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MICRODOSIMETRY OF MONOENERGETIC NEUTRONS¹

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ABSTRACT

Tissue spheres 0.25, 0.5, 1.0, 2.0, 4.0, and 8.0 μm in diameter were simulated using a wall-less spherical counter filled with a propane-based tissue-equivalent gas. Microdosimetric spectra corresponding to these site sizes were measured for five neutron energies (0.22, 0.44, 1.5, 6, and 14 MeV) and the related mean values $\bar{Y}_F$ and $\bar{Y}_D$ were calculated for several site sizes and neutron energies. An elaborate calibration technique combining soft x-rays, a ^{55}Fe photon source, and a ^{244}Cm collimated source of alpha particles was used throughout the measurement. The spectra and their mean values are compared with theoretically calculated values for ICRU tissue. The agreement between the calculated and the measured data is good in spite of a systematic discrepancy, which could be attributed, in part, to the difference in elemental composition between the tissue-equivalent gas and plastic used in the counter, and the ICRU standard tissue used in the calculations.

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1. INTRODUCTION

The distribution patterns of energy deposition in small tissue spheres ($d = 0.1-10 \mu\text{m}$) irradiated by neutron beams have been the subject of extensive experimental and theoretical studies (1-20). Earlier experimental work was primarily motivated by the unique advantages of neutrons as a tool for understanding basic mechanisms in radiobiology (21). The advent of neutron radiotherapy has stimulated further microdosimetric measurements needed to specify beam quality in treatment fields (22, 23). The experimental measurements have been accompanied by theoretical calculations aimed at a better understanding of the interaction of neutrons with tissue, and the subsequent ionization and excitation processes (24). Microdosimetric measurements of neutron radiation are equally important in radiation protection studies, helping to evaluate the radiation quality factor (25, 26, 27, 28).

The energy deposition patterns in simulated small tissue volumes irradiated by neutrons have been measured (1-9) and calculated (10-20). Much of the early experimental microdosimetric data for monoenergetic neutrons were obtained by the Radiological Research Laboratory (RRL) of Columbia University and published in their Annual Reports. There are two distinct groups of data: the first group published in 1967 by Biavati and Boer (1), and covering work from 1964 to 1967, and the later one, published by Rodgers et al., in 1973 (3). Rodgers and Gross summarized the latter report at the Fourth Microdosimetry Symposium (1974),

pointing out, however, that the data were preliminary (6). The disagreement between these two groups of data ranges from 10 to 33 % and prompted us to undertake the comprehensive, systematic measurements presented in this paper. Before 1970, measurements were made with electronics and processing devices that are obsolete by modern standards, and further, the counter calibration was imprecise; some published papers (3, 6) reported only preliminary data that was never confirmed. This situation justified our effort to undertake systematic measurements of energy deposition spectra for a wide range of simulated site sizes and neutron energies. Progress reports describing an early stage of the work done at Radiological Research Accelerator Facility have been published (29, 30) and some preliminary results have been presented at Seventh Symposium on Microdosimetry (7).

Several factors which may influence the results, such as the counter size and the thickness of its wall were investigated in some detail. Substantial improvements in the design of the spherical counter, in calibration techniques, and in electronics, briefly described in the following section, enabled us to measure the spectra with a high degree of reproducibility.

Microdosimetric spectra are presented in the usual notation of $d(y)$ vs $\log(y)$ or $yd(y)$ vs $\log(y)$ (31, 32), and the contributions of gamma rays, protons, alpha particles, and heavy recoils are discussed briefly.

2. EXPERIMENTAL

2.1. The wall-less counter and electronics

A miniature version of the Rossi-type, tissue-equivalent (TE), spherical wall-less counter, described in detail in (3) and (33), was used for our experiments. The counter consists of a 6.35 mm spherical grid, a helix, and the central anode, all contained in a TE plastic sphere, 2.54 cm in diameter. The thickness of the wall of the TE plastic sphere must exceed a critical value which depends on the type and energy of the incident radiation to obtain the secondary particle equilibrium. The small size of the counter enabled us to measure very small simulated site sizes (0.25-0.5 μm).

A low noise FET preamplifier, developed by Radeka (34), extended the measurements down to energy depositions consisting of several ion pairs per event. In addition, a specially designed amplifier with logarithmic response (35) was used to accommodate the extremely broad range of pulse height. The low-noise preamplifier was connected directly to the anode of the proportional counter. Analog pulses from the preamplifier were fed through a logarithmic amplifier into an analog-to-digital converter and stored in the memory of a PDP-8 computer. Data analysis was performed both on- and off-line with specially developed software.

2.2. The neutron source

Monoenergetic neutrons having mean energies of 0.22, 0.44, 1.5, 6.0, and 14 MeV were used to irradiate TE wall-less counters with diameters equivalent to 0.25, 0.5, 1.0, 2.0, 4.0, and 8.0 μm of tissue. The conditions during irradiation were similar to those often used in radiobiological experiments, so that the microdosimetric parameters can be used for biophysical calculations. Data on the neutron source and counter irradiation geometry are shown in Table 1.

TABLE 1. NEUTRON SOURCE DATA AND COUNTER IRRADIATION GEOMETRY

Mean neutron energy (MeV)	Reaction	Approx. γ -ray dose (%)	Counter position	
			Distance to target (cm)	Angle from ion beam (deg)
0.22 $\pm$ 44%	T(p,n)	1	11.5	100
0.44 $\pm$ 10%	T(p,n)	1.5	11.5	120
1.5 $\pm$ 8%	T(p,n)	1	10.4	0
6 $\pm$ 8%	D(d,n)	6	11.5	0
14 $\pm$ 7%	T(d,n)	4	12.0	100

Neutrons were produced using proton or deuteron beams from the Radiological Research Accelerator Facility 4-MV Van de Graaff then at Brookhaven National Laboratory. The charged particle beams were incident on targets consisting of a copper backing with a thin titanium layer in which either tritium or deuterium was absorbed. The target, target holder, and water cooling lines were designed to minimize attenuation and scatter.

2.3. The microdosimetric measurement technique

The experimental technique generally followed that described by Srdoć (33). Two spherical, wall-less Rossi-type counters were used throughout the measurements. The "1-inch" counter, used in previous experiments by Rodgers and Gross (6) was described by them in detail (3). The "1/4-inch" counter has essentially the same design, but scaled down by a factor of 4. However, this smaller counter has two distinct features: a) it is enclosed in a thin Al shell, and b) it has an inner layer, made of TE plastic, which can be changed or removed. In addition, the smaller physical size of this detector improves its operational stability, especially when small tissue site sizes are simulated, because of the higher gas pressure for a given site size relative to the 1-inch counter. Spectra obtained from both counters were compared for a range of neutron energies and simulated site sizes, and no significant differences were observed.

There were no changes in spectral shapes when a 3 mm-thick outside layer of TE plastic was placed on the Al shell; therefore, subsequent measurements were made without it. The thickness of the inner layer of TE-plastic is very critical at high neutron energies. For instance, a 0.25 mm thick TE plastic wall is insufficient to maintain secondary particle equilibrium, resulting in a distorted spectrum of 14-MeV neutrons as shown in Fig. 1, curve 1, in contrast to a 3-mm thick layer, which produces equilibrium (curve 2). Further increases in the thickness of the plastic do not influence the spectrum. The counter was

Fig.1. The influence of the inside layer thickness on the $d(y)$ vs y spectrum, 14 MeV neutrons, $1 \mu\text{m}$ site size. Wall-less spherical counter, 6.35 mm in diameter, Curve 1: 0.25 mm tissue-equivalent plastic layer, Curve 2: 3 mm tissue-equivalent plastic layer.

filled with propane-based TE gas mixture (33). A constant gas pressure was maintained using a pressure and flow regulating device. Very often the measurements were performed in a closed-system mode, since no changes in energy calibration and resolution were detected over a typical period during which data were collected, lasting from 1/2 to 2 hours.

2.4. Counter calibration

Neutron microdosimetric spectra span more than 3 decades in energy. Therefore, it was essential to measure at least two widely separated calibration points within these energy limits to check the linearity of the counter's response. The low energy region was checked with a miniature soft x-ray tube (36) emitting 1.49-keV Al K_{α} -line photons. For higher energies, a finely collimated ^{244}Cm α source was used. For calibration, the energy deposited by α particles in propane TE gas was calculated using Ziegler's data (37). A third calibration source, ^{55}Fe , was used at high gas pressures where the 5.9-keV photons are completely absorbed in the active volume.

Our calibration measurements showed that the voltage between the counter helix and the sphere is critical in maintaining proportionality over the whole energy range (Fig. 2). The generally accepted constant-voltage ratio may lead to serious distortions at the high-energy end of the spectrum. Too low a voltage difference between the sphere and helix causes a loss of ions, whereas too high a

Fig. 2. Calibration of spherical proportional counter with ^{244}Cm alpha source. Horizontal scale: Collecting voltage between helix and sphere. Vertical scale: Energy deposited in the counter. Dashed horizontal lines: Calculated values, based on Ziegler's data (37): 1μ - 84.9 keV; 2μ - 174.9 keV; 4μ - 373.2 keV. The 8μ calculated value (886.3 keV) is out of scale. The ^{244}Cm calibration points are 6-7% lower in the middle of the plateau when the counter is calibrated with soft x-rays (Al K_{α} -line = 1.49 keV) due to the difference in the corresponding W-values.

difference causes a nonlinear counter response. Both phenomena are visible in the plot of charge vs collecting voltage in Fig. 2. Here, the curves correspond to saturation curves of an ionization chamber. The electric charge created by a single event in the spherical volume is too

small to be detected directly by electronics, hence the proportional counter inside the sphere, which multiplies the charge by a factor of $\sim 10^3$. Therefore, instead of ionization current vs voltage, the graph shows the electric charge created by the calibration source after multiplication as a function of the collecting voltage, i.e. the voltage difference between the sphere and the helix. The working voltage is usually chosen in the middle of the flat plateau.

The calibration data reveal several phenomena which are important, particularly in high-energy neutron microdosimetry at simulated tissue sizes of 4 μm and 8 μm . While the agreement between the calibration data is very good for the Al x ray (1.49 keV) and the α peak at small sizes (0.25 to 2.0 μm), systematic discrepancies of 10 and 25 % were observed at TE gas pressures corresponding to 4- and 8- μm site diameters, respectively. In other words, if the 1.49-keV calibration peak is taken as the reference point, the measured α peak was 10 to 25 % lower than the value calculated for these site sizes (Fig. 2). Several factors contribute to this discrepancy: a) the difference in energy per ion pair (W-value) between the soft x-ray photons and the α particles accounts for 6-7 % less ions per ionizing event for α particles at any gas pressure (38); b) an increasing columnar recombination at higher pressures in a densely packed heavy-ion track; and, c) an energy-dependent differential w-value for α particles. Consequently, the data for the 4- μm site diameters are not as reliable as those for the smaller site diameters. The distortion of the high energy part of the

spectra cannot be excluded, however, it is not likely to exceed 10% on the energy scale.

3. RESULTS AND DISCUSSION

The most concise way to present the microdosimetric data is to tabulate mean values $\bar{y}_F$ and $\bar{y}_D$ for the entire range of measured neutron energies and site sizes (Table 2), or to plot these values vs neutron energy, as shown for $\bar{y}_D$ in Fig. 3. More complete information can be obtained from the $d(y)$ vs $\log(y)$, and $yd(y)$ vs $\log(y)$ spectra. In the latter representation, the area delimited by any two values of the lineal energy, y , is equal to the fraction of dose delivered in that interval. The shape of the microdosimetric distributions, as well as changes in their shape with neutron energy or site size are fairly well understood, at least in principle (10, 15, 17, 19).

We give several examples of such spectra. Figure 4 shows the variation of the shape of the $yd(y)$ vs $\log(y)$ spectrum for 0.44-MeV neutrons for site sizes ranging from 1 to 8 μm . While the 0.25, 0.5, and 1.0 μm diameter spectra, (shown separately in Fig. 5, for clarity),

TABLE 2. THE DOSE AVERAGE OF LINEAL ENERGY $\bar{y}_D$ AND THE FREQUENCY MEAN OF LINEAL ENERGY $\bar{y}_F$ IN KeV/ μ m AS A FUNCTION OF NEUTRON ENERGY E_n (MeV) AND SITE DIAMETER (μ m)

E_n (MeV)	$\bar{y}_F$, $\bar{y}_D$	Sphere diameter (μ m)				
		0.25	0.5	1.0	2.0	4.0
0.22	$\bar{y}_D$	-	74.5	69.3	-	-
	$\bar{y}_F$	-	38.5	37.0	-	-
0.44	$\bar{y}_D$	80.5	76.5	73.7	64.1	45.7
	$\bar{y}_F$	56.1	48.1	42.8	37.6	23.0
1.5	$\bar{y}_D$	67.6	64.4	64.8	57.7	47.5
	$\bar{y}_F$	-	-	44.3	33.8	32.0
6	$\bar{y}_D$	60.1	57.5	57.0	48.3	37.9
	$\bar{y}_F$	17.0	17.0	18.0	16.0	-
14	$\bar{y}_D$	-	39.0	39.6	-	67.5
	$\bar{y}_F$	-	9.8	10.1	-	10.1

dominated by proton "crossers", are practically indistinguishable, for larger site sizes proton stoppers become increasingly important. Since a stopper deposits all its energy in the sphere, a further increase in the diameter of the sphere results only in smaller y -values. The spectra above 1 μ m shift to lower lineal values as the site size increases. This shift is accompanied by a broadening of the spectral distributions.

Fig. 3. The dose average of lineal energy, $\bar{Y}_D$, is shown as a function of neutron energy for different sizes of simulated tissue spheres. Full triangles: Calculated values $\bar{Y}_D$ for $1\mu\text{m}$ ICRU tissue sphere according to Caswell and Coyne (19). Experimental $\bar{Y}_D$ values: Open circles, $d = 0.5\mu\text{m}$. Open triangles, $d = 1.0\mu\text{m}$. Experimental points for both site sizes merge at $E_n > 1$ MeV, and are denoted by X. Full circles, $d = 2\mu\text{m}$. Full squares, $d = 4\mu\text{m}$.

Fig. 4. $yd(y)$ vs $\log y$ spectra, 0.44 MeV neutrons. Variation of spectral shape and position with increasing site size from 1 to 8 μm . The 4 μm and especially the 8 μm site size spectra are less reliable due to calibration problems at higher gas pressure (see text).

The physical explanation is simple and straightforward. The initial proton recoil flux, having an approximately rectangular spectrum, is intercepted by spheres of different dimensions. For a sphere diameter equivalent to 1 μm or less of tissue, most of the recoil protons cross the sphere, producing a relatively sharp peak in the $yd(y)$ representation. As the sphere's diameter increases above 1 μm , an increasing fraction of recoil protons deposit their entire energy within the sphere. A further increase in diameter is not accompanied by more energy deposition, and therefore, the whole spectrum is shifted

Fig. 5. $y_d(y)$ vs $\log y$ spectra, 0.44 MeV neutrons. The shape and position of spectra representing tissue spheres of 1, 0.5, and 0.25 μm in diameter do not change appreciably. Their respective mean values y_D change by several percent only (see Table 2).

toward lower y -values. The broadening of the spectrum for larger site sizes is caused by an additional term in the total variance, reflecting the shape of the initial spectrum of the recoil protons.

Figures 6 through 9 show the variation of the shape of the $y_d(y)$ vs $\log(y)$ spectrum with neutron energy and site sizes. Figures 6 and 7 show the change in spectral shape with neutron energy for a site size of 1 μm . The 0.22-, 0.44-, and 1.5-MeV spectra have a sharp proton cutoff

Fig. 6. Comparison of $yd(y)$ vs $\log y$ spectra, $1 \mu\text{m}$ site size, 0.44, 1.5, and 6 MeV neutrons. For a complete description, see Section 3.

Fig. 7. Comparison of $yd(y)$ vs $\log y$ spectra, $1 \mu\text{m}$ site size, 0.22, 0.44, 1.5, and 14 MeV neutrons. For a complete description, see Section 3.

Fig. 8. $y_d(y)$ vs $\log y$ spectra, $1\ \mu\text{m}$ site size, 14 MeV neutrons. Several distinct regions are marked on the spectra, indicating the contributions from protons, alpha particles, and heavy recoils to the total absorbed dose, which is proportional to the area under the curve.

at approximately $145\ \text{keV}/\mu\text{m}$. At higher neutron energies, the recoil proton peak is shifted to lower y -values: for 6-MeV neutrons, the proton peak lies around $17\ \text{keV}/\mu\text{m}$, whereas for 14-MeV neutrons it lies around $10\ \text{keV}/\mu\text{m}$. Neutrons above 1 MeV produce heavy recoils with sufficient energy to be observed in the microdosimetric spectra. A short "tail" belonging to α particles and heavy recoils (C, N, and O) is evident between 150 and $300\ \text{keV}/\mu\text{m}$ on the 1.5-MeV spectrum, becoming more prominent as the neutron energy increases (see Fig. 6, 6-MeV neutrons).

Fig. 9. 14 MeV neutrons, $yd(y)$ vs $\log y$ spectra. The shape of the spectrum for 1, 0.5, and 0.25 μm spheres is practically the same.

For 14-MeV neutrons, the high-energy tail splits into distinct peaks, belonging to alphas and heavy recoils (Fig. 9). The heavy recoils, consisting mostly of carbon and oxygen ions, cannot be separated with our present technique. The variation of the shape of the $yd(y)$ spectra with site size for 14-MeV neutrons is shown in Fig. 9. Again, there is no significant difference between spectra for site sizes from 0.25 to 1 μm . At larger sizes, the general trend is a shift to lower values of y , signifying that a fraction of the heavy particles has a range comparable to 1 μm in tissue.

We attempted to calculate the frequency mean of the lineal energy, $\bar{y}_F$, for several site sizes and neutron energies. In contrast to $\bar{y}_D$, values of $\bar{y}_F$ are very sensitive to the contribution from the lower energy part of the spectrum. To eliminate the contribution of the gamma-ray contamination from the neutron spectra, the subtraction procedure shown schematically in Fig. 10 was applied. The $d(y)$ vs $\log(y)$ spectra

Fig.10. Processing of neutron $d(y)$ vs $\log y$ spectra, 0.22 MeV neutrons, 1 μm site size. 1: Experimental spectrum, neutrons + gamma-rays. 2: ^{60}Co gamma-rays spectrum, 1 μm . 3: Net neutron spectrum, obtained by subtracting the gamma-ray spectrum from the neutron + gamma-ray spectrum.

for 0.22-MeV neutrons and ^{60}Co gamma rays for a $1\text{-}\mu\text{m}$ site size are plotted together, matching the ^{60}Co peak to the peak at low values of y in the neutron spectrum. The shapes of the overlapping sections of both spectra show that energy deposition below $1\text{ keV}/\mu\text{m}$ is almost entirely due to gamma rays. The region between 1 and $10\text{ keV}/\mu\text{m}$ consists of a mixture of a high lineal energy tail of gamma-ray depositions and a low lineal energy tail of short or truncated proton tracks within the sphere. Above $10\text{ keV}/\mu\text{m}$, the spectrum is composed entirely of recoil protons. We note that the spectrum of gamma-ray energy depositions which contaminates the neutron beam is not identical to a ^{60}Co gamma-ray spectrum. However, the difference in the shape of the microdosimetric spectra for gamma rays having a wide range of energies, e.g., ^{137}Cs ($E = 0.66\text{ MeV}$), ^{60}Co ($E_1 = 1.2\text{ MeV}$, $E_2 = 1.33\text{ MeV}$), and ^{24}Na ($E = 2.76\text{ MeV}$), is small, thus justifying the subsequent step of subtracting the ^{60}Co gamma-ray spectrum, leaving the residual spectrum which represents the net neutron energy depositions. The mean value $\bar{y}_F$ changes drastically after subtracting the gamma-ray contamination, however, no significant change in the values of $\bar{y}_D$ was observed after subtracting the gamma component. For instance, in case of 0.22-MeV neutrons, the $\bar{y}_F$ -value increased from $12.2\text{ keV}/\mu\text{m}$ to $37.0\text{ keV}/\mu\text{m}$, while the $\bar{y}_D$ -value changed from $63.9\text{ keV}/\mu\text{m}$ to $69.3\text{ keV}/\mu\text{m}$. The difference in $\bar{y}_D$ -values before and after subtraction of the gamma-ray contamination decreases as neutron energy increases.

Table 2 gives the values of $\bar{y}_F$ obtained after the gamma-ray subtraction. Booz (39) recommended various procedures to extrapolate

events below $y = 0.05 \text{ keV}/\mu\text{m}$, which are usually overlapped by electronic noise. The values obtained were found to change by only a few percent when the "pure" neutron spectra were further extrapolated exponentially to zero lineal energy. Consequently, the $\bar{y}_F$ -values of Table 2 were calculated with no extrapolation. Table 3 compares three sets of experimental data for 14-MeV neutrons. Our data agree fairly well with those of Rodgers and Gross (6). The values of $\bar{y}_D$ first reported by Biavati and Boer were calculated assuming the LET for ^{244}Cm α particles used for calibration was $77.8 \text{ keV}/\mu\text{m}$. In their data from 1965 on, the LET was recalculated as $87.7 \text{ keV}/\mu\text{m}$, using new stopping power data. Therefore, we applied a correction to the older data of Biavati and Boer. First, the old notation was converted into present notation by multiplying the $\bar{y}_D$ values by $3/2$; then, a correction factor of 1.127 was used to correct the α -particle calibration. Remarkably, our data, the Rodgers and Gross data (6), and the LET-corrected early data of Biavati and Boer (1), for 14-MeV neutrons and $1 \mu\text{m}$ site are in surprisingly good agreement, giving an average value $\bar{y}_D = 89.5 \text{ keV}/\mu\text{m}$, and less than a 1% departure of each individual measurement from the mean value. However, for other site sizes, as well as the later data of Biavati and Boer (1), the agreement is not nearly as good. The calculated $\bar{y}_D$ values of Caswell and Coyne (19) for ICRU tissue are generally higher than the

TABLE 3. COMPARISON OF EXPERIMENTAL DATA FOR 14 MeV NEUTRONS: $\bar{Y}_D$ IN keV/ μ m FOR VARIOUS SPHERE DIAMETERS

		This paper	Rodgers and Gross (6)	Biavati and Boer (1)		
				1	2	3
Site size μ m:		0.5	0.5	0.4	0.4	0.49
$\bar{Y}_D$	W-less:	89.0	86.6	-	-	-
	Wall:	-	-	97.8	110.0	113.2
Site size μ m:		1.0	1.0	1.0	1.0	0.99
$\bar{Y}_D$	W-less:	89.6	88.7	-	-	-
	Wall:	-	-	80.1	90.3	103.9
Site size μ m:		2.0	2.0	2.0 ⁺	2.0 ⁺	2.0 ⁺
$\bar{Y}_D$	W-less:	-	86.0	-	-	-
	Wall:	-	-	79.0	89.0	97.0
Site size μ m:		4.0	4.0	4.0 ⁺	4.0 ⁺	4.0 ⁺
$\bar{Y}_D$	W-less:	67.5	72.3	-	-	-
	Wall:	-	-	79.0	89.0	84.0

⁺ Interpolated values

1. Data published in RRL Annual Report NYO-2740-1 (1964)

2. Data published in RRL Annual Report NYO-2740-2 (1965)

3. see Ref. (1).

experimental data by 10 to 25% (see Fig. 3). However, it should be pointed out that 3 out of 4 experimental sets of data (including the early Biavati and Boer data) have consistently lower $\bar{Y}_D$ values than the calculated ones. The discrepancy can be explained at least qualitatively as follows: a) the experimental values need correction for the different values of W, as discussed; b) more precise and complete input data for the calculations are required, as pointed out by Caswell and Coyne (19); and, c) the calculations are for ICRU tissue, not for methane- or propane-based TE gas and TE plastic. While the W correction will definitely help to bring the experimental and the calculated data closer, the effect of the remaining two changes is unknown.

The ultimate use of the microdosimetric data presented in Figs 3-9 or the mean values $\bar{Y}_F$ and $\bar{Y}_D$ must be born in mind. For radiobiological purposes, the uncorrected values may be more relevant since the irradiated samples will absorb both the gamma-rays and the neutron-induced radiation (recoil protons and heavy ions). On the other hand, for comparison with calculated values of $\bar{Y}_F$ and $\bar{Y}_D$, as well as for radiation physics, it is better to have a clear-cut picture of the contribution of each type of radiation to the total absorbed dose and their respective microdosimetric spectra.

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