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Research Article

Lifetime and shelf life of sealed tritium-filled plasma focus chambers with gas generator

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Abstract

The paper describes the operation features of plasma focus chambers using deuterium–tritium mixture. Handling tritium requires the use of sealed, vacuum-tight plasma focus chambers. In these chambers, there is an accumulation of the impurity gases released from the inside surfaces of the electrodes and the insulator while moving plasma current sheath inside chambers interacting with β-electrons generated due to the decay of tritium. Decay of tritium is also accompanied by the accumulation of helium. Impurities lead to a decreased yield of neutron emission from plasma focus chambers, especially for long term operation. The paper presents an option of absorption type gas generator in the chamber based on porous titanium, which allows to significantly increase the lifetime and shelf life of tritium chambers. It also shows the results of experiments on the comparison of the operation of sealed plasma focus chambers with and without the gas generator.

PACS Codes: 29.25.-v; 52.58.Lq

Keywords: Plasma focus; Neutron yield; Tritium-filled plasma focus chambers

1. Introduction

In the area of pulsed neutron sources, a significant niche is occupied by plasma focus (PF) chambers connected as a load to microsecond pulsed current generators (PCGs). A PF chamber has two cylindrical or spherical coaxial electrodes (anode and cathode) separated by an insulator [1]. Electrodes of chambers are usually made of metal, and the insulator is made of glass, porcelain, or of Al_2O_3 as in chambers described below. After launch of PCG, an electric breakdown of the working gas along the insulator occurs in the chambers, then a plasma current sheath (PCS) is formed, which moves under the effect of electromagnetic force along the PF chamber electrodes. The discharge current increases up to a peak value by the time of PCS compression on the chamber axis. Collapse of plasma on the axis results in the formation of pinch generating penetrating emissions [2]. With deuterium (D) or deuterium-tritium (DT) filling of PF chamber flowing, D + D and D + T nuclear reactions occur in the pinch leading to the generation of neutrons with 2.5 MeV and 14.1 MeV energy, respectively [3]. The neutron yield per pulse is determined by the chamber design, the type and pressure of the working gas mixture, as well as the current through the pinch. In general, PF chambers can operate as a pulsed neutron source with 10^5–10^{13} n/pulse yield [4–6].

Dismountable PF chambers are used for research purposes. They are evacuated to a pressure of 10^{-4}–10^{-5} Torr, filled with the working gas and then activated together with PCG. Typically, after a few tens of shots, refilling of PF chambers with the working gas is needed due to desorption of impurities

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from the chamber working surfaces during PCS movement due to their heating and leakage in the dismountable connections. All-Russian Institute of Automatics (VNIIA) uses dismountable refilled chambers with a valve for laboratory research. In standard neutron generators, sealed chambers are used. However, in case of DT mixture, dismountable chambers are not usually applied due to possible leakage of radioactive tritium into the laboratory premises, although for a record yield of 10^{13} n/pulse, a dismountable chamber was used [6]. It should be noted that the DT content (typically 50/50%) of the PF chambers for neutron generation is extremely efficient since the neutron yield in this case is about one hundred times greater than that in the case of D filling due to the larger cross-section of the D + T nuclear reaction [3]. Sealed PF chambers are used for tritium filling. A disadvantage of sealed PF chambers is the accumulation of impurities in the inner volume of PF chambers over time. Presence of even small amount of impurities in the chamber (1%–5%) dramatically reduces the neutron yield and lifetime of the chamber. As for lifetime, we mean the number of PF chamber shots, after which the average neutron yield is below 0.5 from the rated value of the neutron yield defined for each type of PF chambers. The average lifetime of sealed PF chambers by VNIIA is 100 shots. In addition, the shelf life of sealed PF chambers is defined as the maintaining operability of the chamber, when they provide 0.1 neutron yield of the rated value, wherein it is allowed long storage intervals between shots. In other words, shelf life determines the chamber maintaining operability with long storage intervals of a few months or years. Shelf life of sealed PF chambers is 0.5–3 years. This value dramatically decreases the tritium half-life (12.3 years) [7] which should be the main physical reason for value of chambers shelf life. This paper describes the potential significant increase of the lifetime and shelf life of PF sealed chambers by introducing a gas generator (GG).

Today there are facilities for neutron generation based on various types of PF chambers [4–6,8]. There are three types of PF chamber geometry: Fillipov [2], Mather [9] and spherical [10]. Fillipov chambers have an anode diameter d greater than the anode height h; Mather chambers have an anode diameter d less than the anode height h; Spherical chambers have spherical anode and cathode, whose centers are biased relative to each other. The generators by VNIIA have Mather and spherical chambers as shown in Fig. 1. For illustration, chambers in Fig. 1 have the GGs, which will be discussed hereinafter.

2. Theoretical analysis of sealed PF chambers operation

2.1. Features of sealed PF chambers without gas generator

We begin the analysis with a sealed DT gas-filled PF chamber (Fig. 2a) filled with the working gas after manufacture to a predetermined pressure and sealed by cold welding. Oxygen-free copper (99.97%) is used as electrodes of the chambers. When the discharge current flows through the chamber, there is a local heating of the surfaces accompanied

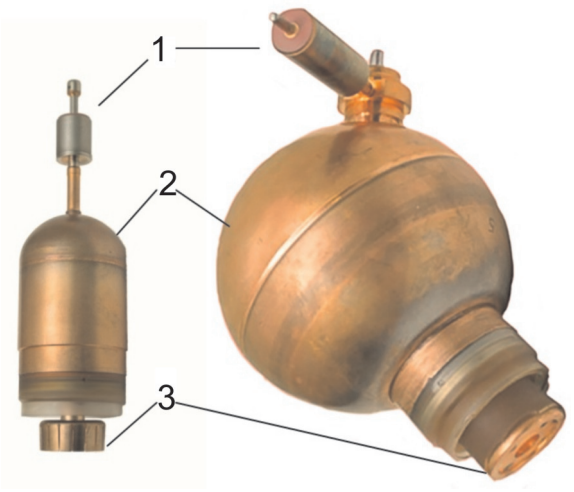

Fig. 1. System of electrodes of PF chambers: PF7 (left) and PF9 (right). 1 – gas generator, 2 – cathode, 3 – anode.

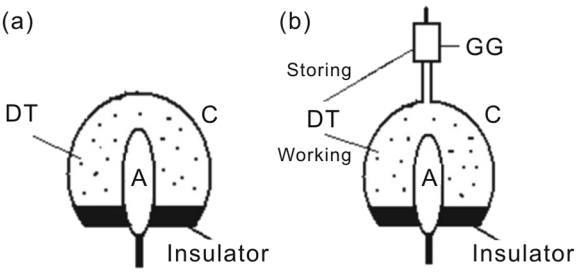

Fig. 2. (a) PF chamber without GG, (b) PF chamber with GG.

by the release of adsorbed gases from the electrodes and the insulator and their accumulation in the working gas. The experiments have shown that when the content of harmful impurities was 1%–2%, the yield decreased by 2–3 times, and when the content of harmful impurities is 10%–15%, the yield decreased approximately by 5–10 times.

When DT mixture is used as the working gas, tritium decays by the following law:

$$N_{\mathrm{T}}(t) = N_{\mathrm{T0}} \exp(-t/t_e), \tag{1}$$

where $N_{\mathrm{T}}(t)$ is the concentration of tritium atoms, N_{T0} is the initial concentration of tritium atoms, and $t_e = 17.7$ years is the time of the reduction of the number of tritium atoms by e times. Electrons of tritium β-decay $\mathrm{T}_1^3 \rightarrow \mathrm{e}^- + \tilde{\nu} + \mathrm{He}_2^3$ having a mean energy of 5.7 keV [7], bombard the surface of the electrodes and the insulator, promoting the release of gases into the chamber volume and reducing the neutron yield, as well as shortening the shelf life of PF chambers. The decay of tritium is accompanied by accumulation of helium (He) in the chamber, thus decreases the neutron yield. Change of the relative concentrations of tritium atoms $N_{\mathrm{Trel}}(t)$ and helium atoms $N_{\mathrm{Herel}}(t)$ in the volume of gas-filled PF chambers while stored for time t at the initial filling with 50/50% of D and T (initial concentrations of deuterium and tritium atoms are equal, $N_{\mathrm{T0}} = N_{\mathrm{D0}}$) can be estimated as:

$$N_{\mathrm{Trel}}(t) = \frac{N_{\mathrm{T}}}{N_{\mathrm{D}} + N_{\mathrm{T}} + N_{\mathrm{He3}}} = \frac{\exp(-t/t_e)}{2}, \quad (2)$$

$$N_{\mathrm{Herel}}(t) = \frac{N_{\mathrm{He3}}}{N_{\mathrm{D}} + N_{\mathrm{T}} + N_{\mathrm{He3}}} = \frac{1 - \exp(-t/t_e)}{2}, \quad (3)$$

where N_{He}, N_{D} and N_{T} are the concentration of He, D and T atoms in the working gas of the PF chambers, $N_{\mathrm{D}}=N_{\mathrm{D0}}$, $N_{\mathrm{T}}=N_{\mathrm{T0}}\exp(-t/t_e)$, $N_{\mathrm{He3}}=N_{\mathrm{T0}}[1-\exp(-t/t_e)]$. The calculation results of the accumulation of $\mathrm{N_{Herel}}$ and the reduction $\mathrm{N_{Trel}}$ within the chamber according to Eqs. (2) and (3) are presented in Table 1.

We assume that neutron yield $Y_{\mathrm{n}} \approx N_{\mathrm{D}}N_{\mathrm{T}} < \sigma_{\mathrm{DT}} >$ [3] (the volume of the generation and the generation time are equal). Then the relative neutron yield of gas-filled PF chambers $Y_{\mathrm{nrel}} = Y_{\mathrm{n}}(t)/Y_{\mathrm{n}}(0)$. It decreases with the storage time due to the reduction of tritium concentration. Accumulation of He in the chamber due to the decay of tritium also affects the neutron yield. The experiments on PF7 chamber have shown that the dependence of the neutron yield on content N_{Herel} in PF chamber is linear. At $N_{\mathrm{Herel}} = 6\%–7\%$, the average neutron yield reduces by one order of magnitude. The experimental function of the relative neutron yield Y_{nrel} vs content N_{Herel} can be accounted for by introducing factor $L(N_{\mathrm{Herel}}) = 0.97–0.14N_{\mathrm{Herel}}$ with reliability of approximation $R^2 = 0.98$. Then the relative neutron yield of DT chambers without gas generator Y_{nrel} is defined as follows:

$$Y_{\mathrm{nrel}} = \frac{Y_{\mathrm{n}}(t)L}{Y_{\mathrm{n}}(0)} \approx \frac{N_{\mathrm{D0}}N_{\mathrm{T0}}\exp(-t/t_e) < \sigma_{\mathrm{DT}} > L(N_{\mathrm{Herel}})}{N_{\mathrm{D0}}N_{\mathrm{T0}} < \sigma_{\mathrm{DT}} >}$$
$$= (0.97 - 0.14N_{\mathrm{Herel}})\exp(-t/t_e). \quad (4)$$

Practice shows that the lifetime of gas-filled PF chambers is several hundred shots. It is likely that improving the process of production and operation of PF chambers (cleansing surfaces, usage of different materials, changing operation modes, etc.) can increase the lifetime of gas-filled chambers. However, the shelf life of DT chambers, even without shots will not exceed two or three years due to considerable accumulation of He in the chamber.

2.2. Features of sealed PF chambers with gas generator

Next we will consider the work of PF chambers with a GG [11]. To increase the lifetime and shelf life of the PF chambers, a gas generator based on titanium was included into the chamber design (Fig. 2b). Titanium was chosen because of its high activity with respect to hydrogen, relatively low activation temperature (~800 °C), sufficient mechanical strength and irreversible absorption of gases except for hydrogen and its isotopes. The absorption of the residual gases by titanium begins at 20 °C. The absorption of hydrogen in the temperature range of 20–300 °C by far exceeds the absorption of other gases. Above 300 °C, the reverse hydrogen release starts, and active absorption of carbon monoxide, carbon dioxide and nitrogen occurs in the temperature region of 400 °C and above.

Before handling GG, it was saturated with D and T hydrogen isotopes using activation procedure [12]. Saturation takes place with coefficient $K_{\mathrm{sat}} = (N_{\mathrm{D}} + N_{\mathrm{T}})/N_{\mathrm{Ti}}$, where N_{D}, N_{T} are the number of bonded atoms of D and T, respectively, and N_{Ti} is the number of titanium atoms. Titanium adsorbs hydrogen isotopes up to $K_{\mathrm{sat}} \sim 2$. After the activation of GG, the hydrogen isotopes are in the titanium "lattice". At room temperature, GG absorbs gases from the volume of the chamber and maintains a high vacuum. Before the PF chamber shots, the GG was heated to the desired temperature by supplying the current, and the working gas was released into the inner volume of the chamber only during run time. In the chambers by VNIIA, GG allows to work at DT filling in the pressure up to 50 Torr. During operation, the working gas pressure can be adjusted by changing the current through the heater. After the chamber operation, the GG cools and absorbs the working gas and impurities that remain inside the GG all the time while the PF chamber is stored.

Three important things should be noted:

1. Tritium is in the operating element of the GG in the bound state almost all the time and therefore β-electrons from tritium decay do not affect the surface of the chamber electrodes and the insulator, thus there is no release of impurity gases into the chamber volume during storage;
2. Tritium in titanium is in the bound state at a saturation factor of 0.5. Studies have shown that at small saturation factor, helium formed by the decay of tritium remains in the bound state in the titanium "lattice" and is not released into the PF chamber volume when heated up to working temperature [13];
3. Titanium irreversibly captures heavy impurity gases like CO, NO, etc. from the chamber volume, purifying the working volume [14]. Wherein the reverse release of heavy gases occurs at higher temperature than for hydrogen isotopes, so the heavy impurities remain in titanium in the bound state during the heating of GG titanium to the working temperature (release of DT mixture) [15].

These facts contribute to increasing the lifetime and shelf life of the PF chambers.

Use of the gas generator allows to maintain constant pressure and constant concentration of hydrogen isotopes in the chamber volume during the whole period of its operation. When the concentration of D and T goes down, the heating of saturated titanium in GG results in additional release of D and T. Maintaining the pressure and concentration in the PF

Table 1
N_{Herel} and N_{Trel} contents within PF chamber during storage time.

Storage life (years)	Concentration	
	$\mathrm{N_{Trel}}$ (%)	$\mathrm{N_{Herel}}$ (%)
0	50	0
1	47.3	2.7
2	44.7	5.3
3	42.2	7.8
4	39.9	10.1
6	35.6	14.4
9	30.1	19.9
12.5	24.7	25.3
18	18.1	31.9

chamber is performed for the operation in a time-matched mode. Pinching time at fixed charging voltage of storage capacitor is determined by the gas pressure and should be close to the point of time of reaching the maximum current to obtain the maximum neutron yield. Release of hydrogen isotopes from the GG can be described by function $K(t)$, which compensates for the decrease in the tritium concentration due to its decay. Therefore, the gas concentration in the chamber with GG remains constant and is equal to the initial concentration $N_0 = N_{D0} + N_{T0}$ ($N_{D0} = N_{T0}$). We assume that function $K(t)$ is equal for D and T. We can write

$$N = [N_{D0} + N_{T0}\exp(-t/t_e)]K(t) = 2N_{D0}, \tag{5}$$

hence

$$K(t) = 2/[1 + \exp(-t/t_e)]. \tag{6}$$

Deuterium and tritium concentrations in the chamber volume are

$$N_D(t) = N_{D0}K(t) = N_0/[1 + \exp(-t/t_e)], \tag{7}$$

$$N_T(t) = N_{T0}\exp(-t/t_e)K(t) = N_0\exp(-t/t_e)/[1 + \exp(-t/t_e)]. \tag{8}$$

Using Eqs. (7) and (8), the relative change of the neutron yield $Y^*_{n_{rel}}$ (t) over the storage time in the case of using GG can be written as:

$$Y^*_{n_{rel}} = \frac{Y^*_n(t)}{Y^*_n(0)} = \frac{4\exp(-t/t_e)}{[1 + \exp(-t/t_e)]^2}. \tag{9}$$

The calculation of the relative neutron yield $Y_{n_{rel}}$ for PF chamber without GG (Eq. (4) with and without coefficient $L(N_{Herel})$ of effect of He impurity) and $Y^*_{n_{rel}}$ with GG (Eq. (9)) is found in Fig. 3. Curve 1 in Fig. 3 shows that the neutron yield $Y^*_{n_{rel}}$ (t) for chambers with GG decreases by only 5% over 6 years, and 10% over 12.5 years. The determining factor for a sharp decline of $Y_{n_{rel}}$ for chambers without the GG is the accumulated He, as demonstrated by comparing curves 2 and 3. In general, these calculations lead to the conclusion that the introduction of GG into the PF chambers could greatly increase their storage time.

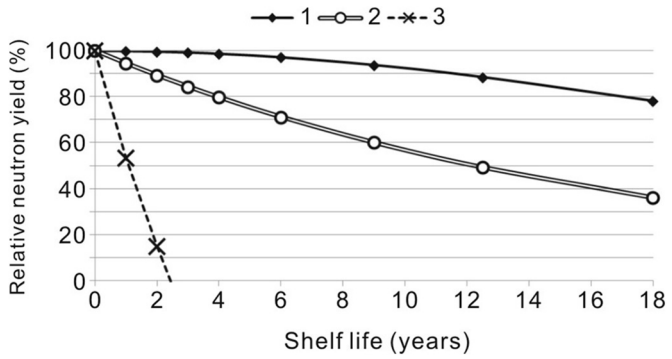

Fig. 3. Calculated function of relative yields of sealed PF chambers vs. storage time: 1 – $Y^*_{n_{rel}}$ for chamber with the gas generator, 2 – $Y_{n_{rel}}$ (excluding accumulation of He) for chambers without gas generator, 3 – $Y_{n_{rel}}$ (considering accumulation of He) for chamber without gas generator.

3. Experimental studies of sealed PF chamber operation with and without gas generator

Experimental studies of sealed PF chamber operation with and without the gas generator were conducted on two neutron generators: ING-103 and ING-104 [16]. PF7 chamber was used in the ING-103 generator (see Fig. 1a), which when filled with DT gas provided an average neutron yield of 10^{10} n/pulse. PF9 chamber was used in the ING-104 generator (see Fig. 1b) with an average neutron yield of 10^{11} n/pulse. These chambers were subjected to endurance tests, and were tested after long storage periods up to 10 years. Neutron yield at each shot was measured by SIVN61 neutron measure device based on silver activation. Instrument error of measuring neutron yield of this unit is <20% with a confidence level of 0.95.

Experimental results with PF7 sealed chamber without the gas generator are shown in Fig. 4. It is evident that the average neutron yield went down by 50% after about 100 shots of the chamber. By the 200th shot, the average neutron yield went down by an order of magnitude, and the PF chamber became basically unsuitable. This result was confirmed at another two PF7 chambers: for each chamber after 200 shots, the neutron yield went down by at least 3–5 times.

PF7 sealed chamber with GG was enabled to perform 1000 shots with the detection of neutron yield. After that, it retained its operability at half of the rated neutron yield of 1.0×10^{10} n/

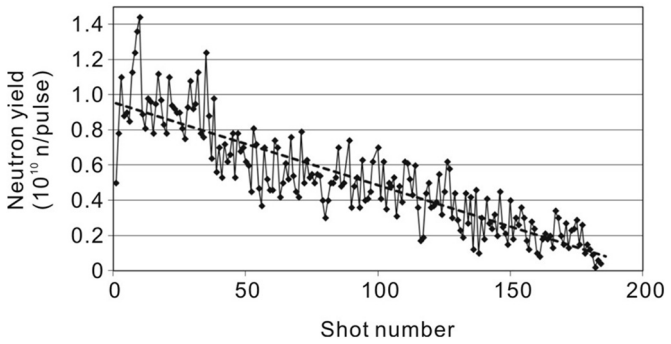

Fig. 4. Neutron yield as a function of the number of shots of PF7 sealed chamber without GG.

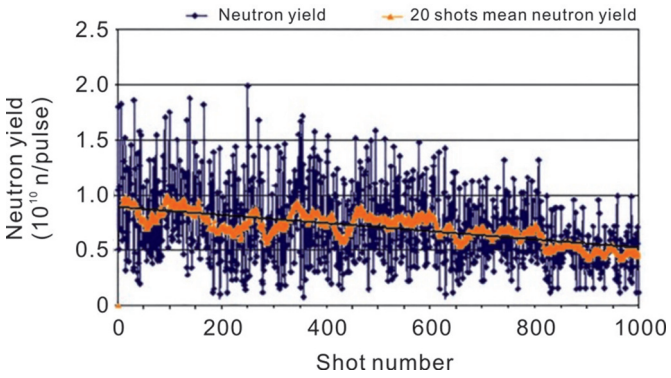

Fig. 5. Neutron yield as a function of the number of shots of PF7 sealed chamber with GG.

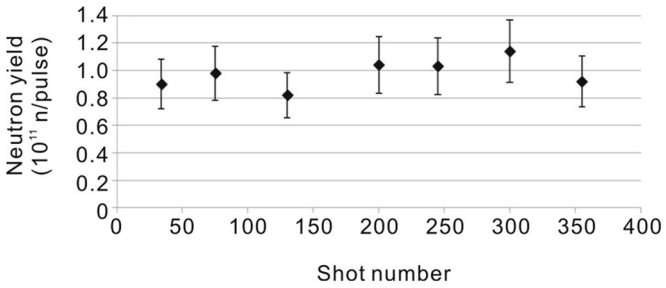

Fig. 6. Neutron yield as a function of number of shots of PF9 sealed chamber with GG.

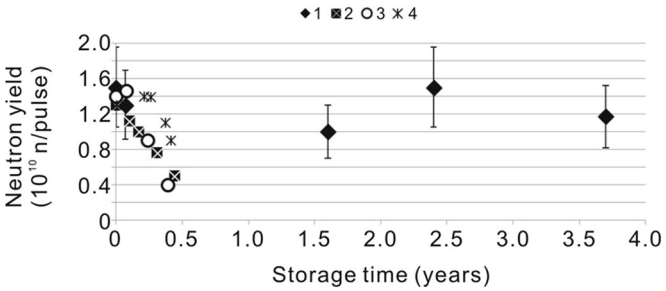

Fig. 7. PF7 chambers shelf life with (1) and without GG (2,3,4).

pulse. Neutron yield as a function of the number of shots of PF7 chamber with GG is shown in Fig. 5. The experimental data allow to estimate the lifetime of PF chambers with GG as 10^3 shots.

The change of PF9 chamber neutron yield with GG when filled with DT within ING-104 is shown in Fig. 6. After 350 shots, neutron yield did not change, and no trend towards its decrease was observed, meaning that the chamber might also have a lifetime of at least 500–1000 shots.

As it was justified above, inclusion of GG into the PF chambers could lead to increased lifetime and shelf life of chambers. Average neutron yields of a batch of PF7 sealed chambers with and without GG as the function of storage time are presented in Fig. 7. Each point on the graph represents the average neutron yield in a series of at least 10 shots. The shelf life of PF7 chambers without GG can be estimated as ~6–12 months; as for PF7 chambers with GG, it is about 5–7 years. As a confirmation of the 5–7 years shelf life for chambers with GG, we present the results of the neutron yield measurement performed in 2016 on PF7 chambers with DT content manufactured in 2007–2010. PF7 chambers with a shelf life of 6–8 years showed an average yield of $(0.9 \pm 0.2)\times10^{10}$ n/pulse; as for the ones with a shelf life of 8–9 years, the average yield is $(0.5 \pm 0.2)\times10^{10}$ n/pulse. The initial neutron yield of these chambers was $(1.4 \pm 0.3)\times 10^{10}$ n/pulse. A similar result was obtained for PF9 chambers. In December 2016, the neutron yield was measured for a batch of four PF9 chambers made in 2006–2008. All four chambers proved workable and had average yields of 8.5×10^{10}, 8.9×10^{10}, 11.9×10^{10}, 9.7×10^{10} n/pulse, corresponding to the average neutron yield for the chambers of this type at the time of manufacture within the error range. Additionally, in this experiment series, deuterium filled (2005–2006) PF9 chambers with GG were investigated. The fact that the decrease of the neutron yield hasn't been observed means that PF9 chambers have high quality surface cleaning from impurities. That provides the chamber properties unchanged for 10 years.

It should be noted that the experiments results demonstrate that PF9 chambers had longer shelf life than PF7 chambers. This might be due to the fact that during the transition from PF7 chamber to PF9 chamber, the internal volume filled with the working gas increased to a greater extent (~6.5 times) than the surface of the electrodes and the insulator (~4 times). Therefore, the release of impurities has lower effect on the neutron yield.

4. Conclusion

The results of the studies of sealed DT filled PF chambers show that the introduction of the gas generator based on the porous titanium increases the lifetime of the chambers from 100–200 to 500–1000 shots, and the shelf life from 0.5–2 years to 8–10 years or more. The gas generator allows to provide a wide range of working gas pressure (up to 50 Torr) in the PF chambers of various design and high vacuum level in the electrode gap of the chambers (up to 10^{-5}–10^{-6} Torr). In general, the introduction of the GG made it possible to use DT filled PF chambers as a reliable, safe source of neutrons with 14 MeV energy and 10^7–10^{12} n/pulse neutron yield range.

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