Characterization of low output portable neutron generators operated at FNSPE CTU in Prague

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***Abstract*—Low output portable DD and DT neutron generators are used at Faculty of Nuclear Sciences and Physical Engineering of the Czech Technical University in Prague for variety of research and educational applications. The paper summarizes some acquired experience related to characterization of these devices. Specifically, it includes determination of the total neutron generator emission, dependence of neutron emission on operational parameters, and characterization of pulsed mode operation. Whereas for ca. 1E8 n/s DT neutron generator, the total neutron generator emission has been estimated via foil activation technique, for 7E6 DD neutron generator the manganese bath technique has been applied. Further, the pulsed mode operation has been analysed over wide range of frequencies utilizing in-house developed set-up based on Red Pitaya Stem-lab. The verification of the set-up for the purpose is provided as well as the discussion of using epithermal (cadmium covered thermal neutron gas-filled detector) and fast neutron detector (of diamond type) for the purpose. Finally, the discussion is provided on measurement of neutron output dependence on operational parameters, as well as on self-activation of the devices and characteristic X-ray produced during their operation.**

***Keywords* —portable neutron generators, manganese bath technique, pulsed neutron source characterisation.**

I. Introduction

Portable electronic neutron generators have found many applications in training, research and industry [1]. At CTU in Prague, they are used for studies related to nuclear material characterisation techniques [2], to drive a subcritical reactor [4], and study its reactivity [4], or its kinetic parameters [5], or as neutron irradiator for training purposes [6] and for training purposes.

Optimizing their use for applications is connected with understanding of their characteristics which depends on device design. As they are electronic devices it is worth to check their parameters by measurements. The parameters to be verified typically include the total emission rate (and eventually its space profile, or flux at given irradiation position), dependence of emission rate on the set-up of high voltage and beam current, characterisation of pulse mode operation, i.e. of time profile of neutron emission during neutron pulses and stability of pulsing frequency, and as well the byproduct radiation caused, i.e. x-rays and self-activation.

II. Materials and Methods

A. Portable neutron generators at CTU in Prague

At CTU in Prague, two portable neutron generators from Thermofisher Scientific are used: the DD neutron generator model P-385 [7] and DT neutron generator of model MP320 [8]. Both devices are rugged, portable electronic devices designed for laboratory or field applications.

The systems are comprised of two main components: the accelerator assembly and the electronics enclosure. These components are connected to each other via a set of cables that are used to send control and monitoring signals. The accelerator assembly and the electronics enclosure are usually mounted separately; however, they may be physically connected using a mounting plate or other hardware, if desired. A warning lamp and HVPS Disable box must also be connected to the system for the system to operate. The High Voltage Power Supply (HVPS) used to accelerate ions is integrated into the accelerator assembly. The system can be completely controlled using a standard RS 232 serial communications interface with a simple terminal program or with the supplied LabWindows Graphical User Interface (GUI) program. Technical parameters of both devices are summarized in table I.

Table I
Basic technical data of used neutron generators [7, 8]

Parameter	DD model P385	DT model MP320
Neutron yield (n/s)	7E6	1E8
Neutron energy (MeV)	2.4	14.1
Max. accelerator voltage (kV)	130	90
Max. beam current (μA)	70	70
Frequency range	250 Hz to 20 kHz, continuous	250 Hz to 20 kHz, continuous
Duty Cycle	5% to 100%, 5 μs pulse width minimum	5% to 100%, 5 μs pulse width minimum
Tube length/diameter (mm)	700 /101.6	558.8/120.6

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B. Manganese bath and foil activation techniques

For very low output portable neutron generator, it seems reasonable to select different approaches for DD and DT tubes to determine the total neutron output. For DT generators the combination of higher neutron production and 14 MeV neutrons enabling use foil activation technique utilizing properly selected threshold reactions. Contrary, 2.45 MeV neutrons from DD generators has not high enough energy to utilize threshold reactions (115In(n,n')115m might be one of very few candidates) and together with significantly lower neutron output it would be doubtful to get reasonable precision even with thick activation foils. Thus, two experimental methods were used to measure the intensity of neutron production by the neutron generator: foil activation method for DT neutron generators and manganese bath method for DD neutron generator. Both methods are based on the similar principle, with the help of a suitable gamma spectrometric system, the activity, respectively neutron reaction rate of the material irradiated by the investigated neutron source is determined. Contrary to utilizing threshold reactions, the manganese bath technique is based on moderation and absorption of vast majority of neutrons emitted from the source and measurement of volumetric activity of the bath. For both methods the irradiated materials were analysed in the NAA laboratory equipped with the semiconductor HPGe (high purity germanium) detector utilised for gamma spectrometric measurements. It is Canberra-Packard HPGe detector (model GC2518) with relative detection efficiency of 25 % and energy resolution of 1.8 keV. Analysis of gamma spectra was performed using Genie 2000 Spectroscopy Software. The matters of absolute detection efficiency ε_γ calibration for relevant source-to-detector distances were performed earlier [9, 10], for thick foils the Labsocs SW was use for the purpose [11]. The reaction rate (RR) which equals to saturated activity can be obtained using the equation ([7]):

$$RR = \frac{S_\gamma \lambda^{t_{real}/t_{live}}}{N_0 \varepsilon_\gamma I_\gamma (1-e^{-\lambda t_{irr}}) e^{-\lambda t_{cool}} (1-e^{-\lambda t_{irr}})}, \quad (1)$$

where S_γ is the area under the full energy peak (FEP) provided by gamma spectrometric system, λ the decay constant of radionuclide, ε_γ the absolute detection efficiency for the FEP, I_γ the intensity per decay, t_{irr} and t_{cool} the irradiation and cooling time, t_{real} and t_{live} the real and live time of the spectrometric measurement and both times are also provided by gamma spectrometric system.

In foil activation technique, the neutron generator emission rate was determined through activation foil saturated activity and comparison with expected activity at given position as calculated by the MCNP6 code [12] and taking into account the spatial distribution of neutron emission based on [13]. Three foil materials were used Al, Nb and Cu. The foils were put to the distance of 7.5 cm from outer tube of neutron generator at 90° from target plane. During measurement, foils were put directly on HPGe surface. Tab. II shows the reaction channels used as well as the products half-lives and energies and intensities of measured gamma lines. Fig. 1 shows the cross sections for these reactions.

Obviously, the most desirable would be reactions with threshold rather closely below 14 MeV to avoid contributions from scattered neutrons, with high cross section and reasonable half-live to get enough counts at the detector, and flat profile of cross-section around 14 MeV. Many candidates have been tested for the purpose, see e.g. [14]. In this work we used Al-27 having flat cross section around 14 MeV and short half-live, copper foil which is considered to be kind of standard for the purpose [15, 16, 17], and Nb-93 with high threshold. Foils had diameter of 25 mm, and thickness of 1 mm. The aluminium foil had weight of 1.38 g and purity of 99.999 %, copper foil weighted 4.4 g with purity of 99.9%, and niobium foil weighted 4.21 g and had purity of 99.9%.

TABLE II
PARAMETERS OF USED REACTIONS

Reactions	Product half-live (s)	E_γ (keV)	Intensity %
Al27(n,α)Na24	53989	843.7	71.8
Al27(n,p)Mg27	567	1368	100
Nb93(n,2n)Nb, m92	10.15 days	934.4	99.07
Cu65(n,2n)Cu64	45724	511(annih.)	35.2

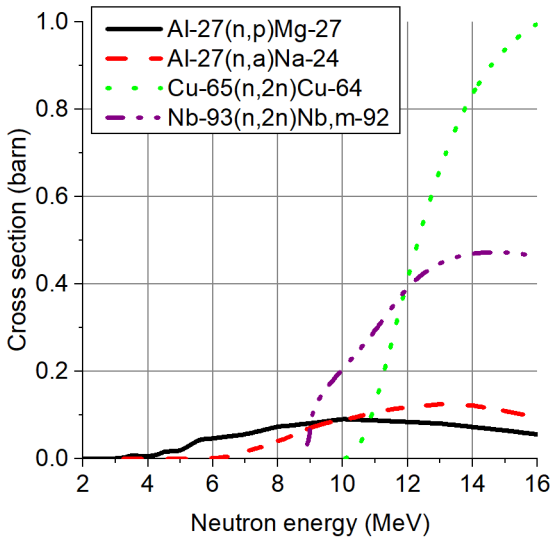

Fig. 1. Reaction cross sections used for DT generator neutron emission rate determination

For the estimation of neutron emission rate we used the determination of saturated activity of the foils based on measurement, calculation of the foil saturated activity per one source neutron in MCNP with the assumption of isotropic emission and neglecting generator internals and surrounding material, and the difference in neutron flux at 90° between models based on isotropic neutron emission and that taking into account generator internals as published in [13] (the difference for the given position reaches ca 20 %).

The manganese bath technique is a proven technique used usually for calibrating radionuclide neutron sources, e.g. [18, 19, 20], although to the knowledge of the authors its use for determination of neutron generator emission rate has not been

published. It is based on the determination of the activity of Mn-56 produced by the irradiation of manganese sulphate with neutrons emitted from calibrated source. The resulting emission rate of the source is determined from the manganese activity and the efficiency of the manganese bath. The manganese bath utilized for the experiment consists of a spherical plexiglass tank filled with an aqueous solution of $MnSO_4$. The aqueous solution of $MnSO_4$ has the density 1.21 g/cm^3 and the concentration of manganese sulphate 0.215 g per gram of aqueous solution. The total volume of the bath is 163.3 litres. The bath (see Fig. 2) was designed as a simple device; It consists of two plexiglass hemispheres, which are water-tightly connected by screws and silicone glue. The sources are manually introduced into the centre of the bath using a dry plexiglass channels. The channels are inserted through the neck at the top of the bath. The channels are optional according to the neutron source size, so that it is placed in the centre of the bath. The bath can be moved using the transport trolley on which is mounted. Mixing of the activated solution is performed by bubbling compressed air through the tank. The dump valve located at the bottom part of the tank serves for sampling of the activated solution. Activated solution is taken into the Marinelli bakers.

The neutron generator tube was inserted into the bath using a special dry channel, so the position of the generator target corresponds to the centre of the manganese bath. The generator produced neutrons inside the bath for more than 21.5 hours. Substantial number of the source neutrons were captured by manganese nuclei forming Mn-56, which β decays to Fe-56 with a half-life of 2.579 hours:

$$Mn55 + n \rightarrow 56Mn \rightarrow 56Fe + \gamma + \beta - , \qquad (2)$$

The energy of gamma rays emitted during the decay process of Mn-56 ranges from 846.77 keV to 3369.60 keV. The activated solution of $MnSO_4$ was taken into two Marinelli bakers after the source switching off.

The activity of the samples was determined based on the measurement by HPGe gamma spectrometric system using the equation 1. The analysis of the irradiated samples was done for three energies of gamma radiation with the highest intensity I_γ these are 846.771 keV (I = 98.9 %), 1810.772 keV (I = 27.2 %) and 2113.123 keV (I = 14.3 %). The resulting activity A_{bath} was determined as a weighted average of the activities corresponding to the observed gamma-ray energies:

$$A_{bath} = \frac{\sum_{i=1}^{n} \frac{A_i}{s_i^2}}{\sum_{i=1}^{n} \frac{1}{s_i^2}}, \qquad (3)$$

where A_i is the activity determined for the gamma-ray energy E_γ^i and s_i is corresponding standard deviation. The neutron emission rate B (s^{-1}) was obtained from the activity A_{bath} and bath efficiency ε_{bath} using the equation:

$$B = \frac{A_{bath}}{\varepsilon_{bath}}, \qquad (4)$$

ε_{bath} reflects neutron losses by absorption (except for manganese) and by leakage from the bath. It can be described by equation (ref):

$$\varepsilon_{bath} = f\,(1 - O - S - L)\,, \qquad (5)$$

The coefficient f is the fraction of thermal neutrons captured by manganese as opposed to those captured by other nuclei. The coefficient O represents the fraction of neutrons lost by (n,α) reactions in oxygen, and (n, α) and (n,p) reactions in sulphur. The coefficient S represents the fraction of source neutrons absorbing by the construction materials surrounding the source (stainless steel capsule, dry channel). The coefficient L describes the fraction of source neutrons escaping the bath. The efficiency of the bath was determined based on the simulation using MCNP code.

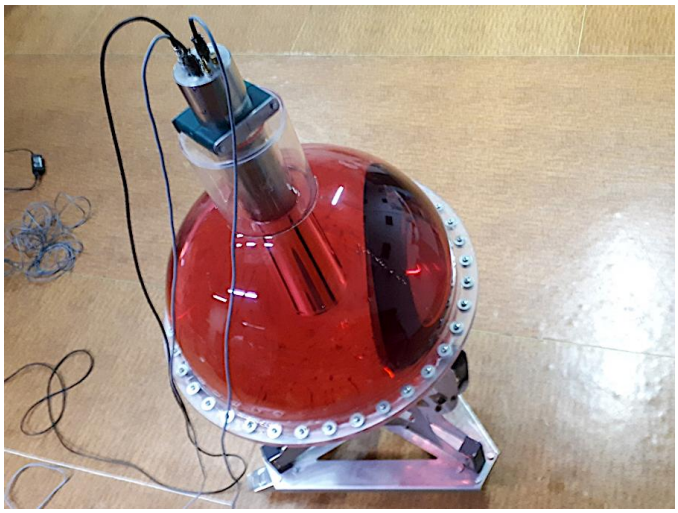

Fig. 2. Manganese bath with installed neutron generator

C. Measurement of dependence on operational parameters

Measurement have been performed for DD neutron generator with fast neutron detector (ZnS(Ag) encapsulated in resin, i.e. using proton recoil reaction) and DA310 analyser. The detector was operated in pulsed mode and have been located 10 cm from the source at 90° from deuteron beam at target plane. Dependence on beam current and high voltage has been measured in the range from 20 µA to 70 µA, and from 60 kV to 130 kV respectively. The time of measurement has been adjusted to reach relative statistical uncertainty of all values at 1%.

D. Characterisation of pulse mode operation

For characterisation of neutron generators operation in pulsed mode Red Pitaya – STEMlab device with in-house SW that evaluates TTL signal from NG ion source is used, either together with output of neutron detector, or standalone to characterize stability of pulse frequency. Diamond detectors have been used for characterisation of whole pulsing range of DT neutron generator [21]. Alternatively, low frequency range (250 Hz and lower) can be also characterized by cadmium covered thermal neutron detector, typically gas-filled one [22].

E. Self-activation, x-ray production

Self-activation and x-ray production of neutron generators have been evaluated using portable HPGe detector. Dose rates after operation have been determined using Rados RDS-120 radiation survey meter.

III. Results

A. Foil activation technique to determine the emission rate

The measurement was performed at maximal allowable parameters of the tube, i.e. 90 kV and 70 μA. Parameters and results of the foil activation measurement are summarized in Table III.

Table III
Parametres and result of the foil activation measurement (uncertainties in brackets)

	Al27 (n,α)	Al27 (n,p)	Al27 (n,α)	Al27 (n,p)	Nb93 (n,2n)	Cu65 (n,2n)
Activation time (s)	3600	3600	3600	3600	3600	3600
Cooling time (s)	300	300	300	300	300	7588
Measurement time (s)	4417	4417	75610	75610	487.8k	176.7k
Net peak area (counts)	2295 (50)	4824 (70)	24942 (160)	4882 (100)	63275 (252)	4882 (100)
Saturated activity (Bq)	462 (21)	306 (16)	445 (18)	308 (17)	1161 (51)	308 (17)
Estimated generator emission rate (n/s)	1.62E8	1.65E8	1.56E8	1.66E8	1.42E8	1.64E8

Regarding uncertainties, the uncertainty of saturated activity determination reaches 5 % (mostly caused by uncertainty in efficiency calibration), calculation uncertainty is about 1 %, uncertainty in determination ratio between neutron emission from isotropic model and model with neutron generator internals taken from [13] was estimated to 10 %. Another 10 % uncertainty is estimated to be connected with foil positioning. Thus, the total uncertainty of neutron generator emissions was estimated to be 15 % at 1 σ. However, it should be noted that the estimation does not include a possible systematic error caused scattering on surrounding materials which was not evaluated (the nearest shielding wall was ca 30 cm from the foil). The value corresponds with the results presented in [13] where for the same type of generator the neutron emission was determined for 90 kV and 60 μA to be ca. 1.56E8 n/s, which assuming linear dependence on beam current would give some 1.82E8 n/s at the parameters used by us (90 kV, 70 μA). Obviously, the two results cannot be easily compared due to lack of data on previous operation, tritium decrease due to radioactive decay, or production uncertainty in the targets. The producer certificate gives emission rate of 9.02E7 n/s for settings of 80 kV and 60 μA which should correspond to ca. 1.49E8 n/s at 90 kV and 70 μA. Finally, the measurement has been performed at the 5 year old tube, so that tritium decay should be considered. Authors do not have study related to their tube, however based on the study [23] performed at different model of DT generator from the same producer, the neutron yield can be considered stable for the first ca 10 years and in the first 4.5 years it can even slightly rise. Taking this into account together with uncertainty of producer check-out record of 25 % and our uncertainty, the measured values correspond with the expectations.

B. Manganese bath technique to determine the emission rate

The results of the experiment are summarized in Table IV. The table contains the determined activity of the manganese bath and the corresponding emission rate, including a comparison with the reference value provided by manufacturer. These values were determined for both collected samples of activated manganese sulphate solution. The resulting values, determined by a weighted average, are given in the last row of the Table IV.

Table IV
Results of the measurement by Manganese bath method

Sample	A_{Bath} (Bq)	s (%)	B (s-1)	s (%)	1 – C/E (%)
1	1.92E+06	0.44	7.40E+06	0.44	-5.73
2	1.86E+06	0.44	7.20E+06	0.44	-2.79
Average	1.89E+06	0.31	7.30E+06	0.31	-4.23

C. Measurement of dependence on operational parameters of DD generator

The results are shown in the fig. 3 and 4 and in the normalized form in the fig. 5 and 6. Values were normalized to the minima of each row. Obviously, the rise of neutron emission to 90° with high voltage is slightly faster for higher values of beam current. The rise with beam current is slightly higher for lower values of high voltage. The dependences were fitted with an appropriate function: linear for dependence on beam current and power for the case of high voltages. The obtained fit parameters are summarized in tables V and VI.

Table V
Fit parametres of neutron emission to 90° for dependance on high voltage ($c * HV^d$)

Beam current (μA)	c (kV^{-1})	d (-)
20	4.90E-05	2.72
25	4.91E-05	2.78
30	4.86E-05	2.83
35	4.79E-05	2.87
40	4.52E-05	2.91
45	4.03E-05	2.96
50	3.99E-05	2.99
55	4.14E-05	3
60	4.57E-05	3
65	4.22E-05	3.03
70	3.92E-05	3.06

TABLE VI
FIT PARAMETRES OF NEUTRON EMISSION TO 90° FOR DEPENDANCE ON BEAM CURRENT (A * BC + B)

High voltage (kV)	a (μA^{-1})	b (-)
60	0,14	0,49
70	0,25	0,39
80	0,4	0,24
90	0,58	-0,51
100	0,82	-2,01
110	1,05	-2,57
120	1,34	-4,51
130	1,71	-8,05

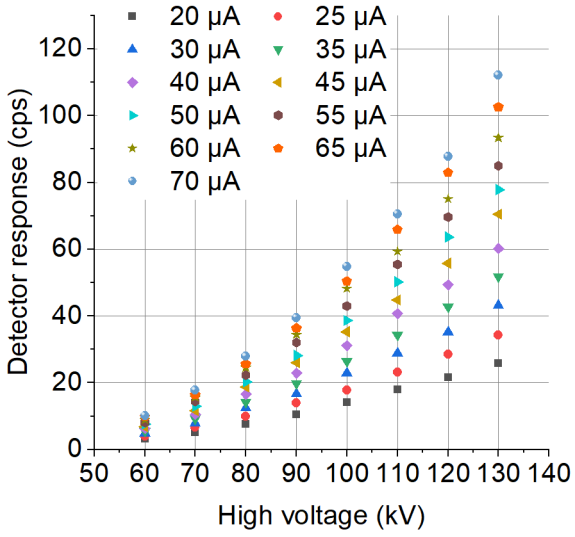

Fig. 3. Dependence of neutron output at 90°on high voltage

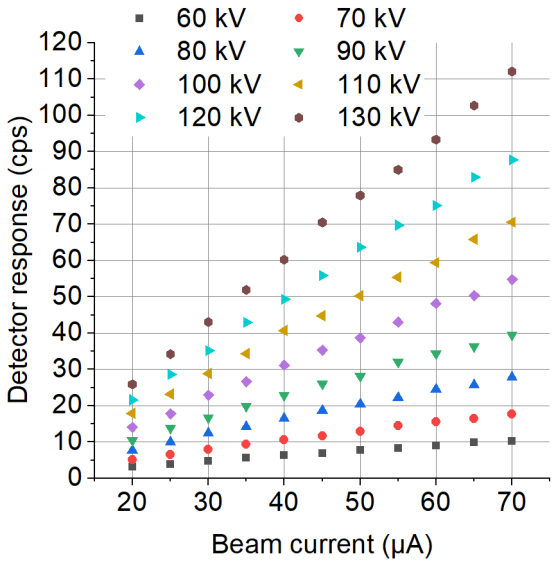

Fig. 4. Dependence of neutron output at 90°on beam current

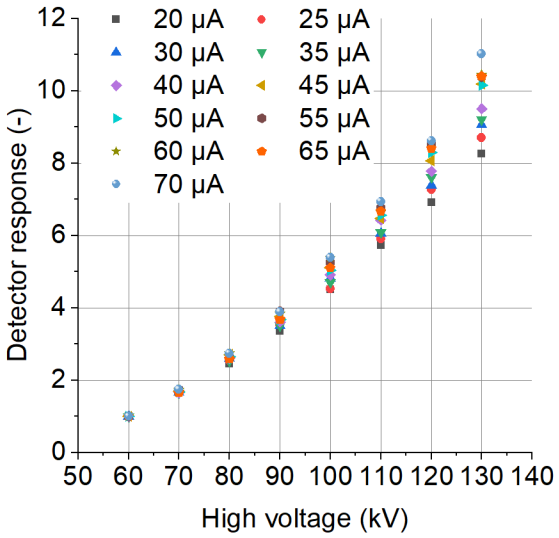

Fig. 5. Normalized dependence of neutron output at 90° on high voltage

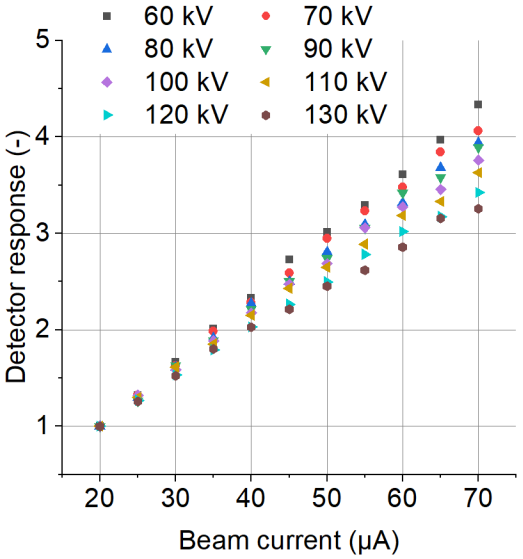

Fig. 6. Normalized dependence of neutron output at 90° on beam current

D. Characterisation of pulse mode operation

Typical result for neutron emission during pulsed mode operation for several frequencies of DT MP320 neutron generator measured via diamond detector for set of frequencies and fixed duty cycle, HV and BC settings is shown in the fig. 7. Red curve indicates idealized rectangular pulse for set pulse parameters; the black is measured one.

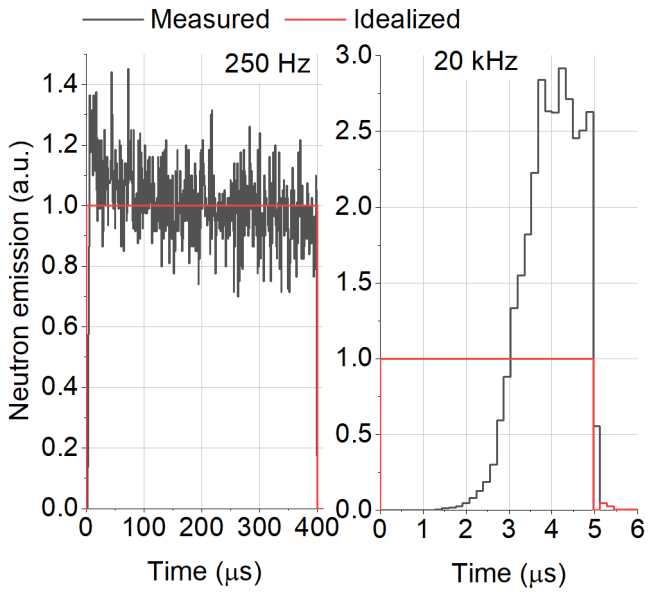

Fig. 7. Typical neutron pulse emission for D-T neutron generator at nominal HV and BC and frequency of 250 Hz and 20 kHz. Adapted from [21].

Typical results of frequency stability measurement are given in fig. 8 and 9 for 100 Hz and 20 kHz respectively. E.g. for 100 Hz operation in 99.62 % of cases the next TTL signal came after 10 000 µs ± 1 µs, however in few cases significant deteriorations has been observed. For 20 kHz operation in 99.39 % cases the next TTL signal came after 50 µs ± 1 µs, however in few cases significant deteriorations has been observed.

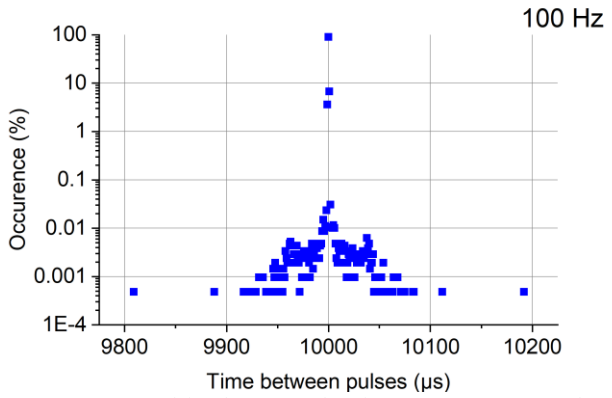

Fig. 8. Occurrences of time between pulses for DT generator operated at 100 Hz and DF 10 %

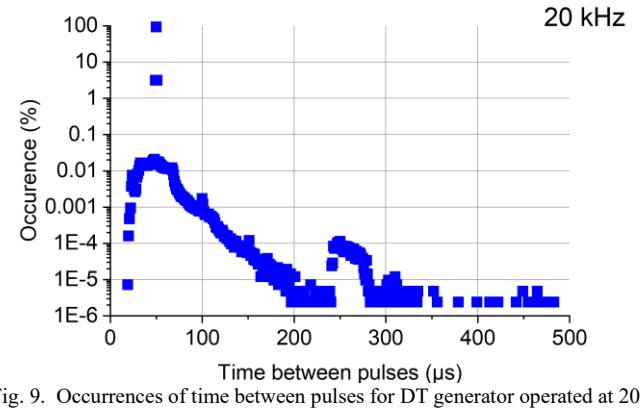

Fig. 9. Occurrences of time between pulses for DT generator operated at 20 kHz and DF 10 %

E. Self-activation, x-ray production

Whereas for ca. 7E6 DD neutron generator the self-activation was found negligible, for ca. 1E8 DT generator weak activation is measurable. E.g. after 1-hour long irradiation, the dose rate at 30 cm from the tube reaches ca. 5 µSv/h at 5 minutes after stop, and ca. 1.5 µSv/h at 30 minutes after stop. Visible gamma lines originates mainly from aluminium and iron activation corresponding to Al-27(n,g), Al-27(n,p) , Al-27(n,α), and Fe-56(n,p) reactions. Although the capture reactions may significantly depend on neutron generator environment, the dose rate results correspond to information provided by producer [24].

During neutron generator operation X-rays of about 80 keV are produced which may need to be handled for some measurements and also it need to be taken into account in radioprotection considerations. However, it can be easily shielded by few mm of lead (see Fig. 10). The results were consistent with findings of [25].

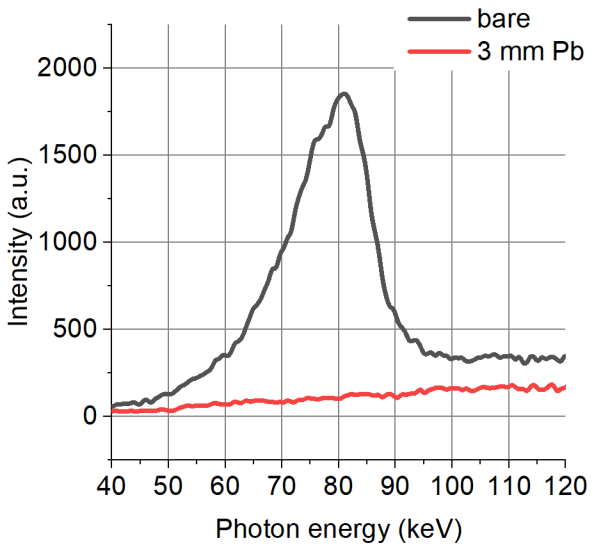

Fig. 10. X-ray peak for bare MP320 DT neutron generator operated at 20 µA and 80 kV and its suppression by 3 mm of lead.

IV. CONCLUSIONS

The detailed understanding of neutron generator behaviour under specific operating conditions enhances their applicability both for training and research purposes and makes results of any application more reliable. The paper summarizes techniques used for characterisation of DD and DT portable neutron generators used at CTU in Prague. To measure neutron generator emission rate, foil-based technique and manganese bath technique were used. Foil-based technique is well established for DT neutron generators; however, its application requires knowledge on spatial distribution of outgoing neutrons. Contrary, the paper shows good applicability of using the manganese bath technique for neutron emission rate determination for low output DD generators. In fact, the applicability of manganese bath technique is not limited to generators of DD type, but the limitation lies rather in the tube geometry (e.g. due to geometry of cable inputs it cannot be used for MP 320 type generator). Further, paper shows typical results of dependence of neutron output on operational parameters (high voltage and beam current set-up). Next, it shows the approach to characterize pulsed mode operation, i.e. the time profile of neutron emission during pulsed mode operation and stability of pulsing frequency. Finally, self-activation and x-ray production was measured and found consistent with producer-provided data.

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