

Influence of High Magnetic Field on Fusion Reactor Blanket Processes

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Abstract. Information about effects of intense magnetic fields (MF) on fusion reactor blanket processes, for the most part on tritium release, is summarized in this study. Methods and experimental setups were developed to investigate these effects. MF effect on tritium release from tritium breeding materials – lithium-containing ceramic pebbles and neutron multiplier – beryllium pebbles depends on the chemical form of tritium localized in the material. Tritium release at thermo-annealing of irradiated Li_4SiO_4 pebbles, Be pebbles and samples from plasma facing surface of Be tiles from JET was tested for effect of MF up to 2.4 T. In ceramic pebbles, where tritium is localized for the most part (86%) as T^+ , MF delays tritium release from the volume of ceramic grains as a result of electrodynamic (Larmor) effect. In Be pebbles, tritium is localized for the most part (85 %) as T_2 , presence of MF has little effect on thermo-annealing. However, a considerable increase in the tritium release by up to 80% can be achieved at thermo-annealing in the presence of simultaneous MF and ionizing radiation. That is related to radiolysis of localized T_2 into pairs of radicals of T^0 and MF-induced spin transformation $\text{S} \rightarrow \text{T}$ in pairs of radicals resulting in an increase of concentration of T^0 – main diffusing particles in beryllium metal. Simultaneous action of temperature, MF and ionizing radiation facilitates also tritium release from a surface layer up to 200 μm of a Be tile, where the most part of tritium is localized as T_2 . Other possible MF effects on physicochemical processes in the blanket zone – corrosion of ceramic and structural materials, tritium permeability, MF-induced e.m.f. in ceramics with ionic conductivity were also analyzed.

1. Introduction

The blanket zone (helium-cooled pebble bed - HCPB) and the first wall (FW) of the vacuum vessel will be the only structures that will be subjected to action of various factors - temperature up to 950 °C, fast neutron radiation of power to 2 $\text{MW}\cdot\text{m}^{-2}$ (about 10^{19} $\text{neutrons}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$) and intense magnetic field (MF) of 7-10 T. At the beginning of development of structures for a future fusion reactor, sufficient attention was not paid to effects of MF, which is a necessary operating factor of a fusion reactor, on physicochemical processes in the blanket zone. The main reason of that is a little energy of MF effect – about 10^{-4} eV per atom at MF of 1 T. Nevertheless, MF may affect considerably processes and chemical reactions in substances. At first, MF may change diffusion path of charged particles in condensed media similarly to the case of electron flux (the Hall effect). MF changes the spin state of pairs of radicals, triplet molecules, reactions of radicals with molecules. MF may cause singlet-triplet transformation ($\text{S} \leftrightarrow \text{T}$). As a pair of radicals in a singlet state for the most part recombines, but that in the triplet state does not react, MF changes kinetics of high-energy chemical reactions (radiolysis, reactions with oxygen, etc.). In the presence of MF, the efficiency of radiolysis of lithium-containing materials (Li_4SiO_4 , Li_2TiO_3 , Li_2ZrO_3 , etc.) – formation of radiation-induced defects and products of radiolysis (colloid lithium, O_2) increased by 20-25%. Unfortunately, the investigations were carried out only for MF of up to 1.4 T, which is far below the real operating MF [1]. Increased formation of radiation-induced defects and products of radiolysis in the ceramics in the presence of MF may facilitate tritium binding in the chemical forms (LiOT, LiT) that delay T release and form high-temperature peaks in curves of tritium release at thermo-annealing [2]. MF effects on tritium release were investigated only in out-of-pile experiments with previously irradiated materials. At present, in-pile experiments with ceramic pebbles in the presence of MF are not carried out because of technical difficulties. In order to assess properly MF effects on tritium release, it is necessary to determine chemical forms of

tritium localized in a crystal lattice of ceramics. Present physical methods are little usable as they change these forms at analysis. Only the lyomethod developed during the last years enables determining a real state of tritium in a matrix [3]. The tritium released at thermo-annealing forms in surface layers of ceramic pebbles as HT or HTO, and it does not reflect a state of tritium diffusing in the volume of grains. At present, there is no information about MF effects on surface processes of tritium release – isotope exchange, chemicondensation reactions, etc.

Investigations of MF effects on tritium release from Be pebbles and samples from plasma-facing surface of Be tiles were started recently [4]. As T^0 is a main diffusing particle of tritium in Be metal grains, but the tritium formed as a result of nuclear reactions is localized as T_2 in bubbles together with helium, there should be no delaying electrodynamic effect of MF on the tritium release. However, simultaneous action of MF and ionizing radiation at thermo-annealing had an unexpected facilitating effect – the efficiency of the tritium release of both irradiated Be pebbles and Be tiles from FW of JET increased. In this study, both previous and new results on MF effects on the tritium release of the two main blanket materials are analyzed.

2. Experimental

2.1. Materials investigated

Relevant materials for ITER and DEMO such as Li_4SiO_4 ceramic pebbles (FZK, Karlsruhe) both unirradiated (only marked with tritium in a photoneutron source) and irradiated in the EXOTIC-8 experiment (1998), unirradiated Li_2TiO_3 ceramic pebbles (CEA), Be pebbles manufactured by the Brush Wellman fluoride reduction process and irradiated in the BERYLLIUM experiment (1994), and Be tiles from JET (Culham, the UK) after DT experiments in 1989-1992 were investigated in this study. Main characteristics of the materials investigated are given in Table 1 In order to test influence of MF on radiolysis of the ceramic pebbles, radiation-induced defects (RD) and products of radiolysis (RP) were generated in the ceramic pebbles and high-temperature changes in their structure were introduced with by ELU-4 at the dose rate of 5 MeV electron radiation up to 80 MGy·h⁻¹ and temperature up to 1250 K, in a MF up to 1.7 T.

TABLE I. Characteristics of the materials investigated

Material	Size, mm	Grain size, μm	Conditions of irradiation	Activity of tritium in 1 g of the material or in 1 cm ² of surface layer
unirradiated Li_4SiO_4 pebbles	\varnothing 0.25-0.63	1-5	$T_{\text{irrad}}=290-300$ K $\Phi_n=2.1 \cdot 10^{18}$ n·m ⁻²	
irradiated Li_4SiO_4 pebbles	\varnothing 0.25-0.63	20-40	$T_{\text{irrad}}=900$ K $\Phi_n=2.4 \cdot 10^{25}$ n·m ⁻² Li-burnup 11%	0.9-1.6 GBq·g ⁻¹
Be pebbles	\varnothing 1.6-2.1	40-200	$T_{\text{irrad}}=780$ K $\Phi_n=1.24 \cdot 10^{25}$ n·m ⁻²	0.6-1.5 GBq·g ⁻¹
Be tiles	-	-	T_{irrad} up to 600 K D-T plasma	5 kBq·cm ⁻²

2.2. Lyomethod for determination chemical forms and distribution of localized tritium in blanket materials

In order to estimate and understand influence of MF on tritium transfer processes in matrix it is necessary to know in what kind of chemical forms tritium is localized. At present available methods (SIMS, laser ablation) don't allow to determine all forms in volume without their changing in the time of analysis. Tritium released and determined at thermoannealing process can't give the answer about its initial chemical forms also. Original lyomethod was developed. It is based on dissolution of a sample with constant linear dissolution rate in the presence of scavengers of chemically active particles in dissolving layer. Dissolution doesn't change the content and status of chemically active particles in solid matrix. Detection both the products transferred into solution and in gaseous phase is going simultaneously. Scavengers systems for dissolution are shown in the TABLE II, by simple equations it is possible to calculate amount of each chemical form of tritium separately.

TABLE II. CHARACTERISTICS OF THE MATERIALS INVESTIGATED

No.	Sample	Scavenger system	T ₂ , HT	T ⁺	T ⁰	T ⁻
1.	Li ₄ SiO ₄	1M NaNO ₃ , pH > 10	gaseous	liquid phase	liquid phase	liquid phase
2.	Li ₄ SiO ₄ , Be pebbles, tiles	0.4M H ₂ SO ₄ + 0.5 M C ₂ H ₅ OH	gaseous	liquid phase	gaseous	gaseous
3.	Li ₄ SiO ₄ , Be pebbles, tiles	0.4M H ₂ SO ₄ + 0.5 M Na ₂ Cr ₂ O ₇	gaseous	liquid phase	liquid phase	gaseous

Chemical forms and distribution of tritium in Be pebbles and pieces of Be tiles were analyzed in special dissolution set up (FIG.1). The samples were dissolved in a special cell blown with Ar at first; it serves as purge gas also. The amount of hydrogen evolved at the dissolution was measured by means of a thermal-conductivity detector (a catarometer), and the amount of tritium released was measured by a meter TMH 2. Only pure Ar flows through the comparison camera of the catarometer with the same flow rate (4 L·h) as through the cell of dissolution and an analysis camera of the catarometer. After complete dissolution of a Be pebble the amount of tritium transferred into the solution (in a T⁺ form) was determined by a liquid scintillation method; the effect of Co-60, Zn-65, etc. was prevented by distillation the solution.

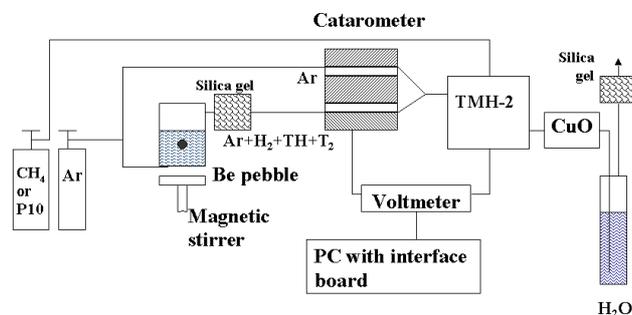


FIG.1 Scheme for dissolution experiments of Be materials

Be dissolution in 2 M H₂SO₄ only enables to separately determine T⁺. T⁰ reacts in a dissolution layer with H⁰ from the Be reaction with H⁺, forming HT, which together with the localized T₂ releases as gas, in Be isn't T⁻ form. Therefore, Cr₂O₇²⁻ ions were chosen as a

scavenger to transform T^0 into a T^+ form. In separate experiments with dissolution of non-irradiated Be, it was stated that the presence of 0.25 M $K_2Cr_2O_7$ in 2 M H_2SO_4 decreases the amount of hydrogen evolved by 87.5%. In Fig. 2, it is shown for the case of the hydrogen evolved, because the sum of the hydrogen evolved without the scavenger was assumed to be equivalent to the amount of beryllium in the pebble. The effect of the scavenger $Cr_2O_7^{2-}$ on the amount of the hydrogen evolved can be determined quantitatively from the sum of the hydrogen evolved in the presence of the scavenger and the Be pebble mass. In the case of tritium T_2 (HT), similar calculations are not possible because each Be pebble has a different initial amount of tritium. Thus, in such dissolution, the T^0 localized in Be transform by 87.5% into T^+ , and by 12.5% into HT as a result of the reaction with H^0 . Detection the chemical forms of tritium in Li_4SiO_4 ceramic pebbles were supplemented by scavenger system 1M $NaNO_3$ with $pH > 10$ where T^0 changes to T^+ . The distribution of tritium in pebbles was determined by lyomethod in the other special setup in hydrodynamic regime also, in these case pebbles were dissolved in distillate water, electroconductivity of solution was measure continuously and tritium in solution was detected discrete step by step.

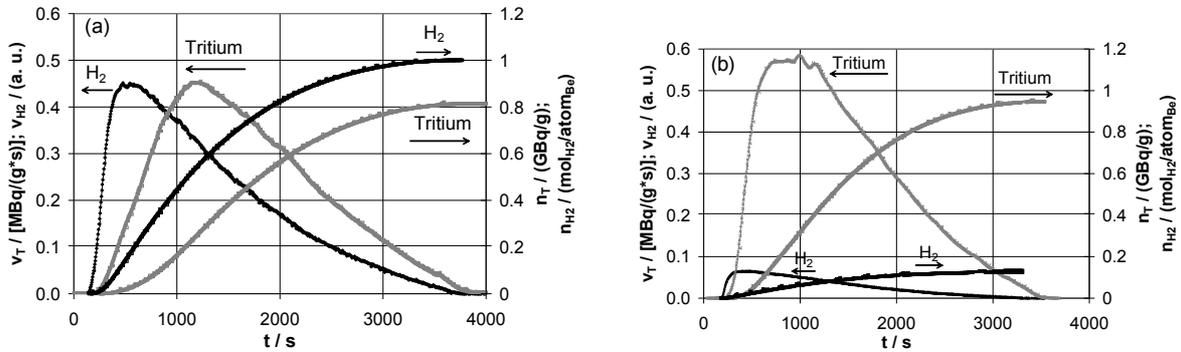


FIG. 2. Tritium release rate, v_T [MBq/(g·s)], tritium sum release, n_T (GBq/g), (both the tritium curves are grey lines) hydrogen evolution rate, v_{H_2} (in arbitrary units), amount of the hydrogen evolved, n_{H_2} (the ratio of gram-molecules of the H_2 evolved to gram-atoms of the initial pebble Be), (both the hydrogen curves are black lines) as functions of time, t (s), at the dissolution of non-treated Be pebbles: (a) in 2 M H_2SO_4 ; (b) in 2 M H_2SO_4 + 0.25 M $K_2Cr_2O_7$.

2.3. Thermoannealing method

Influence of MF on tritium release from samples was investigated in the thermal magnetic rig (FIG.3) The treatment of the samples at isothermal regime ($T = 500^\circ C$, $t = 30$ min) with and without fast electrons radiation ($P = 14$ MGy·h $^{-1}$) and magnetic field ($H = 1,7$ T) was performed in order to test an influence of different parameters on the tritium release of beryllium pebbles and tiles at thermoannealing processes. Tritium release was made separate under action of temperature (T), temperature and MF (T+M), temperature and radiation (T+R) and under all three parameters simultaneously (T+M+R). In order to estimate influence of MF of tritium release from Li_4SiO_4 separate experiments were made in another thermal magnetic rig up to 2.4 T.

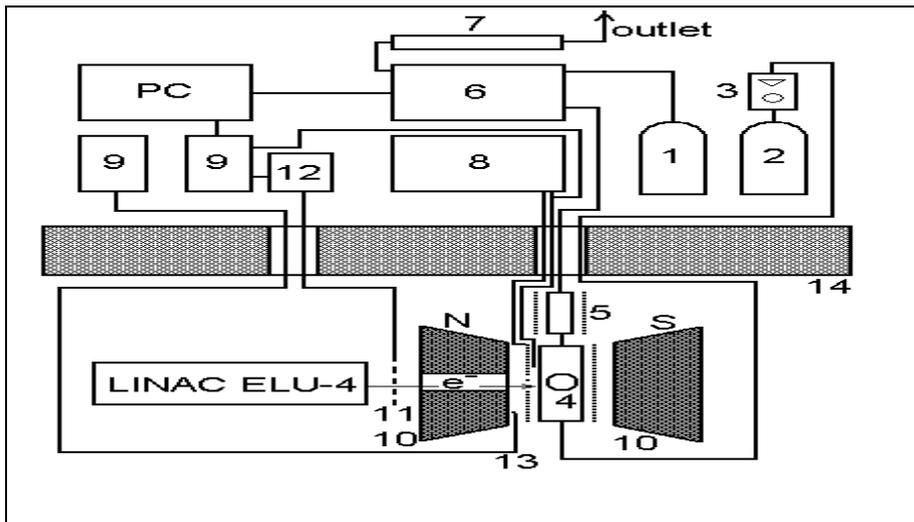


FIG. 3. Thermal magnetic rig on the base of a linear electron accelerator
1,2 gases (Ar, He, P10), 6, 8, 9,10,11,12,13 – instruments for parameters [5].

3. Results and discussion

3.1. Influence of MF on generation scavengers of tritium in ceramic materials

One of the main tasks of the HCPB is tritium breeding in lithium containing ceramic and its full release in flow of purge gas. Tritium forms in nuclear reactions of lithium with neutrons and initially it is localized in vacancies of cations as T^+ . Such a form of tritium is optimal for diffusion through a volume of grain to surface, where tritium desorption takes place by isotope exchange or chemicondensation. However, in processes of radiolysis of ceramics different radiation defects (RD) of an electron or hole type can develop (F^+ , F_o , F_n^o , centre, O^- , O_2^- , etc.), as well as radiolysis products (RP), e.g., colloidal lithium and molecular oxygen - they all can catch tritium in forms T^+ , T^o or T_2 and further develop compounds fixed with matrix – LiT, LiOT. Diffusion is inconvenienced if tritium is scavenged in such kind, as result higher temperature is necessary for thermo dissociation and for release of tritium.

Formation of RD and RP was tested by irradiation of Li_4SiO_4 pebbles in different temperatures with or without MF. That accumulation obtained of RD decreases by increasing the temperature of irradiation and at temperatures > 600 °C localization of RD in ceramics wasn't observed [6]. MF increases concentration of RD up to 20-25 % for Li_4SiO_4 and up to 15 -20 % Li_2TiO_3 (irradiation by fast electrons up 200 kGy at room temperature). RP forms at absorbed doses 100 kGy at irradiation temperature 200-300 °C, colloidal lithium accumulates most effectively. At radiolysis > 600 °C forms other RP - O_2 , Li_2O , Li_2SiO_3 . MF facilitates formation of RD and RP and in such a way can stimulate capture of tritium by matrix and necessity to increase annealing temperature and time up to full release of tritium [3].

3.2. Chemical forms of tritium localized in blanket materials

Distribution of abundance of localized tritium forms in blanket materials is significant to assess effects of influence of MF. Electrodynamics effects can affect only charged particles of tritium (T^+ , T) at jumping in diffusion process by vacancies, the effect of changing spin in radical pair can modify concentration of T^o in matrix. Four kinds of localized forms of tritium (T^+ , T , T^o , T_2) are possible in ceramics, three forms - (T^+ , T^o , T_2) in metallic beryllium. In ceramics tritium is localized in cation vacancies mainly in the chemical form T^+ , but it can be bounded as LiOT also. Distribution of abundance of chemical forms of tritium changes

negligible at thermoannealing processes for ceramics. The main localization form of tritium in Be is T_2 . Tritium is localized in microbubbles as mixture of $T_2 + He$ gases in a volume of grain. The main diffusion agent for tritium in beryllium is T^0 , which migrates by interstices of crystal lattice. The T^0 forms at thermal dissociation process of T_2 or as a result of the reaction of T^+ with electrons of band. In Be tiles the main localization form of tritium is T_2 . In a subsurface layer (up to 200 μm) under action of plasma on FW, T^+ , T^0 , T_2 form - in deeper layers tritium is localized as T^+ as a result of nuclear reactions.

TABLE III Distribution of abundance (%) of localized tritium forms

No.	Material	T^+	T^0	T_2 (HT)	T^-
1.	Li_4SiO_4	86 ± 3	----	9 ± 2	5 ± 1
2.	Be pebbles	5 ± 1	10 ± 3	85 ± 3	-----
3.	Be tiles	10 ± 2	16 ± 3	73 ± 3	-----

3.3. Distribution of localized tritium in blanket materials

There is different distribution of localized tritium in different blanket materials. The tritium concentration in separate layers of pebbles or surface layers of Be tiles was determined using the lyomethod – dissolving samples in a kinetic regime at a constant rate of dissolution, measuring gaseous tritium forms (HT, T_2) with a gas flow detector, and thickness of the layer of the matrix dissolved being determined from the electrical conductivity of the solution (for the ceramics) or from the hydrogen evolved (for Be pebbles). In the ceramic pebbles, the most part of tritium is localized in subsurface layers up to 30 μm of the pebbles. The distribution of tritium correlates with the distribution of radiation-induced defects (RD) in a pebble matrix testifying the localization of T^+ in cation vacancies. In the Be pebbles, tritium is localized for the most part in a central part of a pebble, the tritium concentration decreases sharply in a subsurface layer (*Fig. 4.*) The non-uniformity of the distribution of tritium remains after thermo-annealing of the irradiated Be pebbles.

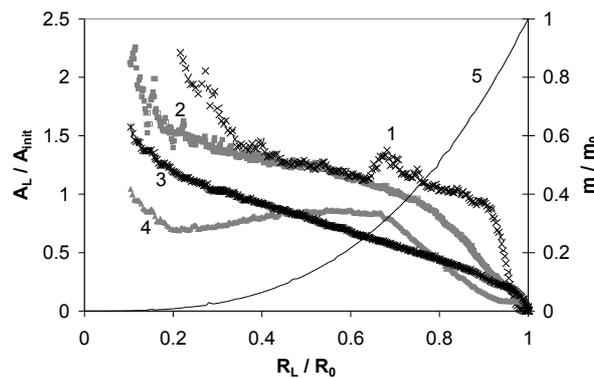


FIG. 4. Relative distribution of the localized tritium (T_2+T^0) along a radius in different Be pebbles treated under the following conditions: 1 – unannealed, $A_{init}=1373 \text{ MBq}\cdot\text{g}^{-1}$, $F_{ta}^{gas}=0$; 2 – 1123 K 2 h, $A_{init}=1076 \text{ MBq}\cdot\text{g}^{-1}$, $F_{ta}^{gas}=25\%$; 3 – 1123 K 2 h, MF 1.7 T, $P=14 \text{ MGy}\cdot\text{h}^{-1}$, $A_{init}=890 \text{ MBq}\cdot\text{g}^{-1}$, $F_{ta}^{gas}=52\%$; 4 – the same treatment as for 3, $A_{init}=900 \text{ MBq}\cdot\text{g}^{-1}$, $F_{ta}^{gas}=53\%$. A_L – tritium activity ($\text{MBq}\cdot\text{g}^{-1}$) in the dissolved layer of radius R_L . R_0 – initial radius of the pebble. 5 – the part (m/m_0) of the initial mass of the pebble, m_0 , that remained after its dissolution to mass m corresponding to radius R_L : $(m/m_0) = (R_L/R_0)$.

In surface layers of the Be tiles subjected to the action of D-D and D-T plasma in JET, gaseous forms of tritium (T_2 , T^0) are localized in a layer to 200 μm with a maximum at the distance of 10 μm from the initial plasma-facing surface. The tritium in the ion form (T^+) amounts a little part (5%) of the total tritium. T^+ is localized non-uniformly in subsurface

layers to the depth of 1-10 mm. It is a characteristic feature that, at thermo-annealing, both tritium release through the surface into gas phase and tritium diffusion into the volume of a tile take place.

3.4. Influence of of MF on tritium release at thermo-annealing

Present experimental techniques do not enable testing of MF effect on tritium release under real conditions of irradiation of ITER or DEMO blanket materials. MF effects were investigated at thermo-annealing of pre-irradiated ceramic or Be pebbles at a constant temperature or at a constant rate of temperature increase both without and in MF, without and under ionizing radiation. Fast electrons, which excite a matrix similarly as fast neutrons but do not produce tritium, were used as the ionizing radiation in these experiments. In the case of the ceramic pebbles, MF affects appreciably the tritium release [1] – in MF of 2.4 T, the efficiency of the tritium release decreases by 40%, the time of the tritium release increases. The delaying MF effect is proportional to the squared MF intensity. That correlates well with the value calculated from the lengthening of the diffusion path of T^+ in the volume of ceramic grains [1]. It is significant that the grain size of the Li_4SiO_4 ceramic pebbles increased considerably from 5-10 μm to 30-40 μm at their irradiation in the EXOTIC-8 experiment. In the case of Be pebbles, the presence of MF decreases slightly the efficiency of tritium release at thermo-annealing. That may be related to recombination of T^0 diffusing in the grain volume. The diffusing T^0 radicals, at they encounter, form pairs of radicals $T^0 \cdots T^0$ with uncorrelated spins. 75% of the uncorrelated pairs of radicals are in a triplet state, 25% of them are in a singlet state. Then at the encounter of T^0 without MF, only the singlet pairs, which are 25% of all the pairs, recombine forming a slowly diffusing T_2 . Spin transformation ($T \rightarrow S$) takes place in the presence of MF. That increases the number of the singlet pairs, facilitates the recombination of the pairs of radicals, and decreases the concentration of fast diffusing T^0 . Simultaneous irradiation in MF considerably increases the tritium release at thermo-annealing of the Be pebbles. That is related to radiolysis of the main form of localized tritium – T_2 forming a pair of radicals $T^0 \cdots T^0$ with correlated spins in a singlet state, which without MF easily recombine into T_2 . MF changes the spin state of the correlated pair of radicals ($S \rightarrow T$), the recombination may not take place, and the concentration of T^0 in the matrix increases (Fig. 5). In the Be tiles, tritium is localized for the most part as T_2 . However unlike the highly irradiated Be pebbles, tritium in the Be tiles does not form microbubbles, but occurs as separate molecules in the grain volume. Radiolysis of the separate molecules takes place more efficiently forming the singlet pairs of radicals $T^0 \cdots T^0$. There is a greater possibility to prevent their recombination and to transfer into a matrix as separate T^0 facilitating the tritium release.

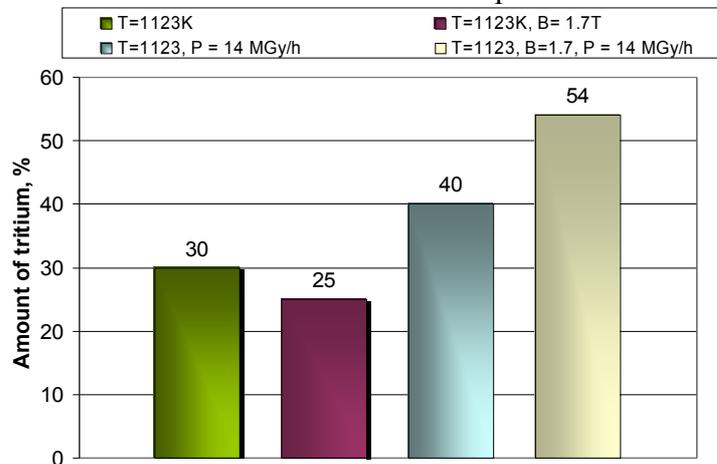


Fig.5. Release of tritium at different parameters from the irradiated Be pebbles

In the presence of MF under these conditions, MF changes the spin state of the pairs into the triplet state, all the radical pairs formed at radiolysis dissociate, and the tritium release increases considerably. The efficiency of radiolysis of gaseous T_2 is high ($G = 17$ molecules / 100 eV). However, there are no data about radiolysis of T_2 localized in a solid matrix. The tritium release at the combined thermo-annealing at a constant temperature with simultaneous MF and radiation is shown in TABLE IV

TABLE IV Effect of additional MF and radiation (R) on tritium release from Be pebbles and tiles at thermoannealing

No.	Sample	Ratio of the tritium released with MF and/or R to the tritium released only with temperature		
		T+M	T+R	T+M+R
1.	Be pebbles	0.8	1.3	1.8
2.	Be tiles	1.3	5.0	7.0

Thus, under the real operating conditions of HCPB, MF together with ionizing radiation will facilitate the tritium release from Be materials and decrease the tritium retention.

4. Conclusions

The results obtained give evidence that MF can act in two different ways on tritium release from the main fusion reactor blanket materials – ceramic and Be pebbles and on tritium release from FW Be tiles. Delaying role of MF on tritium release from ceramic pebbles with a little grain size (5-10 μm) will be negligible. Nevertheless, if the grain size in initial material is larger and radiation-stimulated grain growth up to 30-50 μm under operation conditions of blanket zone, decreasing of tritium release will be observed. As delaying effect of MF is linearly proportional to the grain size and to the squared MF intensity, then from larger grains and at real intensity of MF a part of tritium cannot release from the inside volume of grain to its surface. From previous calculations [formulu raksts] the critical grain size at MF intensity 5 T is 70 -80 μm . Influence of MF on tritium release from Be materials (pebbles, tiles) at thermo-annealing process under action of temperature is negligible, it is connected with the delaying of diffusion of the little amount of localized T^+ . MF doesn't affect T_2 and T^0 diffusion, which are the main localization forms of tritium in Be. MF simultaneously combined with temperature and radiation significantly facilitates T release. It is possibly connected with formation of radical pair T^0-T^0 in a singlet state at the radiolysis of T_2 . Under influence of MF transformation of spins (S- \rightarrow T) will take place. Pair of radicals in the triplet state cannot recombine, produced radicals diffuse in matrix and facilitates T release. In order to achieve high release of tritium, an optimization of the doze rate and MF intensity is foreseen in further investigations.

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