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On partition of the excitation energy between the fission fragments in spontaneous fission of ^{252}Cf

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ABSTRACT

In this work the correlation has been investigated between the neutron numbers and average excitation energies for the complementary fission fragments in the spontaneous fission of ^{252}Cf. The results of the experimental work on the average neutron number, total kinetic energy distribution of the pair of fragments, and the average energies of neutrons and gamma quanta emitted from the fission fragments are used. Calculated results are compared with the results of the other calculations and measurements.

Keywords: Nuclear fission, ^{252}Cf , neutron number, excitation energy, correlation

1. Introduction

Mechanisms of fission and neutron emission from fission fragments stipulate neutron energy and angular distribution in fission of heavy nuclei. Different neutron characteristics such as neutron average energy, average neutron multiplicity and etc., measured and calculated at the different fragment masses and kinetic energies are due to these mechanisms. The detailed analysis of these quantities measured in different experiments promotes understanding of nuclear fission peculiarities, in particular especially for partition of the excitation energy between the fragment pair.

Neutron yields from individual fission fragments and correlation between neutron numbers emitted from complementary fission fragments are the basic characteristics used in studying the problem and a number of experimental and theoretical works [1-15] have been devoted to this goal.

Theoretical study of the problem depends on the model used to describe excitation energy dependence on nuclear temperature, usually within the Fermi gas model [13-14], or within the generalized super-fluid model [14]. In addition to these models, the constant temperature level density approximation has great importance in description of nuclear level density versus excitation energy. A number of indications are available on the constant temperature behavior of nuclear level density on the excitation energy [16-18]. Consequently, the results of such investigations are sensitive to the model chosen [19]. However, the problem may be well analyzed on the base of experimental data and empirical expressions describing these data, which is the aim of this work.

As stated above, in the study of excitation energy partition between the fission fragments, number of neutrons emitted from fragment is the main quantity because each emitted neutron carries out $\sim(6-7)$MeV excitation energy from fragment and thus, the contribution to the fragment excitation is mainly determined by the average number of neutrons yielded from the fragment of mass number, A. Additionally, gamma emission carries the energy of fission fragment which is less probable compared to neutron emission. The ratio

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of neutron and gamma widths (Γ_n/Γ_γ) is $\sim 10^2$ at the fragment excitations $\sim$6MeV and angular momentum value $5-6\hbar$ [20]. Thus, competition between neutron and gamma emission may be valid for the last neutron emitted from the fragment. Therefore, gamma rays are usually supposed to be emitted after all possible neutron emissions and their contribution to the fission fragment excitation energy must be taken into the account for the accurate calculation of fission fragment excitation energy.

First neutron yield experiments in neutron induced fission of ^{233}U, ^{235}U, ^{239}Pu and in spontaneous fission of ^{252}Cf were analyzed in the work of Terrell [1] (and references therein) and it was stated that the lowest number in neutron emission corresponds to the formation of near magic nuclei with charge and neutron numbers of $Z \approx 50$, $N \approx 50$ (masses 82 and 128), respectively and it was suggested "that magic and near magic fragments have low excitations, and consequently emit almost no neutrons, because of greater rigidity against distortion from near-spherical shapes". Later experiments, using more dedicated neutron detection techniques have measured neutron yields dependent on the fission fragment mass and the total kinetic energy of fragments pair. The gamma emission yields were also measured in such experiments, review and analysis of which are given in the work of Nifenecker and others [7]. In the work [8], the mean value of neutron number and neutron number variances as a function of mass and total kinetic energy of fission fragments in spontaneous fission of ^{252}Cf were measured and detailed analyses of similar measured data investigated in works [2-5] are given. Experimental work [12] investigated the partition of excitation energy between the fragment pairs in 12MeV proton induced fission of ^{232}Th, measuring primary and secondary fission fragment mass (yields) and total kinetic energy of primary fragments. Part of the results of this work, which were presented for asymmetric fission model are used for comparison with our results. Below in the sections 2 - 4, different independent experimental data analyzed and then presented our prescription for calculation of partition of excitation energy between the fragments in spontaneous fission of ^{252}Cf. Discussion and conclusion are given in Section 5.

2. Correlation between the average neutron numbers at the definite mass split

Correlation between the average neutron numbers at the definite mass split for a given total kinetic energy of fission fragments may be calculated by the use of the experimental data on neutron average number $\nu(A)$ for the fragment of mass number A at total kinetic energy E_K of fragments pair (by $\bar{\nu}(A)$, we denote average of this mean number over the total kinetic energy distribution). The measured in the work [8] mean values of neutron number are dispersed from the average light ($A_L = 108$) and heavy ($A_H = 144$) fragments of which, σ_{ν_L} and σ_{ν_H}, are $\sigma_{\nu_L} = 0.639$, $\sigma_{\nu_H} = 0.832$ at the most probable total kinetic energy $E_K^P = 190\ MeV$. Corresponding covariance $cov(\nu_L, \nu_H)$ and correlation coefficient $\rho(\nu_L, \nu_H)$ of these neutron numbers are $cov(\nu_L, \nu_H) = -0.38$, $\rho(\nu_L, \nu_H) = -0.714$. This result means that there is significant anti correlation between the excitation energy of heavy and light fragments, i.e., the higher the excitation energy of one fragment, the lower the one of another fragment at the fixed total kinetic energy of fragment pair. However, one of most important characteristics used in many applications of prompt fission neutron spectrum is the correlation coefficient, at all possible fission fragment kinetic energies, i.e. correlation at the definite mass split.

The correlation coefficient at the average mass split, $A_L = 108, A_H = 144$ in the spontaneous fission of ^{252}Cf may be calculated by the use of experimental data for the average neutron number, dependent on the fission fragment total kinetic energy (Fig. 1) and for the total kinetic energy distribution $p(E_K)$ of this fragment pair (Fig.2), [8].

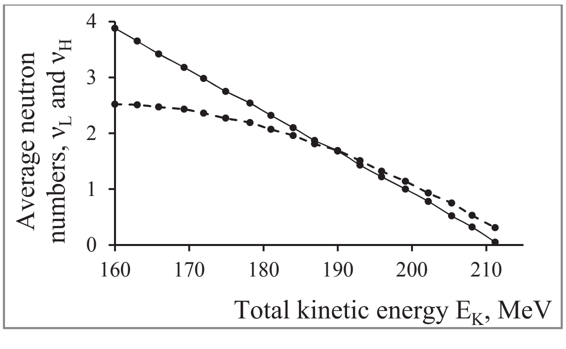

Fig. 1 Experimental average neutron values ν_L (solid curve), and ν_H (dashed curve) dependent on the total kinetic energy of fission fragments for the

mass split A_L=108, A_H=144 in the spontaneous fission of ^{252}Cf [8].

The covariance and the correlation in two discrete set of variables $\{\nu_L^i\}$ and $\{\nu_H^i\}$ having distributions (or probabilities) $p(\nu_L^i)$ and $p(\nu_H^i)$ are [21],

$$cov(\nu_L,\nu_H) =\sum_i\left(\nu_L^i-\bar{\nu}_L\right)\left(\nu_H^i-\bar{\nu}_H\right)p\left(\nu_L^i\right)p\left(\nu_H^i\right) \quad (1)$$

$$\rho(\nu_L,\nu_H)=\frac{cov(\nu_L,\nu_H)}{\sigma^2(\nu_L)\sigma^2(\nu_H)} \quad (2)$$

where dispersion σ and average number $\bar{\nu}$ are determined by

$$\sigma^2\left(\nu_{L,H}\right)=\sum_i\left(\nu_{L,H}^i-\bar{\nu}_{L,H}\right)^2 p\left(\nu_{L,H}^i\right), \quad (3)$$

$$\bar{\nu}_{L,H}=\sum_i \nu_{L,H}^i\, p\left(\nu_{L,H}^i\right), \quad (4)$$

for the light fragment L and heavy fragment H, separately. Neutron yield, dependent on total kinetic energy in the above formulae, is the same as the total kinetic energy yield $p\left(\nu_L^i\right)=p\left(\nu_H^i\right)=p(E_K^i)$, and is normalized as $\sum_i^{18} p\left(\nu_{L,H}^i\right)=100$ (18, is the number of experimental data in the Figs. 1, 2).

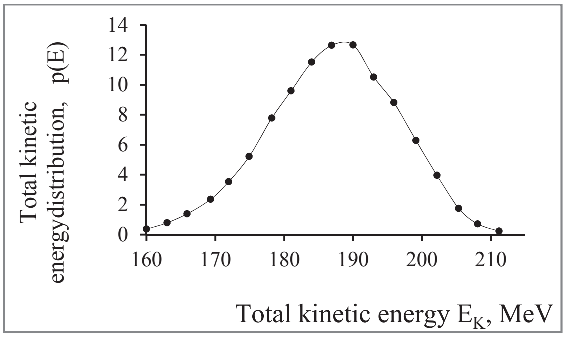

Fig. 2 Experimental total kinetic energy distribution of the pair of fission fragments in spontaneous fission of ^{252}Cf for the mass split A_L=108, A_H=144 [8].

The calculated values for the average neutron numbers are found to be $\bar{\nu}_L=1.89$, $\bar{\nu}_H=1.76$, which are very close to the measured results $\bar{\nu}_L=1.88$, $\bar{\nu}_H=1.74$ in the work [9]. Neutron number dispersions and correlation coefficient are found to be $\sigma^2(\nu_L)=0.685$, $\sigma^2(\nu_H)=0.697$ and $\rho(\nu_L,\nu_H)=0.030$, respectively. Correlation coefficient, measured in the work [8] has value of $\rho_{exp}(\nu_L,\nu_H)=-0.064$, the statistical experimental error of which is 50%. Thus, the calculated value agrees well with this value. This result indicates that there is a negligible correlation between the average neutron numbers from complementary fission fragments when one integrate over total kinetic energy distribution of fragment pair. In terms of the excitation energies, there is almost zero correlation between the excitation energies of fragment pair. Apparently, this result means that independent excitation energy distribution in the average light and heavy fission fragment pair is valid for other fragment pairs and it is very important in calculation of neutron energy spectrum in fission [23, 16].

3. Correlation between the average neutron numbers of heavy and light mass groups

The average number of neutrons emitted from the fragment of mass A at all its possible kinetic energies is another important characteristic in deducing the excitation energy partition between the fission fragments. Here we investigated the global characteristic-correlation in average neutron numbers including all complementary fission fragment masses irrespective to their total kinetic energies. The coefficient is determined as

$$\rho=\frac{\sum_i\left[\bar{\nu}(A_L^i)-\bar{\bar{\nu}}(A_L)\right]\left[\bar{\nu}(\mathrm{A_H^i}-\bar{\bar{\nu}}(\mathrm{A_H})\right]}{\left\{\sum_i\left[\bar{\nu}(A_L^i)-\bar{\bar{\nu}}(A_L)\right]^2\sum_i\left[\bar{\nu}\left(A_H^i\right)-\bar{\bar{\nu}}(A_H)\right]^2\right\}^{1/2}}, \quad (5)$$

where $\bar{\nu}(A_L^i)$ and $\bar{\nu}(A_H^i)$ are the average neutron numbers of i-th complementary fission fragments A_L^i and A_H^i, respectively. Experimental data of the work [9] for this quantity are illustrated in the Fig.3. $\bar{\bar{\nu}}(A_L)$ and $\bar{\bar{\nu}}(A_H)$ are the average neutron numbers in the light and heavy fragments group, respectively and are determined by

$$\bar{\bar{\nu}}(A_L)=\frac{\sum_i^N\bar{\nu}(A_L^i)}{N},\ \bar{\bar{\nu}}(A_H)=\frac{\sum_i^N\bar{\nu}(A_H^i)}{N}, \quad (6)$$

N is the number of fragments in each group.

We divide the whole mass range of fission fragments into three groups-mass ranges substantiating the monotonically behavior overlooked in the mass distribution of fission fragment in the spontaneous fission of ^{252}Cf [9, 10]. These three mass ranges are,

a) $A_L^i(73-106)$, $A_H^i(146-179)$,

b) $A_L^i(106-112)$, $A_H^i(140-145)$,

c) $A_L^i(112-126)$, $A_H^i(126-140)$.

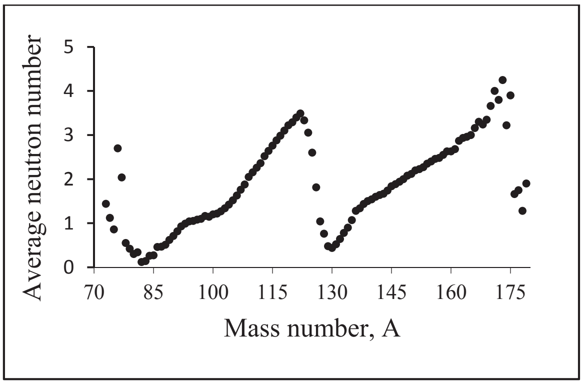

Fig. 3 Experimental average neutron number dependent on the fission fragment mass number in the spontaneous fission of ^{252}Cf [9]

In the range a) mass yield of complementary fragments is continuously increasing, whereas in the range c) it is continuously decreasing and, in the range b) fission fragment mass yields are approximately same probable.

Calculated correlation coefficients are $-0.633, -0.994, -0.940$ in the mass ranges a), b) and c), respectively. The lowest correlation coefficient (-0.994) is found in the mass range b) that means almost complete anti correlation in the emitted from complementary fragments neutron numbers. A relatively slower anti correlation in the mass range a) is due to irregularities in $\bar{\nu}(A)$ behavior in Fig.3. Correlation coefficient estimate for the whole mass range $A_L^i(73-126), A_H^i(126-179)$ (54 fragment pair) is found to be -0.883. Note that experimental data have errors $10-50$ % at the mass numbers less than $A=80$ and higher than $A=172$. If neutron number data for these masses are excluded, then the coefficient of correlation calculated for the whole mass range (47 fragment pair) has value of $\rho=-0.965$. These results say that total average number of neutrons emitted by a pair of fragments approximately is constant, in agreement with the result of Terrell analysis [1] and with the results of [7]. Indeed, the total average number of neutrons, $\bar{\nu}^{tot}(A)=\bar{\nu}_L(A)+\bar{\nu}_H(A)$, illustrated in Fig.4 versus light fragment mass confirms this result in the fission fragment mass range 82-108; it is slightly increasing starting from mass 109 up to mass 116 and then is slightly decreasing.

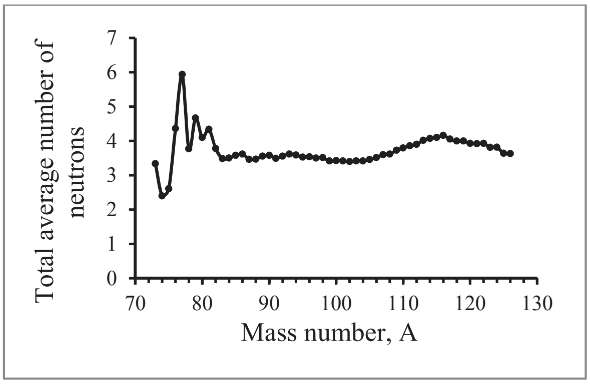

Fig. 4 Total average number of neutrons dependent on the light fragment mass.

At the fission fragment masses below 82 experimental errors, as mentioned above are high and therefore calculated values of average excitation energies at these masses are not used for concluding remarks. Some inaccurate generalization of this result can be made for the excitation energy, if the contribution of gamma emission is neglected and if it is supposed that every neutron carry out the same excitation energy from fragment. If all light and heavy groups of fission fragments are replaced by a single light and single heavy fragment, the excitation energies of light and heavy fragments are anti-correlated even the total kinetic energy of fission fragments is distributed like Gaussian. The sum of excitation energies of light and heavy fragments is constant in spontaneous fission of ^{252}Cf, even though energy release in fission is participated in the kinetic energy and excitation energy forms. In other words, the difference between the energy release and total kinetic energy of fission fragments for the different mass splits is unchanged. However, the energy of the emitted photons from fission fragments and accurate value of the excitation energy carried out by emitted neutron must be taken into account accurately, for the calculation of the excitation energy of fission fragments.

4. Calculation of the average excitation energy of the fission fragments and energy partition between the complementary fragments

To calculate the correlation between the average excitation energies of the complementary fragments we use the expression for the excitation energy of fission fragment as in Refs. [1, 22, 23]

$$\bar{E}_X=\bar{\nu}(A)\cdot[\bar{B}_n(A)+\bar{\varepsilon}(A)]+\bar{E}_\gamma(A). \quad (7)$$

Here $\bar{B}_n$, is the neutron binding energy averaged over the cascade neutron emission and charge distribution for the fission fragment of mass A, $\bar{\varepsilon}(A)$ is the emitted neutron average energy in the center of mass system of fission fragment, $\bar{E}_\gamma(A)$ is the average energy of the gamma quantum emitted from fragment of mass A. The $\bar{\nu}(A)$ and $\bar{\varepsilon}(A)$ values are taken from [9]. We use the estimation given in work [2] for the average neutron binding energy, $\bar{B}_n(A)$ of fission fragment of mass number A. Mean gamma energy is calculated by the use of the expression [11]

$$\bar{E}_\gamma(A) = \left(6.6867 - 0.15578\frac{Z_f^2}{A_f}\right)\cdot\bar{\nu}(A) + \left(0.11127\frac{Z_f^2}{A_f} - 2.2408\right)MeV \qquad (8)$$

for a fission nucleus of charge number Z_f and mass number A_f. This reduces to the expression [7]

$$\bar{E}_\gamma \cong [0.75\bar{\nu}(A) + 2]MeV \qquad (9)$$

for ^{252}Cf spontaneous fission, which is the approximation used in description of the experimental data (see [7]). Behavior of $\bar{E}_\gamma(A)$ as a function of A, is similar to the behavior of of $\bar{\nu}(A)$. The results of the calculations of the average excitation energy in the mass range (89-167) are shown in Fig.5 (solid curve) (experimental data for $\bar{\varepsilon}(A)$ available in work [9] starts at $A = 89$). The calculation result of work [14] (dashed curve), based on an assumption that total excitation energy is partitioned between the fission fragments in the same ratio as the numbers of prompt neutrons emitted by these fragments, is also shown in this figure. Some discrepancy is seen in the heaviest mass regions of the light and heavy fission fragments.

The correlation between the average excitation energies of complementary fission fragments is calculated like the correlation coefficient in neutron average multiplicity (see Eq.(5)) and its value is -0.855. We see that the anti correlation between the excitation energies is lower than that for average neutron numbers (compared to -0.965).

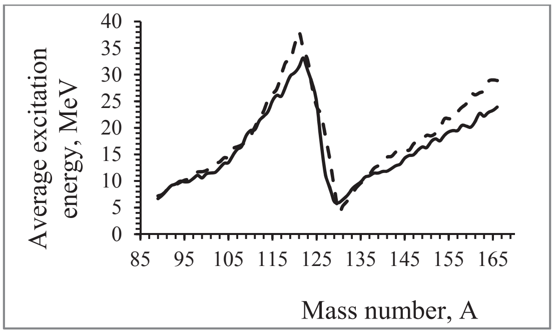

Fig. 5 Average excitation energy of fission fragments dependent on the fragment mass calculated by the use of Eqs. (7, 9) (solid curve) in comparison with the calculation result of the work [14] (dashed curve).

The ratio of fission fragment excitation energy to the total excitation energy of complementary fragments is another characteristic of excitation energy partition between the fission fragments. This ratio has the form

$$R = \frac{\bar{E}_X(A_L)}{\bar{E}_X^{tot}}\,. \qquad (10)$$

We calculate total excitation energy from the relation

$$\bar{E}_X^{tot} = \bar{E}_X(A_L) + \bar{E}_X(A_H) \qquad (11)$$

and the result is given in Fig.6.

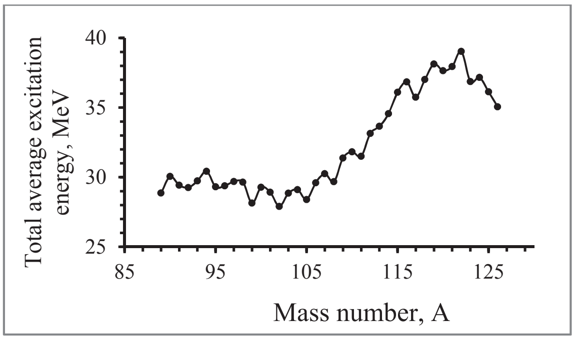

Fig. 6 Average total excitation energy of the pair of fission fragments dependent on the light fragment mass calculated by the use of Eqs. (7, 9, 11)

The total average excitation energy of complementary fission fragments is found in the range $28 - 30MeV$ in the light fragment mass range (89-108). Then, it increases up to 39 MeV at the mass number $A = 122$ having maximum value, and then decreases to the value of $35MeV$ at the symmetric mass split. The increase in the total excitation energy starting of the mass 109 is due to the increase of all factors-total average number of neutrons, neutron average binding energy, and neutron average energy in the center mass system. These results may be compared with the experimental data of work [12], on $12 MeV$ proton induced fission of ^{232}Th related with asymmetric fission mode of compound system ($p + {}^{232}Th$). In work [12] the measured average total excitation energy for asymmetric fission mode has values ranging between $(28-32)\,MeV$ in the light fragment mass range (84-100). This result is in agreement with our calculation result for the mass range (89-109). At higher masses, the experimental data illustrate (at $A > 100$) similar behavior in total excitation energy.

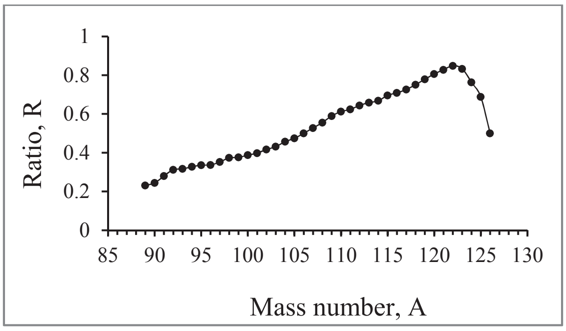

Fig. 7 The ratio (R) of the fission fragment average excitation energy to the total average excitation energy of complementary fragments versus the light fragment mass calculated by the use of Eqs.(9,10)

The result of the calculation of the ratio R, given by Eqs.(10)-(11) is shown in Fig.7. As it is seen in the figure, starting with light fragment mass $A = 106$ up to symmetric mass split at $A = 126$ light fragments carry out excitation energy higher than the heavy fragment ($R > 1/2$) when 85% (maximum) share of the total excitation comes to the light fragment at A=122. However, this means that at this mass number the relation

$$1 - \bar{E}_X(A_L)/\bar{E}_X^{tot} = \bar{E}_X(A_H)/\bar{E}_X^{tot} \qquad (12)$$

becomes minimum, i.e. heavy fragment of mass $A = 130$ carries 15% of total excitation energy. Generally, in the fission fragment mass range $(89-122)$ the ratio R is gradually (approximately, linearly) increasing with the average slope of 0.018. This slope in work [12] was found to be 0.02.

5. Discussion and conclusion

The partition of the excitation energy between the fragment pair in the spontaneous fission of ^{252}Cf is investigated in terms of the correlation coefficients between the average neutron number integrated over the total kinetic energy distribution of fission fragments at the definite mass split, and between the neutron average number from complementary fragments in a whole mass distribution range of fission fragments. It is shown that there exists, approximately zero correlation between the neutron numbers at the definite mass split corresponding to the average light and heavy fragment masses $A_L = 108$, $A_H = 144$. In terms of the excitation energy, this means that the excitation energies in complementary fragments in fission are distributed independently. This fact is used for claculation of the prompt fission neutron spectra [23,16]. Whereas, there is approximately, complete anti correlation between the average neutron numbers including all fission fragment masses. This means that the equation $\bar{\nu}_L + \bar{\nu}_H = const.$ approximately holds through the whole fission fragment mass range.

The correlation between the excitation energies of complementary fission fragments calculated in a whole mass range becomes weaker compared to that of in neutron average number, because of influence of both the neutron average binding energy and the neutron average center of mass energy. In fact, the correlation coefficient for the excitation energies is -0.855.

Thus, the mechanism of total excitation energy partition between the fission fragments may be analyzed in detail by the use of experimental data on the neutron numbers and the average gamma energies dependent on the fission fragment mass and on the total kinetic energy of fragment pair. There is no dependence on theoretical prescriptions in such study and result does not depend on them but it is determined by experimental data only. Therefore, the results in such a study may, in principle, be used to test different theoretical models such as nuclear state equation and nuclear level density dependence on excitation energy.

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