DECAY HEAT OF IRRADIATED NUCLEAR FUELS – A STATUS REPORT FROM THE NEA WPNCS

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ABSTRACT

Decay heat from spent nuclear fuel (SNF) is a crucial quantity for many aspects of the back-end fuel management (including but not restricted to interim storage, transport and final repository), for the design of safety systems in case of reactor transient or loss of coolant. In most cases, decay heat remains a calculated quantity, mostly depending on burnup, cooling time, fuel type and enrichment, and irradiation history. It has been measured in a limited number of SNF cases (PWR and BWR, UOX fuels), with characteristics not systematically overlapping with today's needs. Direct validation of decay heat calculations therefore relies on a restricted set of experimental data, partially correlated, and exhibiting large gaps in burnup and cooling times coverage.

The Working Party on Nuclear Criticality Safety (WPNCS) of the OECD Nuclear Energy Agency has established a subgroup (SG12) to reflect the state-of-the-art and current needs in terms of decay heat evaluation, covering code simulations capabilities, standard methods, nuclear data libraries and availability of experimental data. It aims, for example, at discussing the current understanding, including biases and uncertainties, for decay heat estimations, arising from calculation methods and the quality of nuclear data libraries, or that of fuel

fabrication and irradiation data. From the experimental aspect, the subgroup is also assessing the needs for new measurements, helping to prioritize in terms of SNF burnup, fuel types, and cooling time. Such perspectives, leading to robust estimations of calculated uncertainties and biases, are at the heart of the SG12.

KEYWORDS

Decay heat, Validation, Spent Nuclear Fuel

1. INTRODUCTION AND BACKGROUND

Spent fuel characterization (SFC) is one of the key activities in demonstrating that Spent Nuclear Fuels (SNF) meet the safety requirements related to the back end of the fuel cycle. This back end generally consists in moving the SNF from reactor core to wet storage (spent fuel pool), then from wet to dry storage, and finally towards reprocessing facilities or deep underground final repository. Additionally, the decay heat characteristics of fuel assemblies during their irradiation also play an important role in case of core loading and reactor transients, or accident such as loss of coolant. In these examples, the cooling time of interest for SFC spreads over several orders of magnitude, from seconds after reactor shutdown up to 10^5 years. Such range of time is in itself a challenge regarding our understanding of the evolution of the SNF with the change of environment, technical and administrative requirements, and finally with ensuring that the SNF knowledge goes through time and societies. There are no stricter requirements in any other industry with respects to its waste management.
Many quantities besides decay heat are of importance for the characterization of SNF. They concern the fuel and cladding behavior, radioactivity, gamma and neutron source strengths, nuclide migration and corrosion. Regarding decay heat, it is to be understood as the power released from the fuel that originates from the decay of unstable nuclides, eventually emitting alphas, gammas and betas, or decaying by spontaneous fission. In the reactor core, decay heat provides around 7% of the overall core power throughout the cycle. After reactor shutdown, it naturally evolves with cooling time, decreasing from about 7 % of the thermal reactor power (in the order of megawatts) at the shutdown time, down to fractions of watts, as presented in Fig.1 for two different SNF assemblies.

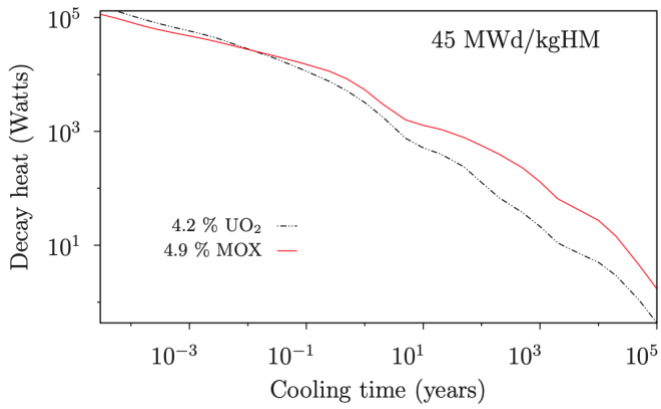

Figure 1. Example of calculated decay heat for two types of SNF assemblies.

A large number of the SFC quantities, besides decay heat, have the same origin, being the "source term", representing the nuclide inventory, usually considered at the end of life of the fuel assembly. A nuclide inventory is simply a list of nuclides (for instance ^{244}Cm) and their concentrations (*e.g.* given in grams per metric tons of initial heavy metal), at a given time. Because the same source term is used, the decay heat is indirectly correlated with criticality and radiation protection (shielding) studies.
Decay heat is a key parameter for the thermal design of spent fuel transport and storage casks as well as the back-end fuel cycle facilities to demonstrate safety, hence with an impact on the cost. Too high

temperatures can contribute to the degradation of fuel and cladding, with heavy consequences to criticality safety, potentially up to melting in certain hypothetical accident configurations, or in the damage of the engineered barriers or of the host rock formation in the case of a geological repository.
Its level of knowledge, such as best estimates and uncertainties, has direct and important consequences on various design options, and is therefore under study for more than 80 years. After the first steps towards the understanding of decay heat [1]-[2], systematics (*i.e.* analytic formula) were first developed (see for instance [3]), slowly followed by more precise calculations based on the summation method [4]. More recently, international efforts were framed at the Nuclear Energy Agency (NEA) with dedicated meetings and benchmarks [5]-[7]. Today, the interest in decay heat is still very high, for similar reasons as in the past, with the additional need of increasing our confidence in estimations (better understanding of uncertainties and biases), for instance in support of design optimization procedures. Within this context, a new NEA subgroup was launched under the Working Party of Nuclear Criticality Safety (WPNCS), called SG12, in order to establish our current state of knowledge and to assess potential computational or experimental gaps with respect to today's SNF characteristics. A short summary of the subgroup's outlook is presented in the following, with first the experimental and modeling aspects, followed by recommendations for future developments.

2. DECAY HEAT ESTIMATION

The decay heat estimation relies manifestly on code prediction, due to the large number of SNF worldwide and the requirements for their characterization. But code prediction implies prior code validation, and validation signifies experimental values to compare with. As presented in the next three sections, assessing the calculation methods and measurements landscape allows to set the boundaries of the current knowledge, and to frame the needs for the future. We will then go through some details of measurements and codes, which will help to understand the proposed needs in section 3.

2.1. Experiments

As in many other fields, measurements are a necessary step for the understanding of a specific phenomenon, its modelization and the validation of the codes of interest. In the case of the decay heat for integral SNF assemblies, three dedicated experimental facilities have been operated worldwide, among which only one is still in use. In the US, the majority of the calorimetric measurements was performed at the GE-Morris operation facility in the 1980s [8]-[10]. The decay heat was deduced from the increase of the water temperature inside the calorimeter where an assembly was first placed, combined with calibration curves and measurements of the escape gamma radiations, thanks to monitors outside the calorimeter. A second calorimeter was located at the Hanford Engineering Development Laboratory (HEDL), at the EMAD facility, on the Nevada Test Site. This calorimeter, also operated in the 1980s, was a boil off type calorimeter [11]-[12], evaluating differential steam condensate collection rates to extract decay heat. Neither of these two facilities exist nowadays, and the only remaining operated calorimeter is at the Clab facility, in Oskarshamn, Sweden, operated by SKB [13]. Decay heat is also deduced measuring the temperature increase of the water in the calorimeter, compared to a calibration curve and combined with gamma radiation monitors outside the calorimeter. A list of measured assemblies can be found in [8] and newer measurements are listed in [14] as well as in a future EPRI report, expected to be published in 2023.
These calorimetric measurements come with a description of the fuel assemblies and their irradiation conditions. It is therefore possible to use them for code validation. Naturally, as these fuel assemblies were measured more than 15 years ago (except for the five assemblies reported in [14]), their characteristics do not systematically encircle today's fuel enrichment or discharged burnup (see section 3). For instance, the initial enrichments of the measured PWR assemblies were from 2.1 to 4.0 wt%, with average burnup values from 20 to 51 MWd/kgU, whereas today, fuel enrichments are closer to 5.0 wt% and assembly averaged burnup values at discharge are regularly reaching values higher than 51 MWd/kgU. Some types of fuels are also not measured at all: LWR MOX and enriched reprocessed uranium (ERU) fuels, but also VVER and CANDU types of fuels.

Apart from these measurements on full-length assemblies, other types of measurements are also of interest. The first group of measurements are the single fission pulse experiments, providing gamma and beta heat rates per fission for a number of individual fissionable nuclei, with dedicated monoenergetic neutron sources. Such measurements, comparable to several ones performed in the MINERVE reactor [15]-[16], have been performed for more than half a century, and were compiled in [17] and [18]. One of the main advantages is that the measurement time starts at a few seconds after the fission burst, and can last up to one day, leading to unique and valuable data for code validation in the event of core transients.

A last set of measurements, being mostly used for the validation of nuclear data libraries, consists of the Fusion decay heat experiments [19], which can be used to fulfil the gap of experimental data for fission plant structural materials and for materials under high energy irradiation conditions.

Finally, the estimation of SNF decay heat can be correlated with post irradiation examination (PIE) and the measurements of specific nuclide concentrations [20], initially often performed in the framework of criticality studies. Even if in the framework of criticality studies not all nuclides relevant for decay heat are necessarily measured, those measurements serve to validate the methods (neutron transport and depletion code) used to derive the nuclide inventory and ultimately the decay heat. For completeness, one can also mention proprietary experiments such as MERCI in France [21], and full-scale measurements for fast reactors as on JOYO [22] and Superphénix [23].

As a last remark, it is worth mentioning the ongoing projects for the design, building and the use of new calorimeters in Switzerland and in France. This would help in filling the gaps in the currently available decay heat measurements.

2.2. Prediction and validation with best-estimate codes

Decay heat predictions are essential given the number of worldwide SNF awaiting for final repository, reprocessing, or simply in storage facilities. As safety needs to be demonstrated with sufficient margins, code validation is mandatory; uncertainties and biases in the predictions then translates in provisions or additional constraints on the design, with an associated economic cost. Among the different types of codes, best-estimate ones present the advantage of considering the least approximation possible (for nuclear data, geometry, irradiation history, calculation methods), such as based on Monte Carlo transport codes (e.g. Serpent, MCNP, VESTA, COMPASS, ALEPH-2), or deterministic codes (e.g. ORIGEN, Polaris, CASMO5, DARWIN2.3).

It is convenient to separate calculated cases as a function of the availability of experimental data. In the case of decay heat values large enough to lead to overwhelming measurement obstacles, one has to rely on code prediction without direct methods of assembly-scale validation. This is for instance the case for short cooling periods, related to core transients. The other extreme case concerns long cooling time, for which no measurements can yet be performed (e.g. more than hundreds of years cooling). Whereas this second case is of less practical importance (decay heat values are then in general small enough to be far below limits of interest), predictions for shorter cooling time (below a few days) can represent direct safety concerns. In such a case, indirect code validations for SNF can be performed based on single fission pulse experiments or fixed fission rate experiments, for instance using compilations from [17] or [18], as presented in [24] or with integral data experiments (e.g. Fusion Neutron Source and Frascati Neutron Generator) as presented in [25]. One of the recent notable improvements based on these compilations, combined with new measurements on nuclear structure, is related to the reduction of the impact of the so-called Pandemonium effect [26] on beta decay data from conventional nuclear data libraries by their substitution with total absorption spectroscopy measurements [27] (underestimation of the decay heat due to the lack of specific transitions), as indicated in Fig. 2. As observed for the JEFF library, the agreement between calculations and compilations is greatly improved with the latest release, and it will also reflect on the SNF decay heat calculations, with better prediction for the gamma decay following the fission of a nucleus of ^{239}Pu (note that for the electromagnetic contribution in the case of the thermal fission of ^{235}U, not all differences are not yet resolved for short cooling times) [28,29].

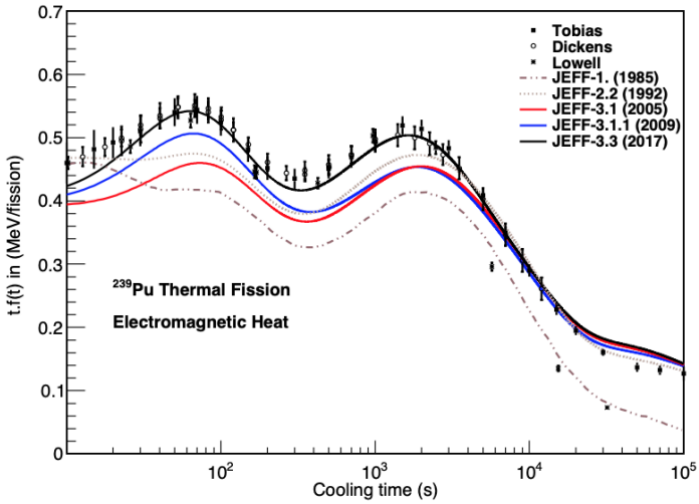

Figure 2. Electromagnetic decay heat component for a pulse irradiation of ^{239}Pu with thermal neutrons.

Regarding the direct calculations of decay heat for SNF, there is a relative abundance of publications comparing calculated (C) and experimental (E) values (expressed in the following with C/E ratios). We cannot list them all here, and to mention only three recent examples, interested readers are referred to the work of Ilas and Liljenfeld [30], Jansson et al. [14], and Shama et al. [31]. Such calculations are based on a variety of best-estimate codes, nuclear data libraries and methods, and contrary to the previous example with a fission pulse, they concern the estimation of the full SNF assembly decay heat. They are almost systematically based on the available description of the same irradiated fuel assemblies, as indicated in Section 2.1: [8]-[13]. Naturally, the conclusions of the published comparisons between C and E are not the same, however they are relatively similar. Fig. 3 (left) summarizes in a simplified manner the results of many published C/E ratios. As observed, the general agreement between C and E values is on average good, with a value of 1.004 for almost 1500 published ratios. The standard deviation of this distribution can be considered large, being 5.4 % (the judgement of large or not is depending on prior expectations), but given the variety of publications, being part of validation or research studies using different codes and nuclear data libraries, it can be expected to observe such value. For comparison, values in [30] (Clab only data) are 1.002 ± 0.012 (1σ) for PWR and 0.997 ± 0.024 for BWR, whereas [31] (Clab and GE-Morris data) gives 1.007 ± 0.071 for both PWR and BWR cases.
From this perspective, one can conclude that the SNF decay heat for measured cases can be reproduced without noticeable bias.

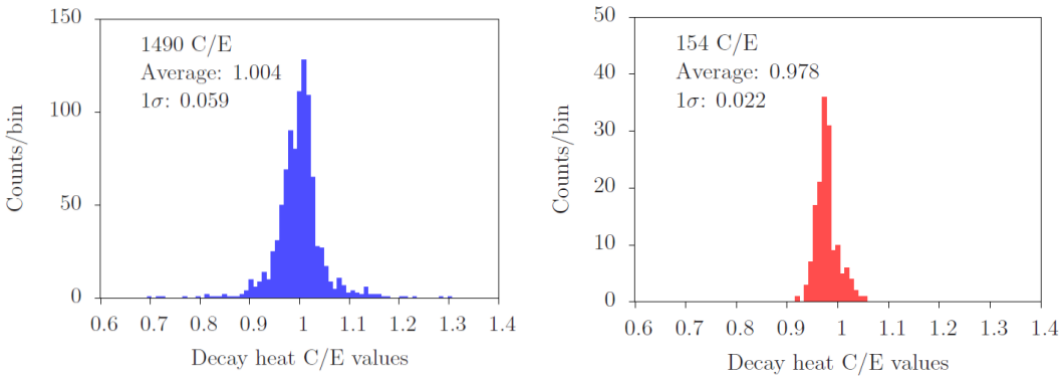

Figure 3. Histograms of the ratios of calculated (C) over experimental (E) decay heat values. Left: from literature studies for calorimetric measurements. Right: blind test for 5 SNF from [14].

This was well encouraging, until the latest published results, based on calculations performed without access to experimental decay heat values, indicated different prediction power [14]. In this publication,

the design and irradiation characteristics of five PWR assemblies were provided to a number of participants, performing the SNF decay heat calculations, without having access to the measured values. About 30 different calculations were performed for each assembly, based on different codes, nuclear data libraries, and users. The results of this study are summarized in Fig. 3 (right). As observed, the standard deviation is more than twice smaller than for the simple data collection presented in Fig.3 (left). Nonetheless, there is a noticeable bias, with, on average a C/E value or -2.2 %. Without analysing in details these results (it was also not done in [14]), this illustrates the complexity of the predictions, as well as the evaluation of measured quantities, based on a small set of experiments. An additional possible source of dispersion is the so-called "user effect", as observed in a number of benchmarks, due to modelling choices (partly to compensate for calculation code limitations), understanding of the system description, and eventually different interpretations.

2.3. Predictions with standard methods

Instead of using best-estimate codes, users have the possibility to apply standard methods, such as the ANS-5.1 from 2014 [32], U.S. Regulatory guide (RG) 3.54, revision 2 from 2018, the ISO 10645 from 2022 [33], the Japanese standard from 1991 [34] or the DIN 25463-1 from 2014 [35]. These methods make use of predefined equations with adjustable parameters, coupled with irradiation characteristics (burnup values, cooling time, fission rates), as well as fuel characteristics (e.g. initial enrichment). Although one does not need to run explicit calculations as applied with best-estimate codes, input quantities required by some standards such as fission rates per burnup step and for the most important actinides are not easily guessed, without a first assessment based on more detailed simulation.
One characteristic of these methods is that they contain a certain degree of conservatism. The application of the four standard methods (ANS, DIN, ISO and RG) are presented in Fig.4 for the same SNF assemblies for which experimental values are available.

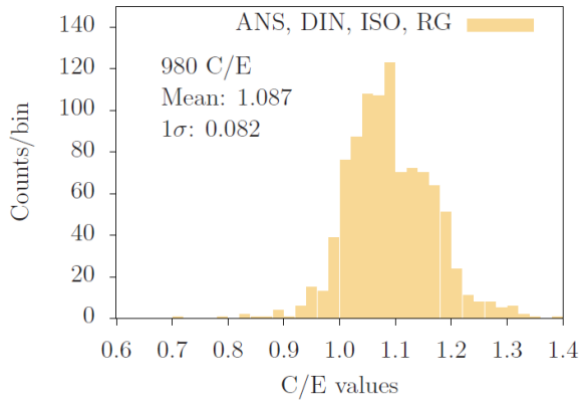

Figure 4. Same as Fig.3 (left), but with C coming from standard methods.

As observed, the average C/E values is higher than in the best-estimate calculations, indicating an amount of conservatism. The standard deviation is nevertheless of the same order of magnitude than the bias, showing that for some SNF, the C/E values are smaller than 1, implying that standard methods do not systematically lead to decay heat values higher than the experimental ones. This also indicates the entanglement of decay heat estimation with standard methods: there is no certainty of conservatism.

2.4. Calculated uncertainties

Calculated uncertainties on SNF decay heat can have numerous sources, including nuclear data, irradiation history, assumptions in calculation methods and modelling and manufacturing tolerances. Depending on the assumptions on these quantities, for instance the uncertainty on the assembly burnup value, different decay heat uncertainties can be obtained. This is de facto what happens when scanning published values, where large variations between calculated uncertainties can easily be obtained. In the case of nuclear data, this is partly due to the lack of correlations between fission yields in the available libraries, leading to different assumptions by the authors. Generally, a better description of the

uncertainties associated to nuclear data or technological data, for both standard deviations and correlations, would be of high interest. If the C/E values as presented in the previous sections tend to be very similar and centred around 1, uncertainties on the other hand often evolve in wider scales.

3. CURRENT AND FUTURE NEEDS

As mentioned, the needs with respect to SNF decay heat are related to the prediction power (bias) and uncertainties, for instance to provide justified confidence intervals. Such confidence intervals can be used for the design of storage facilities or for safety calculations during a core transient. Section 2 has depicted the current state of knowledge, eventually showing current limitations regarding the prediction of the SNF decay heat and its uncertainties. As indicated, there is a consensus on the ability to reach limited biases, with the constraint that the available measurements are limited in number and originates from only three facilities (of which only one is currently active). The current situation for calculated uncertainties is less obvious, as different assumptions for input quantities are often considered.
It was also indicated that the range of applicability of the code validation does not systematically overlap with today's fuel characteristics. Fig. 5 compares for PWR UO_2 assemblies cases with measured decay heat (with a calorimeter, full dots) and cases from an existing set of SNF (open dots). Four of the most-relevant quantities are used in this figure: assembly burnup and initial enrichment, as a function of the estimated total decay heat and cooling time. For the measured SNF, the cooling time corresponds to the measured time, and for the existing set, a hypothetical date of 2040 is chosen.

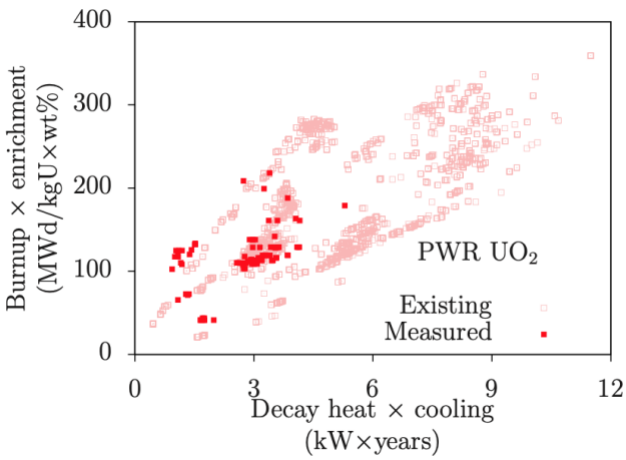

Figure 5. Comparison between an existing set of SNF with cases including decay heat measurements.

It can be observed that the overlap is not systematic, especially in the right and top parts of the figure, i.e. for long cooling time and high burnup (combined with high enrichment). There is therefore a clear need for additional measurements (with small experimental uncertainties and reliable uncertainty estimates), especially with more modern fuel assemblies. This includes higher average burnup (for instance up to 70 MWd/kgU), higher initial ^{235}U enrichment (at least up to 5 wt% for LWR needs, although HALEU fuel, or High-Assay Low-Enriched Uranium fuel, with enrichment up to 20 %, can be used in a close future in advanced reactors), and shorter as well as longer cooling times.
Independently of the acceptable biases and uncertainties, such existing validation concerns specific assembly designs (BWR and PWR without VVER, no CANDU design) and to UO_2 fuel type (no MOX and ERU). There is a clear lack of measurements (and validation) for the mentioned designs and fuel types, potentially affecting operational margins for specific facilities. Finally, Accident Tolerant Fuel (ATF) are already in use in commercial reactors, and no decay heat measurements currently exist for this new fuel or cladding type.

4. GOALS OF THE WPNCS SUBGROUP

In regards of the information described in the previous sections, a new subgroup was launched at the NEA under the Working Party on Nuclear Criticality Safety [36]. The goals of this group, called subgroup 12 (or SG12) is as follows:

- Gather interested participants from different horizons: industries, technical support organizations, waste management organizations, and safety authorities, in order to exchange information on decay heat for existing SNF: current knowledge, interest, needs.
- Raise the awareness of the current prediction capabilities, and limitations due to the lack of experimental data.
- Establish a state-of-the-art report regarding the decay heat for existing SNF, leading to discussions on existing biases and uncertainties, the impact of nuclear data libraries, assumptions in modelling, or irradiation history.
- Finally organize a decay heat benchmark, based on a fuel assembly with measured values, eventually to be started with a new dedicated subgroup.

The SG12 has started in January 2022 and is running for two years. About 50 participants from 12 countries and two international organizations have joined. It is naturally linked with national and international programs on SFC, such as the European projects called EURAD and EURAD-II, as well as with the International Atomic Energy Agency Coordinated Research Project on Spent Fuel Characterization (T13018).

5. CONCLUSION

Decay heat is one of the most important quantities from SNF characterization perspective. Like in criticality-safety, its understanding is of relevance over a very broad time range, from a few seconds up to one million years after reactor shutdown, for core transients and fuel cycle back-end. It can be measured and calculated, leading to specific validations, biases and uncertainties. Additionally, following the limited set of available experimental values, the validation of codes does not cover yet the range of existing SNF characteristics (high burnup, high enrichment, but also MOX fuel, ATF, VVER and CANDU designs). The subgroup SG12 of the WPNCS aims at assessing our understanding regarding the measurements and calculations of the SNF decay heat, with the long-term objective of providing recommendations (or evaluations like the ICSBEP handbook) of decay heat for specific SNF cases. Before reaching this ambitious goal, we have presented the current state of knowledge, experimental needs, and indicated different points of focus. Consequently, the SG12 is supporting new measurements, continuing modelization for best estimates and uncertainties, as well as preparing the path for future decay heat benchmarks.

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