with atmospheric pressure in the inner volume of target, the amount of volatile nuclides in this volume will be about 1,000 times less than that into vacuum.
8. Calculations performed are approximate and need further correction. But they clearly demonstrate the fact that the problem of volatile radionuclides release from solid target should be considered closely. In the connection with this arrangement and performance of relevant experiments to be carried out at one of accelerators now on operation seems to be quite reasonable.

5. References

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