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Assessment of Nuclear Data Needs for Advanced Reactor Demonstrations: Application to the Molten Chloride Reactor Experiment (MCRE)

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ABSTRACT

A number of advanced reactor concepts are planned for near-term demonstrations including microreactors, larger demonstrations and space nuclear systems. These reactor concepts are based on a wide variety of reactor technologies, including sodium, gas, and salt cooling. An overlooked area in the development and ultimate startup of these reactors is addressing nuclear data needs that allow confident prediction of the criticality, safety requirements, and operation of the reactors. In this paper we try to address the problem of assessing nuclear data needs and possible remedies to reduce the existing uncertainties for advanced nuclear reactors. A methodology for defining these needs is described. The case of the Molten Chloride Reactor Experiment (MCRE) has been considered and the related investigation highlights the specific needs for reducing uncertainty on the ^{235}U capture and ^{35}Cl (n,p) reactions. Relatively inexpensive integral experiments are indicated as possible solutions for significantly reducing the current associated uncertainties.

KEYWORDS: Nuclear data, uncertainty reduction, target accuracy assessment, advanced reactors, integral experiments.

1. INTRODUCTION

A number of advanced reactor concepts are planned for near-term demonstrations including microreactors, larger demonstrations and space nuclear systems. These reactor concepts are based on a wide variety of reactor technologies, including sodium, gas, and salt cooling. An overlooked area in the development and ultimate startup of these reactors is addressing nuclear data needs that allow confident prediction of the criticality, safety requirements, and operation of the reactors.

Many advanced reactor projects under development could require significant nuclear data support, including: the VTR (Versatile Test Reactor) [1,2], the molten salt fast reactor [3], advanced gas-cooled reactors [4], different types of microreactors [5], and nuclear reactors for propulsion in space [6]. Current evaluated nuclear data files for several isotopes produce quite large uncertainties (e.g., more than 1% Δk/k on critical masses and several tens of percent for safety reactivity coefficients).

These types of uncertainties substantially increase the required safety margins of core and fuel cycle assessments, and consequently, significantly raise the costs of any advanced reactor systems aiming at optimizing fuel cycles and management of radioactive waste for enhanced safety and economy.
In this paper we will assess the nuclear data needs of the Molten Chloride Reactor Experiment (MCRE). First, we will describe the employed methodology, then the practical case of the MCRE will be investigated, by first providing the uncertainty on the k_{eff}, and then assessing the corresponding nuclear data needs. Finally, some recommendations are provided by using possible integral experiments for reducing the current nuclear data uncertainties.

2. METHODOLOGY

In order to assess the nuclear data needs the TAR (Target Accuracy Requirements) methodology described in [7,8] is adopted.
In the following we provide a brief description of the methodology we will employ for establishing the mentioned data needs. One of the key quantities to be used in this are the sensitivity coefficients that allow quantification of the variation of an integral parameter due to a change in a given cross section. They can be viewed as first derivatives of the integral parameter with respect to a cross-section variable. These quantities are computed using a new developed version of the SERPENT code [9].
Using a covariance data matrix (i. e. standard deviations on diagonal, and correlations on off-diagonal term) $\boldsymbol{D}$ and sensitivity coefficient arrays (in other words derivatives of a change of a cross section with respect to an integral parameter) $\boldsymbol{S_R}$ for an integral parameter $\boldsymbol{R}$, one can calculate the uncertainty □$\boldsymbol{R}^2$ on the integral parameter using the sandwich formula:

(1)

A successive step is the assessment of target accuracy requirements. Target accuracy assessments are the inverse problem of the uncertainty evaluation. To establish priorities and target accuracies on a data uncertainty reduction, we adopt a formal approach by defining target accuracy on the design parameter and finding out required accuracy on data. In fact, we can obtain the unknown uncertainty data requirements by solving a minimization problem where the sensitivity coefficients in conjunction with the existing constraints provide the quantities needed to find the solutions.
We can obtain the unknown uncertainty data requirements $\boldsymbol{d_i}$ (e.g., for variables $\boldsymbol{i}$ not correlated among themselves) by solving the following minimization problem for the functional $\boldsymbol{Q}$:

(2)

with the following constraints:

(3)

where $\boldsymbol{N}$ is the total number of integral design parameters, $\boldsymbol{S_{ni}}$ are the sensitivity coefficients for the integral parameter $\boldsymbol{R_n}$ and are the target accuracies on the $\boldsymbol{N}$ integral parameters; $\boldsymbol{\lambda_i}$ are "cost" parameters related to each $\boldsymbol{\sigma_i}$ and should give a relative figure of merit of the difficulty of improving that parameter (e.g., reducing uncertainties with an appropriate measurement).
When we take correlations into account, the constraints of Eq. (3) become:

(4)

where , as in Eq. (3), represents the uncertainty related to the standard deviations of the selected variables:

(5)

are the correlation terms among the selected variables with both $\boldsymbol{i}$ and $\boldsymbol{i'}$ going from 1 to…$\boldsymbol{I}$:

(6)

$\boldsymbol{Corr_{ii'}}$ is the correlation value between variable $\boldsymbol{i}$ and $\boldsymbol{i'}$. are the correlations among the selected and unselected variables:

(7)

where $\boldsymbol{d_j}$ are standard deviations that are not variables but considered constants and $\boldsymbol{K}$ is the total number of constant terms correlated to variable $\boldsymbol{i}$. Finally, $\boldsymbol{P_n}$ represents the constant residual uncertainty for integral parameter $\boldsymbol{R_n}$ due to the unselected variables (i. e. cross sections with very low contribution to total uncertainty).

The following tasks are executed for obtaining the accuracy requirements for the pertinent nuclear data of the advanced reactor under consideration:

- Computation of sensitivity coefficients for the integral parameters.
- Calculation of the covariance matrix for a series of isotopes and reactions needed for the uncertainty quantification. A seven-energy group [10] structure (see Table I), useful for meaningful cross section accuracy requirements, has been adopted.
- Uncertainty quantification on the selected reactors and integral parameters.
- TAR for the reactor and integral parameters under consideration.

The major neutronic design parameters for which sensitivities and target accuracies are computed generally include, but are not limited to: critical mass, Doppler coefficients, coolant reactivity coefficients, control and safety rod worth, burn-up reactivity loss per cycle, delayed neutron fractions, power peaks, and secondary coolant activations.

3. APPLICATION TO MCRE

The nuclear data needs assessment has been limited to advanced reactors of the MCFR (Molten Chloride Fast Reactor) type. These reactors are under consideration to be built by TerraPower, first at an INL location within four years, and then for a demo reactor to be built in the following years.

The TAR nuclear data need methodology has been applied to the MCRE system using k_{eff} as the integral parameter. The MCRE will be the world's first critical fast-spectrum salt reactor. MCRE will demonstrate safe reactor control under a variety of operation conditions providing empirical nuclear data to support design and licensing of a MCFR Demonstration Reactor. The MCRE is a low power experiment to be carried out at National Reactor Innovation Center (NRIC)'s Laboratory for Operation and Testing in the United States (LOTUS) facility at the INL MFC (Material Fuel Complex) site.

Table I. Energy group structure

Group	Upper Energy (eV)
1	**$1.96403\ 10^{7}$**
2	**$2.23130\ 10^{6}$**
3	**$4.97871\ 10^{5}$**
4	**$6.73795\ 10^{4}$**
5	**$2.03468\ 10^{3}$**
6	**$2.26033\ 10^{1}$**
7	**$5.40000\ 10^{-1}$**

The sensitivity coefficients were generated using the SERPENT code and a model derived from Figure 1. The MCRE uses a HEU (93% enriched) chloride salt. The reflectors, both radial and axial, are made of MgO. The ENDF/B-VII.1 library was used for the k_{eff} calculation and for generating the 7-group cross section covariance matrix.

One important addition was made for the ^{35}Cl (n,p) reaction. Because of the lack of covariance data by ENDF/B-VII.1 in the first group of energy for this reaction a value of 50% was adopted for the standard deviation and a correlation of 50% with the second group of energy was also introduced. It is as well worth mentioning that the covariance data available for this reaction in the second group are limited to the

ground state.
Table II shows the k_{eff} uncertainty (1 σ), expressed in pcm, computed for this model. In red are highlighted the larger contributors to the total uncertainty. The sums are made in a statistically way. The ^{235}U $\mathbf{s^{cap}}$ is the largest contributor followed by the ^{35}Cl $\mathbf{s^{(n,p)}}$. Also, the ^{24}Mg $\mathbf{s^{el}}$ and the ^{235}U $\mathbf{c}$ give significant contributions.

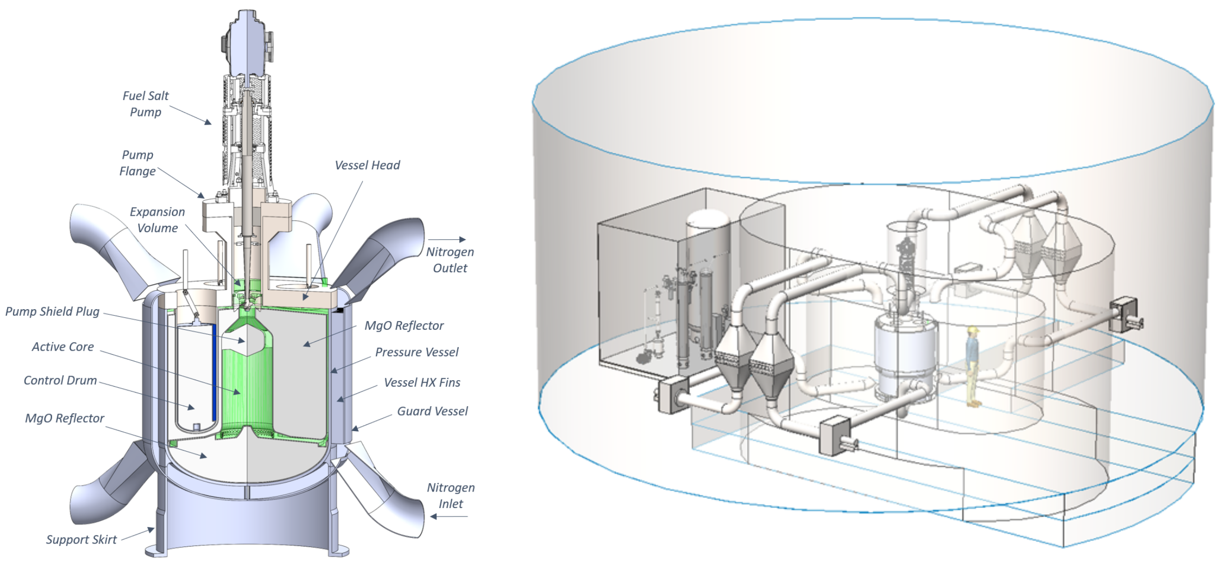

Figure 1. MCRE HEU salt.

Next step was to apply the TAR methodology imposing a constraint of 300 pcm for the total uncertainty on the k_{eff} of the reactor. Table III shows the initial and final, after applying the TAR methodology, uncertainty and their components as identified in Eq. (4). It is interesting to note that the negative sign in the ***F*** and ***P*** components occurs because negative contributors to the total uncertainty are excluded by the selection process. A total of 45 nuclear data were selected that are responsible for 100.7 % of the total uncertainty. The fact this value is larger than 100% is because the discarded nuclear data give a negative contribution to the total uncertainty. Table IV illustrates the target accuracy requirements for the first 20 nuclear data, where ranking is done by the largest contribution to the uncertainty reduction. The total amount of uncertainty reduction associated to these 20 nuclear data is of the order of 98%.
If we focus on the first five nuclear data requirements of table IV, which are responsible of ~92% of the total uncertainty reduction, it can be said that the needed reduction for the standard deviations is quite significant going from several tens of percent to just one or two precents.

4. CONCLUSIONS AND RECOMMENDATIONS

An attempt has been made to address the problem of assessing nuclear data needs for the case of the MCRE. The related investigation has highlighted the specific needs for reducing uncertainty on the ^{235}U capture and ^{35}Cl (n,p) reactions. It is quite unrealistic that this type of reduction could be achieved today only by differential measurements, but complementary integral experiments will be needed. This is even truer when knowing that for integral experiments the spectra can be adapted to match those of the target reactors. Relatively inexpensive integral experiments can be proposed as possible solutions for significantly reducing the current associated uncertainties.

Table II. MCRE k_{eff} uncertainty values (pcm) by isotope and reaction using ENDF/B-VII.1 data in a 7-energy group structure.

ISOTOPE	s^{el}	s^{inel}	s^{cap}	P_1EL	s^{fis}			$s^{(n,p)}$	SUM
^{16}O	230	10	46	23	0	0	0	0	236
^{23}Na	94	70	15	24	0	0	0	1	121
^{24}Mg	454	83	30	0	0	0	0	0	463
^{25}Mg	90	12	27	0	0	0	0	0	95
^{26}Mg	76	6	1	0	0	0	0	0	76
^{35}Cl	7	0	15	0	0	0	0	964	964
^{37}Cl	6	0	1	0	0	0	0	0	6
^{52}Cr	6	1	1	0	0	0	0	0	6
^{56}Fe	15	7	11	4	0	0	0	0	20
^{58}Ni	3	1	4	0	0	0	0	0	5
^{235}U	76	155	2131	28	201	95	278	0	2168
^{238}U	-7	21	5	1	2	4	2	0	21
TOTAL	537	191	2132	43	201	95	278	964	2435

Regarding the large uncertainty attached to the ^{235}U capture, they derive by differential measurements used by the evaluators for generating the covariance matrices. Use of integral experiments, like well documented benchmarks, could result in lower values. However, integral experiments of the separated effect type can be even more effective. To this end, irradiation experiments like the proposed MANTRA-2 (Measurements of Actinide Neutron Transmutation Rates at ATR) [11] could be used. The MANTRA-2 experiment is an irradiation in the Advance Test Reactor (ATR) reactor that will gather information for the capture cross section in the fast spectrum for the irradiated materials derived by the transmutation rate of the irradiated sample. Filters can be used for mimicking the fast spectrum of the target reactor.

The information that can be obtained from the post-irradiation analysis is related to the evaluation of the reaction rates (mainly capture) for a given isotope. In particular, the analysis of the experiment is based on the relationship between the atom build-up due to the transmutation and the related microscopic cross-sections.

Table III. MCRE k_{eff} uncertainty TAR results. Components defined by Eq (4).

		G	*C*	*F*	*V*	*P*	Total
Uncertainty (pcm)	Initial	1834	1620	-190	2440	-159	2
	Final	240	243	-39	339	-159	3

Table IV. MCRE k_{eff} uncertainty TAR results in terms of required standard deviation needed for obtaining a total uncertainty of 300 pcm on the k_{eff} uncertainty. The total uncertainty reduction associated to the shown 20 nuclear data amounts to ~98%.

Rank	Nuclear Data	Energy Range (eV)	Current (%)	Required (%)	Rel. Unc. Reduc. (%)
1	^{235}U □^{cap} Gr. 4	$6.74x10^4$ to $2.03x10^3$	33.2	1.2	47.5

2	^{235}U □^{cap} Gr. 3	4.98x10^{5} to 6.74x10^{4}	24.3	1.4	25.2
3	^{35}Cl □$^{(n,p)}$ Gr. 1	1.96x10^{7} to 2.23x10^{6}	50.0	2.0	13.9
4	^{235}U □^{cap} Gr. 2	2.23x10^{6} to 4.98x10^{5}	17.1	2.1	3.4
5	^{35}Cl □$^{(n,p)}$ Gr. 2	2.23x10^{6} to 4.98x10^{5}	11.2	2.1	1.7
6	^{24}Mg □^{el} Gr. 2	2.23x10^{6} to 4.98x10^{5}	5.3	1.2	1.6
7	^{24}Mg □^{el} Gr. 3	4.98x10^{5} to 6.74x10^{4}	5.6	1.2	1.3
8	^{235}U □^{cap} Gr. 5	2.03x10^{3} to 2.26x10^{1}	6.0	1.5	0.8
9	^{235}U c Gr. 1	1.96x10^{7} to 2.23x10^{6}	5.2	1.4	0.6
10	^{235}U c Gr. 3	4.98x10^{5} to 6.74x10^{4}	6.8	1.8	0.4
11	^{24}Mg □^{el} Gr. 4	6.74x10^{4} to 2.03x10^{3}	2.6	1.4	0.3
12	^{16}O □^{el} Gr. 2	2.23x10^{6} to 4.98x10^{5}	2.0	1.0	0.3
13	^{235}U □^{inel} Gr. 1	1.96x10^{7} to 2.23x10^{6}	17.3	3.6	0.3
14	^{235}U □^{el} Gr. 3	4.98x10^{5} to 6.74x10^{4}	2.5	2.5	0.3
15	^{16}O □^{el} Gr. 4	6.74x10^{4} to 2.03x10^{3}	2.0	1.2	0.2
16	^{16}O □^{el} Gr. 3	4.98x10^{5} to 6.74x10^{4}	2.0	1.2	0.2
17	^{235}U □^{el} Gr. 2	2.23x10^{6} to 4.98x10^{5}	3.7	3.4	0.1
18	^{23}Na □^{el} Gr. 2	2.23x10^{6} to 4.98x10^{5}	6.8	2.5	0.1
19	^{235}U c Gr. 2	2.23x10^{6} to 4.98x10^{5}	2.1	1.8	0.1
20	^{235}U □^{inel} Gr. 2	2.23x10^{6} to 4.98x10^{5}	6.9	3.1	0.1

For isotopes for which the descendant, obtained via neutron capture, is stable or has a long radioactive period, the most accurate experimental technique for obtaining information on the integral capture cross section is to determine the variation in composition that results from the flux irradiation of a pure sample.
In order to evaluate the isotopic ratios of the transmutation products the Multi-Collector Inductively Coupled Plasma Mass Spectrometry (MC-ICP-MS) can be used. This technique has insured uncertainties lower than 0.5% (2 σ level of confidence) in measuring the isotopic ratio to the respect of that of the sample with a sensitivity as low as 10^{-6} [12].
Filters can be used for mimicking the spectrum of the target reactor. For the MANTRA-2 the design shown in figure 2 was proposed, where a filter in a tube consisting of a 5 mm thick region of 97% ^{10}B

enriched B_4C is preceded by a 1-mm-thick region of Cd. Figure 3 shows the normalized ^{235}U capture cross section that would be obtained in the MANTRA-2 spectrum and in the MCRE.

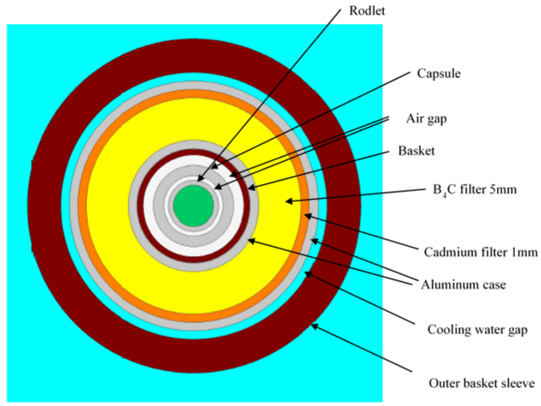

Figure 2. MANTRA-2 experimental configuration

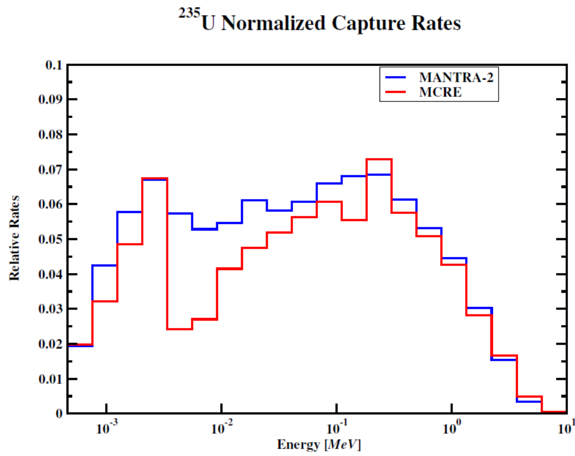

Figure 3. Comparison of normalized ^{235}U capture rates

Concerning ^{35}Cl (n,p), new differential measurements [13,14] have been recently performed and disagree quite significantly between them. Therefore, in [14] it is recommended that a full re-evaluation should be carried out. Hopefully, this will result in more reliable values with a lower uncertainty attached to this reaction. In parallel, it would be possible to perform useful integral experiments similar to what has been proposed for the MASSIMO (Measurement in Adapted Spectra of Spectral Indices and Material Oscillation) experimental campaign [11]. In this case a sample of chloride salt can be irradiated for a sufficient time (likely, few days) so that the daughter resulting from the ^{35}Cl (n,p) reaction ^{35}S (half-life of 87.4 days) has enough activity in terms of β^- decays that can be measured by either an ionization chamber or a scintillator, and in turn compared with the calculated corresponding values.

Again, filters can be used for obtaining the appropriate spectrum. Figure 4 shows one of the proposed filters of the MASSIMO program in an experimental dry assembly at the NRAD reactor, which contains a depleted uranium (DU) region followed by 2mm thick B_4C (97% ^{10}B enriched) region. Figure 5 compares the ^{35}Cl (n,p) normalized reaction rates obtained in the core region of the MCRE and in the sample region of the experimental NRAD assembly. The two sensitivity profiles are quite similar, but the experiment is more sensitive at higher energy, while the MCRE sensitivity is more pronounced between 1.5 and 3 MeV. With a suitable modification to the filter, for instance replacing 1 mm of B_4C with an appropriate moderator, the spectrum could even better match that of the MCRE.

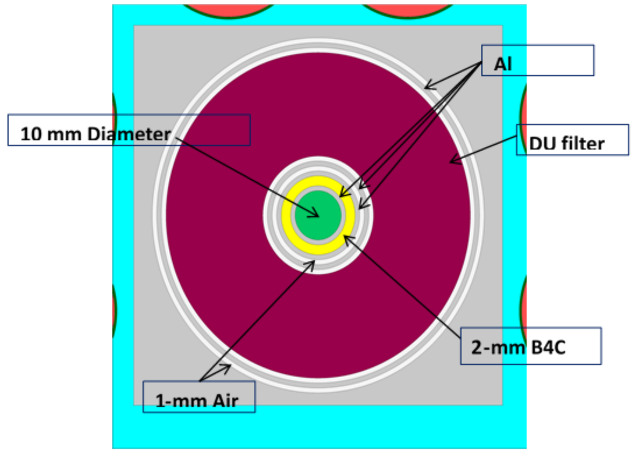

Figure 4. MASSIMO NRAD experimental dry assembly

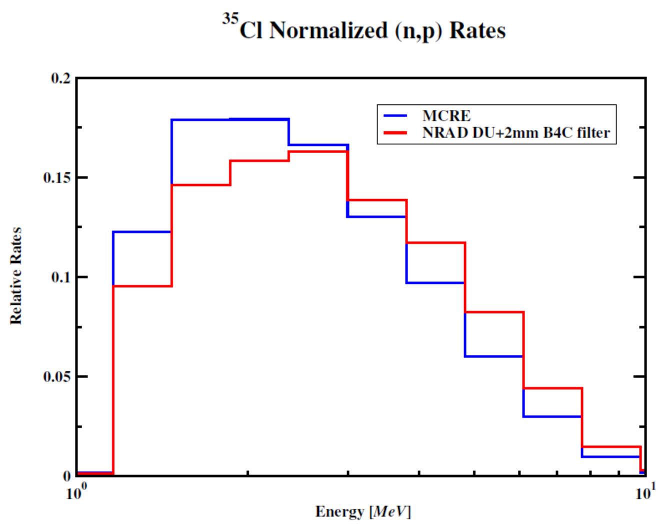

Figure 5. Comparison of normalized ^{35}Cl (n,p) rates

Given the multiple advanced reactor projects that have been flourishing in the last years and the possible use of not so conventional isotopes, it is likely that more nuclear data needs will arise as the neutronic designs of the systems incorporate uncertainty analysis. It would be useful to extend the same nuclear data needs assessment done here for the MCRE to other advanced reactors that have serious likelihood of being built. In order to perform further nuclear data assessment, access to more details about the neutronic design of the candidate advanced reactors will be needed.

A possible suggestion/recommendation could be that of developing and building, a dedicated and flexible experimental facility that could carry out many different types of experiments (integrals, elemental, separate effects, and development of measurement techniques) that could support a variety of needs associated to the design of advanced systems. A relatively small in size coupled thermal/fast system could be foreseen for this type of mission. In fact, such a type system would be able to simulate different neutron spectra ranging from thermal to epithermal, and fast.

ACKNOWLEDGMENTS

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