Nuclear Data Development Related to the Th-U Fuel Cycle in China

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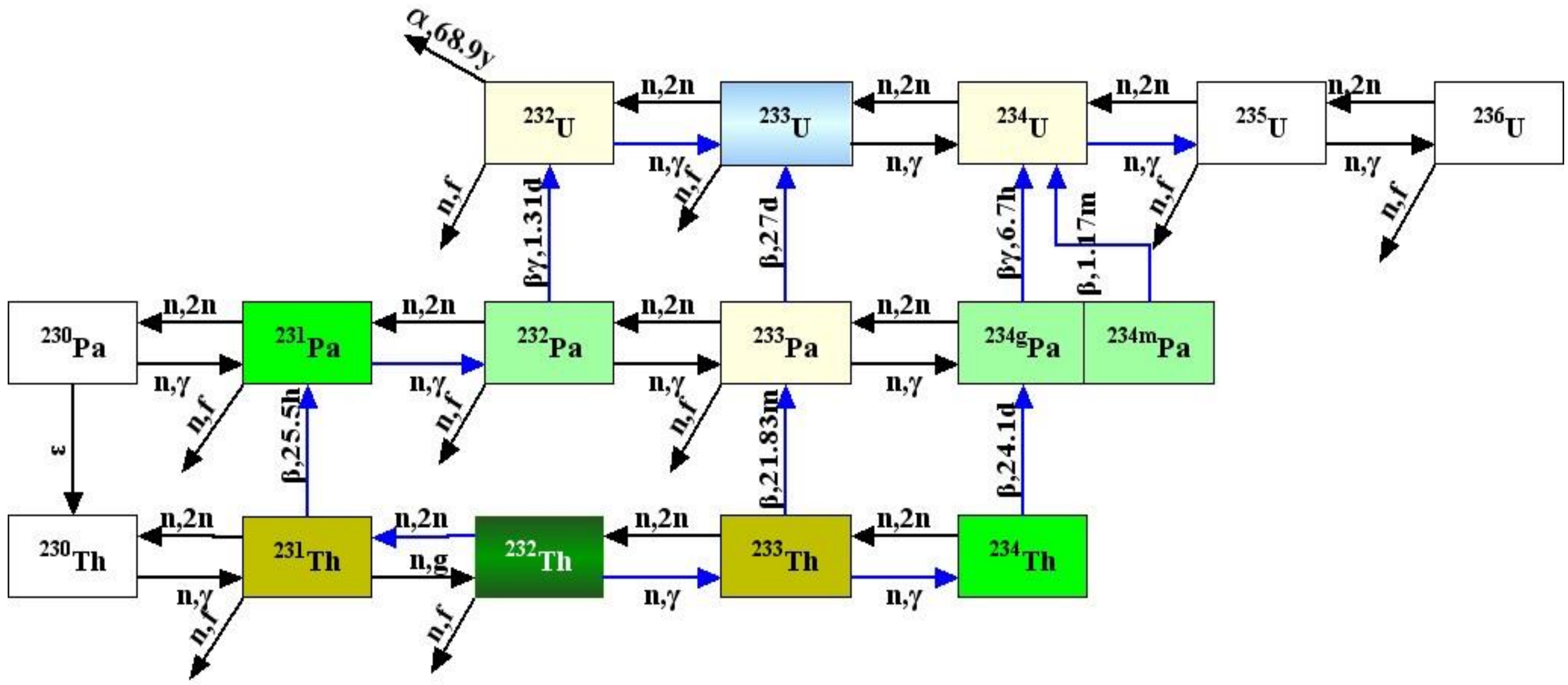
1. Introduction

- **In recent year, the Th-U fuel cycle, which is regarded as a supplement of the U-Pu fuel cycle, draws the nuclear community's attention.**
- **The top priority requirement of improving the nuclear data used in neutronics studies of thorium reactor.**
  - **The general propose nuclear reaction data for Th, Pa and U;**
  - **Decay data of the mass chain A=232,233 and 234;**
  - **Neutron induced fission product yields for ^{232}Th and ^{233}U, et al.**
- **To meet the requirement, quite limit research activities have been carried on in CIAE since 2010, including:**
  - Measurement of fission product yields induced by 14MeV n + ^{232}Th;
  - Study systematics of the thermal cross sections of actinides;
  - Evaluation and validation nuclear data for ^{233}U, ^{232}Th and et al.



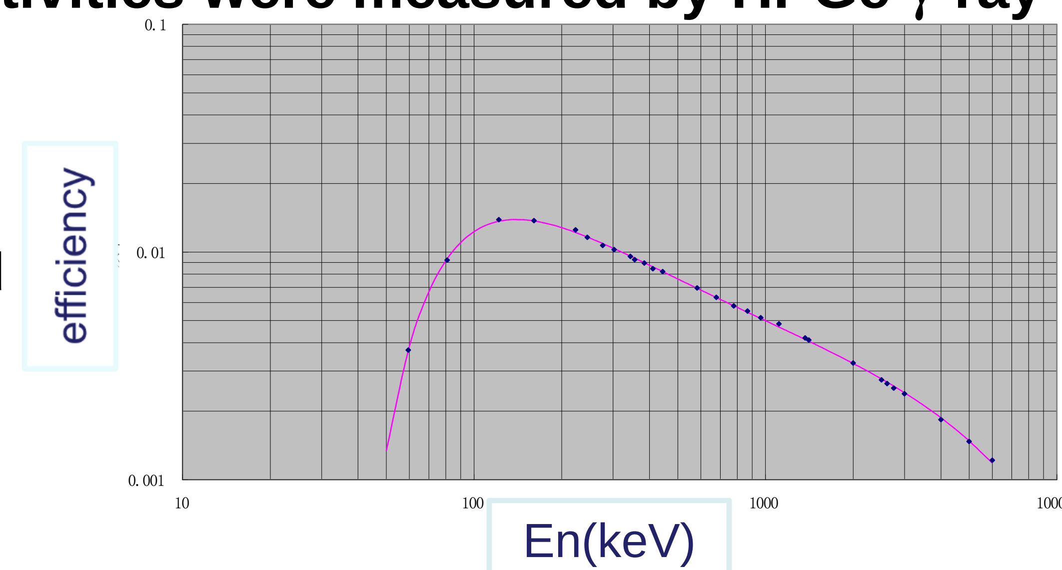
The transmutation chain of Th/U fuel cycle

- 12 key nuclides (Colored in the Figure)
    - Decay data, fission yields and general purpose reaction data.

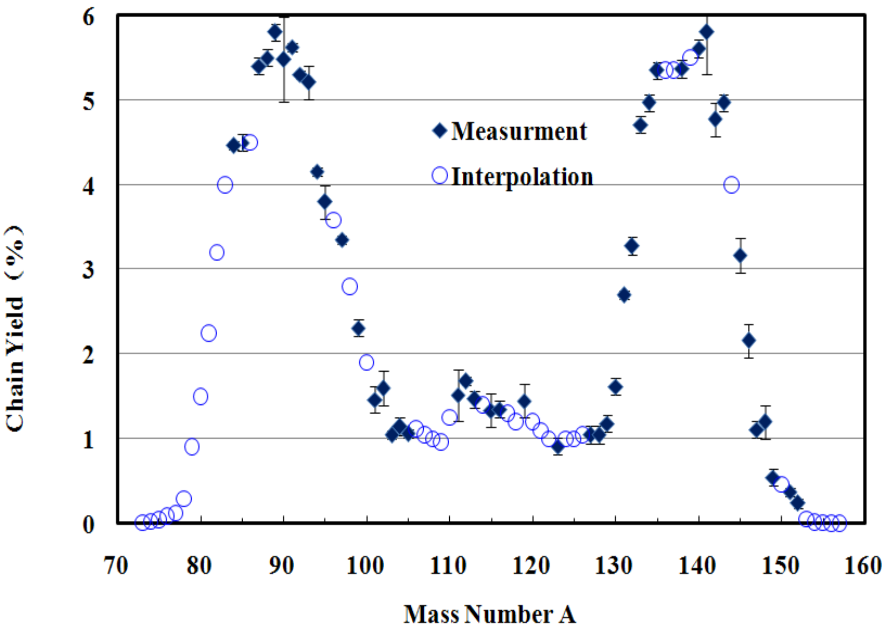
2. Measurement of nFP Yields for ^{232}Th

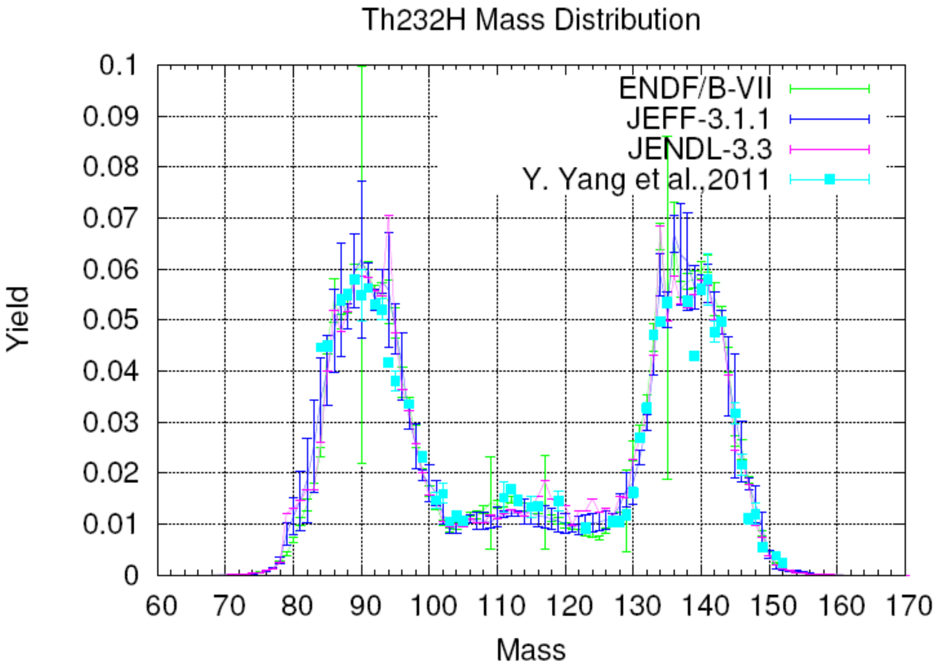
- **Fission Product Yields of ^{232}Th induced by 14MeV neutrons were measured by direct gamma spectrum method.**
  - Sample: 1 gram, 100% ^{232}Th, with Φ=22mm, covered by Al foils;
  - Neutron source: 14 MeV DT source with neutron flux nearly 10^{10} /cm^2/s on the sample;
- **Fission product activities were measured by HPGe γ-ray spectrometry.**

  - The efficiency curve from 59.5keV up to 5MeV was obtained by combination of measurement and M-C simulation.



- **Nearly 70% of the Yields were directly measured.**
  - Unmeasured product fields were interpolated or extrapolated.
  - All mass fields were normalized by 200%.
  - When $80 \le A \le 100$ and $130 \le A \le 150$, the relative uncertainties are less than 5% for most of the products measured.
  - Discrepancies > 20% were found when compared with evaluations.
- **The accuracy of the evaluated data may be not as good as we expected.**





3. Systematics study on thermal XS

- **It is difficult to evaluate thermal XS for some Th and Pa isotopes**
  - Short of theoretical method.
  - Evaluation is mainly based on experimental data.
  - Short of good experimental data for Th and Pa.

- **Systematics of the cross sections at thermal energy are necessary.**
  - No good systematics of thermal fission and capture XS for fission nuclides is available.

3.1 Systematics study on thermal fission XS

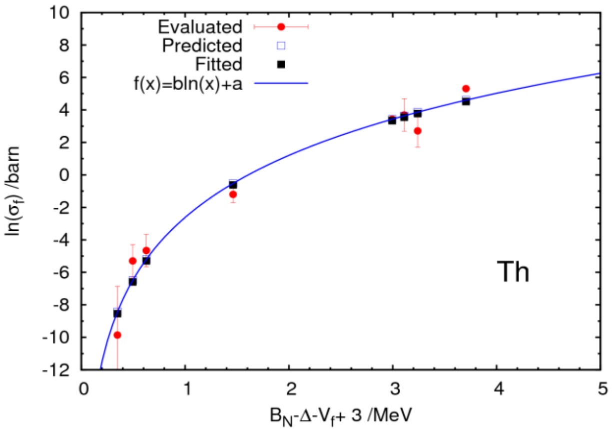
- **An experiential formula was assumed from Bohr-Wheeler formula.**

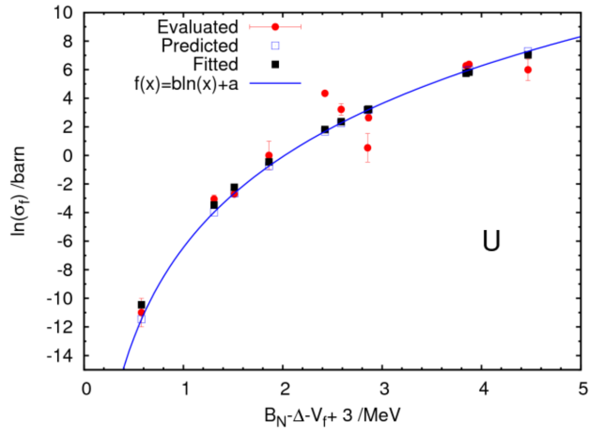
$$\ln\sigma_f = b\ln(B_N - \Delta - V_f + c) + a$$

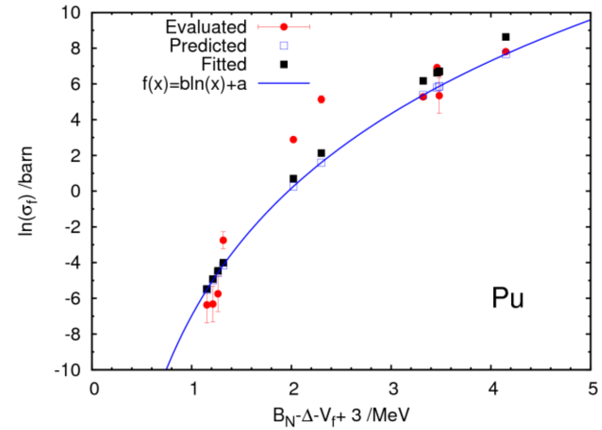
- **Thermal fission cross sections and uncertainties for 70 actinides were recommended.**
  - from ^{227}Th to ^{254}Cf;
  - CENDL-3.1、ENDF/B-VII.0、JENDL-4.0 and “Atlas of Neutron Resonances” (fifth edtion,BNL-325).

- **A set of single-peak fission barriers was also recommended.**
  - RIPL-3, CENDL-3.1 and JENDL-4.0.

When set parameter c = 3, the (n,f) XSs can be illustrated by the formula quite well.



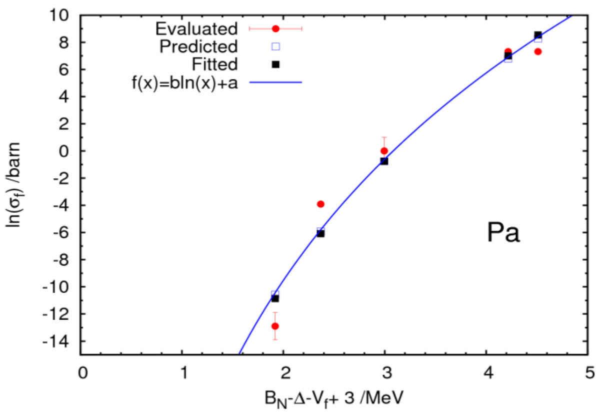


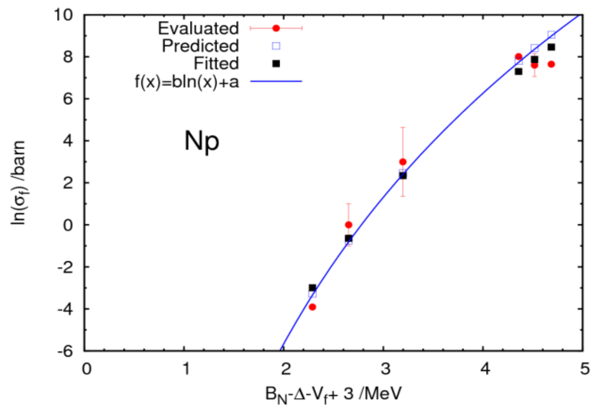


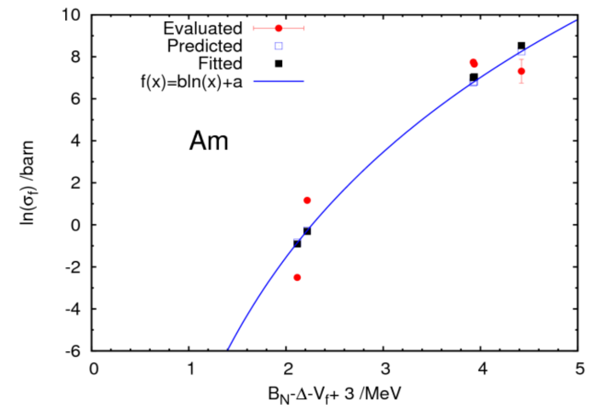
Even Z

Both odd-even effects of proton and shell effect in the neutron dependence are observed.

Odd Z



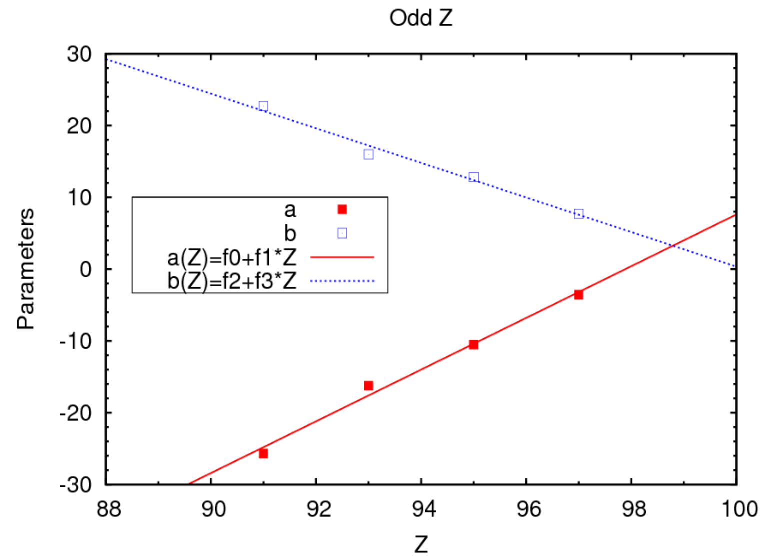


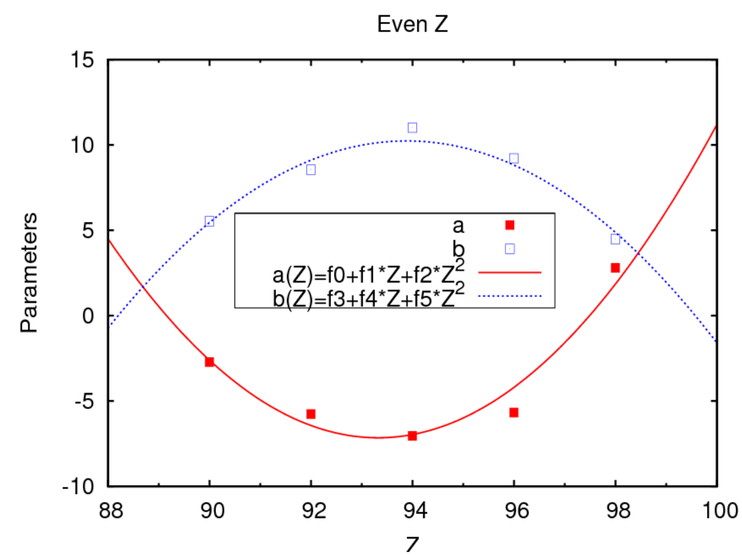


- **parameter a and b for each Z can be obtained by L-S fitting.**
- **When charge number is**
  - Odd: a/b can be illustrated by linear function of Z;
  - Even: a/b can be illustrated by quadratic parabola function of Z.
- **Then, a and b for each Z can be calculated in a general way.**
- **Final formula**

$$\ln\sigma_f = b(Z)\times\ln(B_N - \Delta - V_f + 3) + a(Z)$$

- **Thermal (n,f) XS becomes predictable by systematics.**



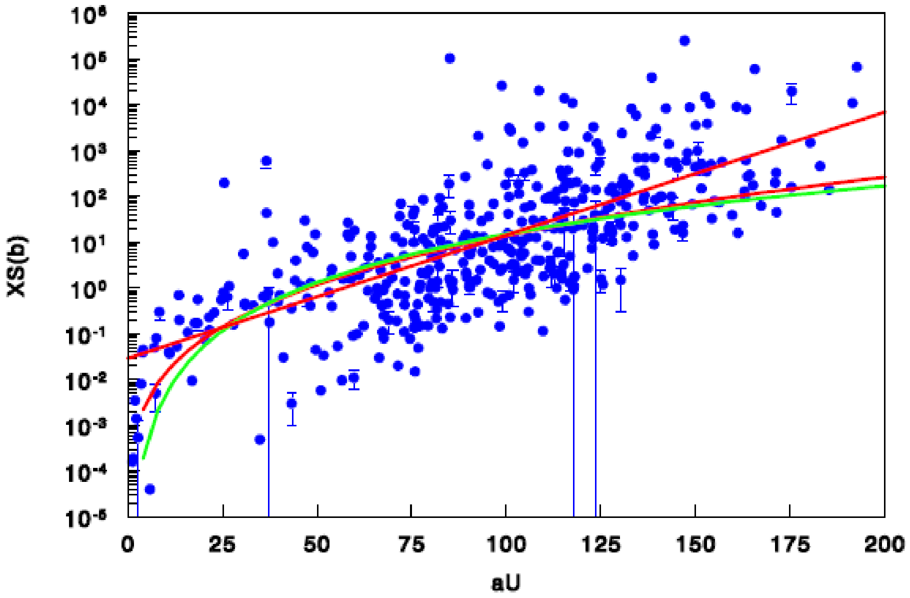


3.2 Systematics study on thermal capture XS

- **No systematics of thermal capture XS for actinides**
  - The Kopecky formula is established for 30keV.

Kopecky formula

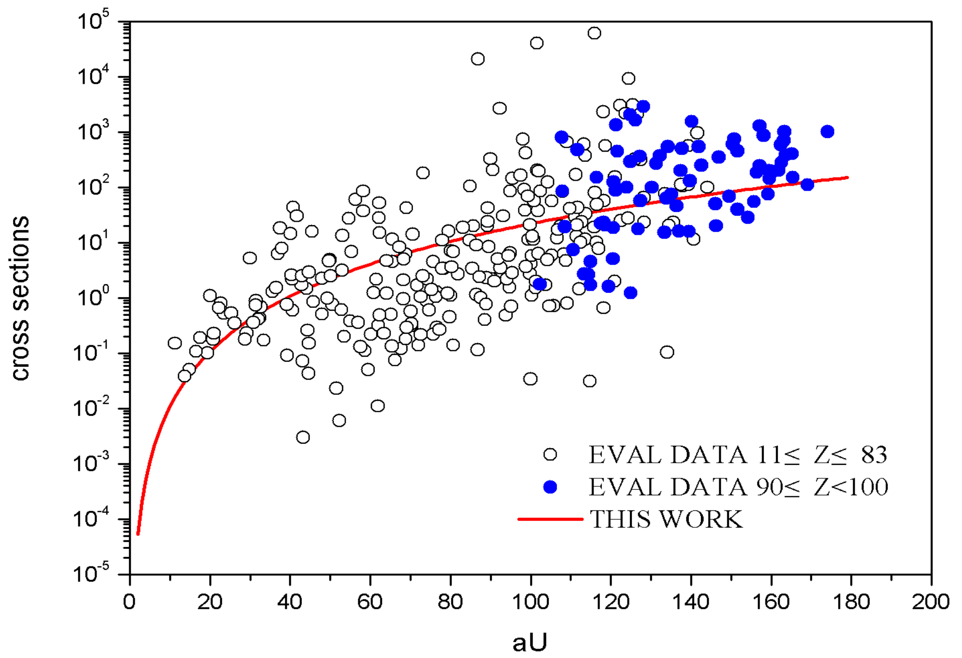
$$\sigma_{n\gamma} = C_1(aU)^{C_2}$$



(from EAF2009)

Thermal capture cross sections plotted against *aU.*
Blue points: evaluations
Red and green curves: systematics calculations

- **Try to decrease the divergence of the data but no significant improvement was observed.**
  - Update experimental database used in evaluation;
  - Update level density parameters of actinides in systematics formula;



$\chi^2 = 153726$ Kopecky

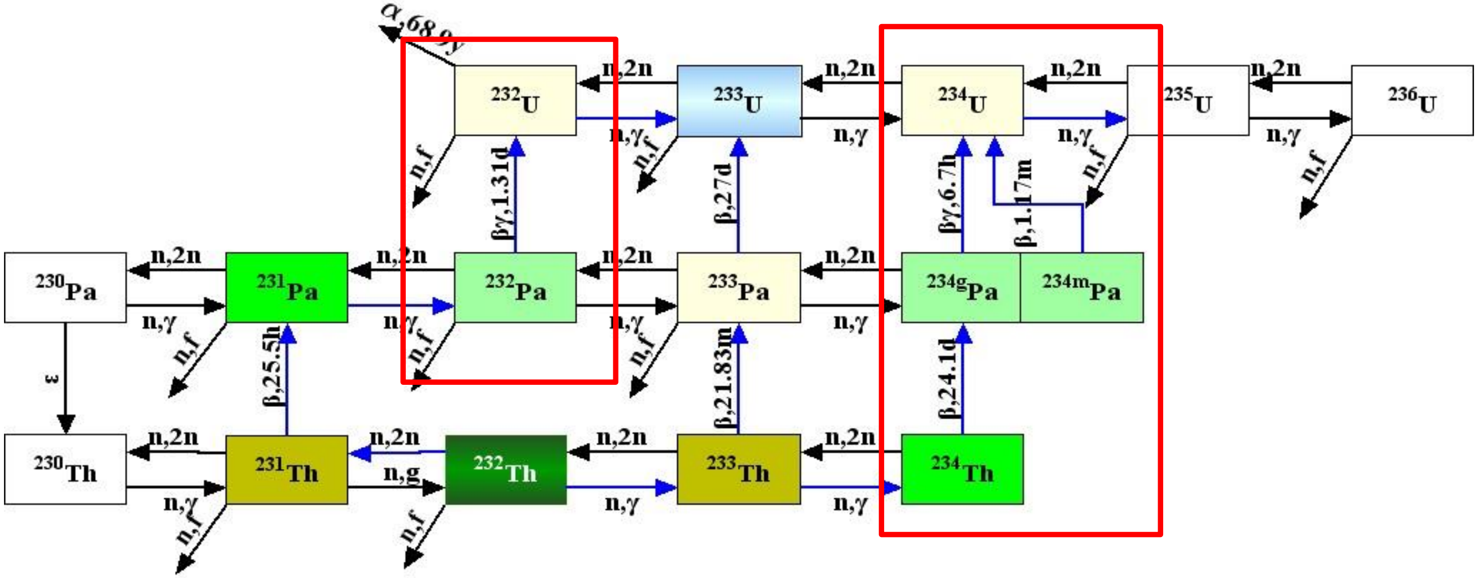
$\chi^2 = 48791$ This work

- **There is still no effective systematics to calculate thermal capture XS.**

4. Nuclear Data Evaluation Related to Th-U fuel cycle

4.1 Nuclear Decay Data and Structure Data

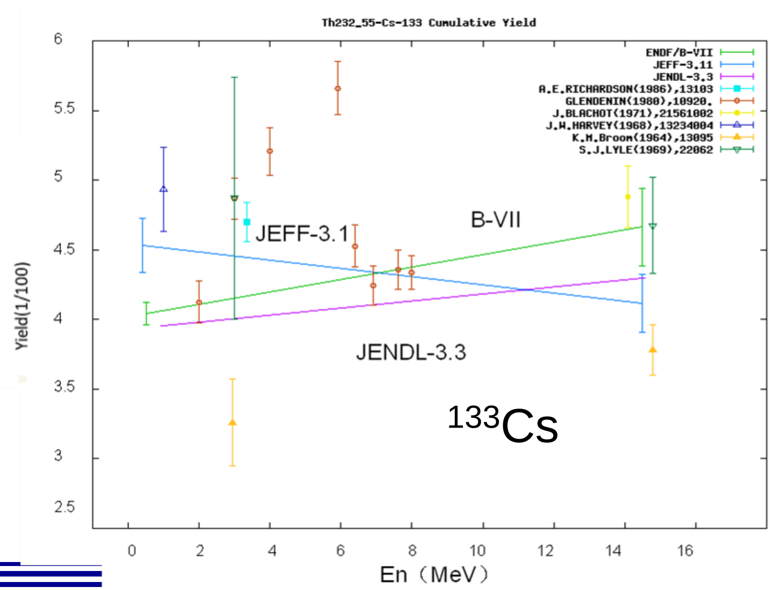
- Nuclear decay data and structure data in mass chain A=232 and A=234 related to Th-U fuel cycle have been re-evaluated.

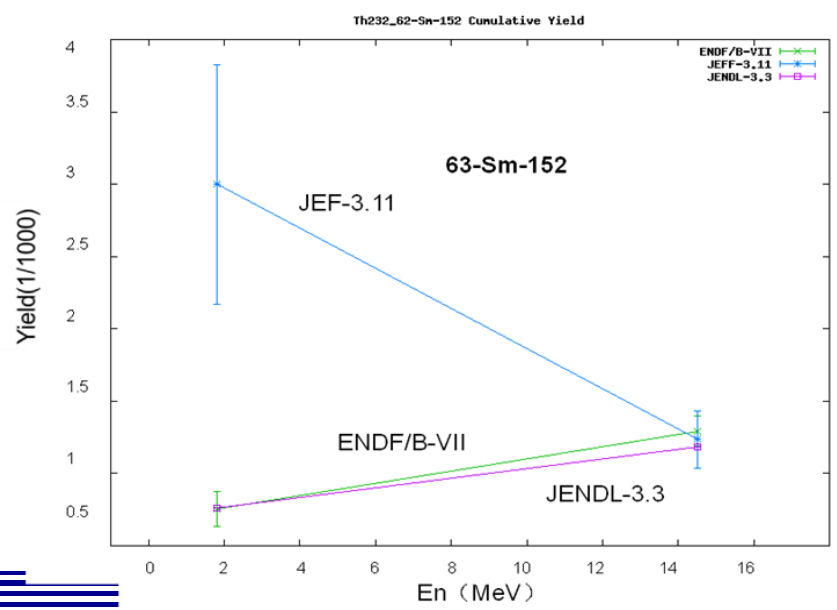


- No significant change of half-lives, except uncertainties and decay scheme.

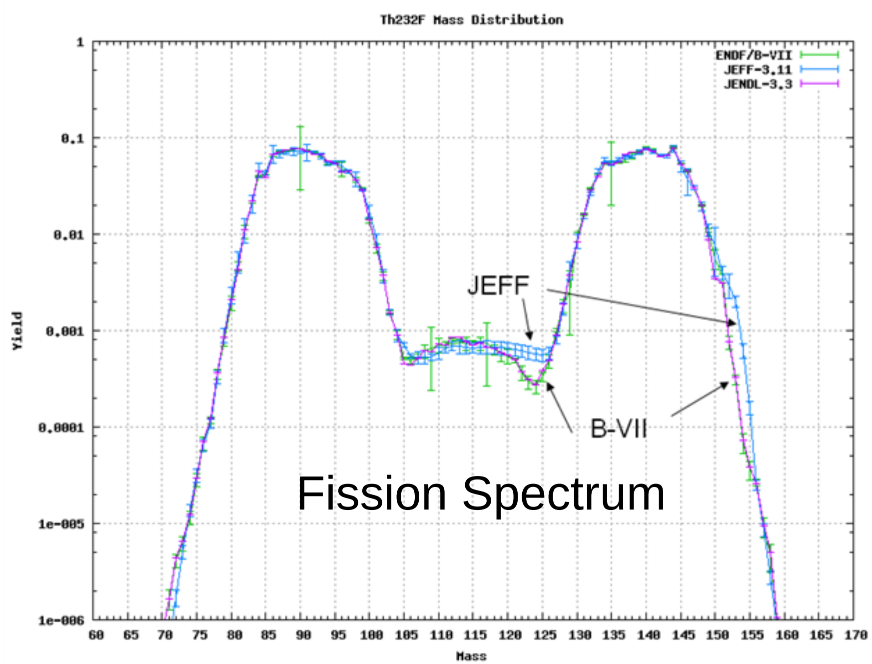
4.2 Evaluation of nFY for ^{232}Th

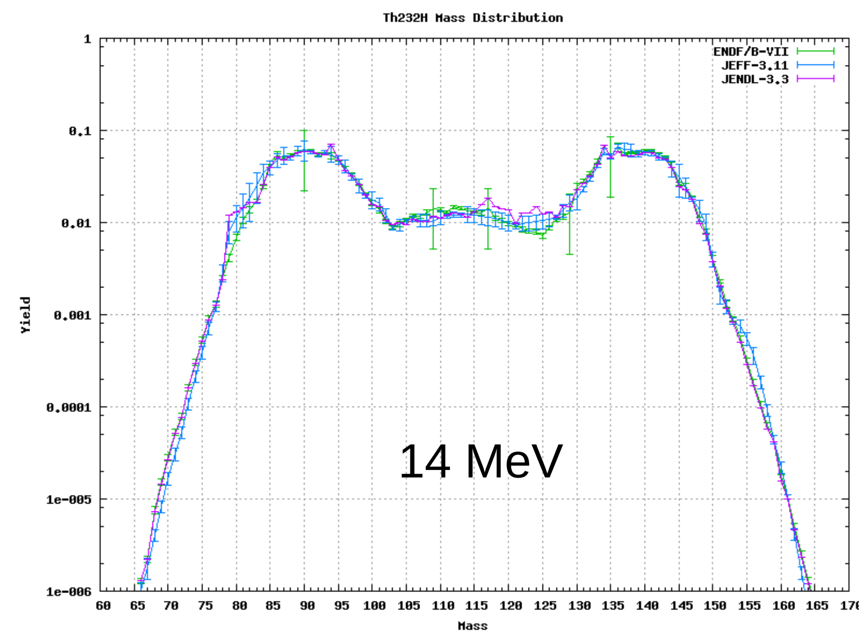
- **Accumulated fission yields and mass distribution were evaluated though comparison of the evaluations from ENDF/B-VII.0, JEFF-3.1.1 and JENDL-3.3.**
  - evaluation mainly focused on the yields that interested in burn-up credit calculation.
    - ^{95}Mo, ^{99}Tc, ^{101}Ru, ^{103}Rh, ^{109}Ag, ^{133}Cs, ^{147}Sm, ^{149}Sm, ^{150}Sm, ^{151}Sm, ^{152}Sm, ^{143}Nd, ^{145}Nd and ^{153}Eu.
  - For ^{133}Cs, JEFF is recommend; For ^{152}Sm, ENDF/B-VII.





- **The final evaluation is a combination of B-VII and JEFF, with the yields of ^{152}Sm and ^{153}Eu as well as the yields on the same chains adjusted.**



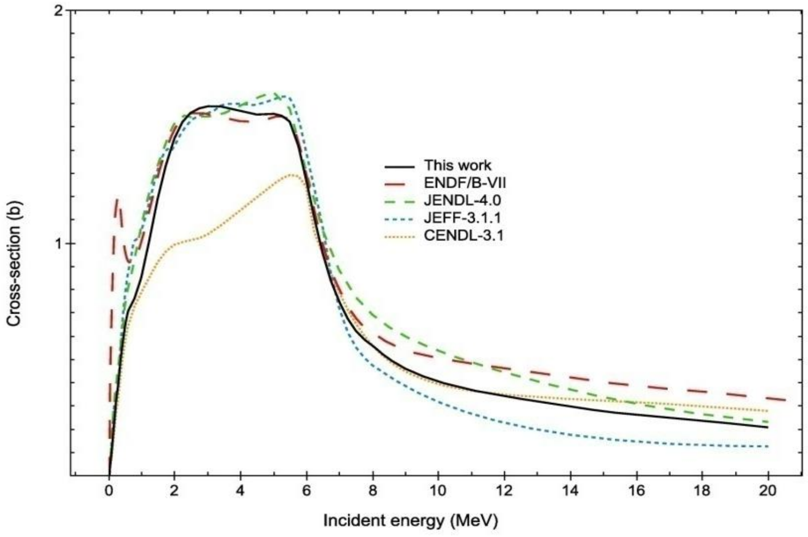


Mass Distribution

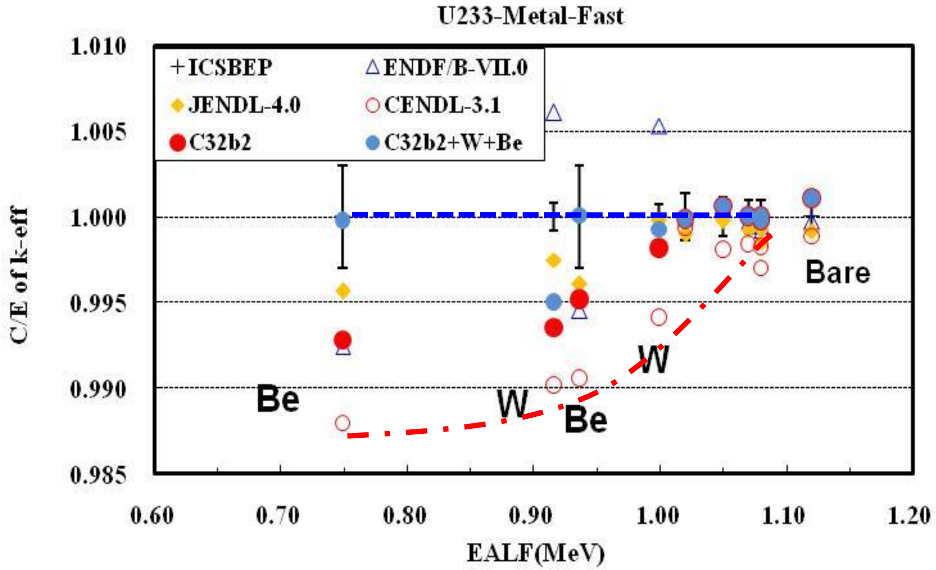
4.3 Nuclear Reaction Data Evaluation Related to Th-U

(1) Re-evaluation of neutron reaction data for ^{233}U

- **According the feedbacks from benchmarking results, an updated evaluation for ^{233}U has been recommend.**
  - (n, el) and (n, inl) cross sections have been improved;
  - fission cross sections, resonance parameters and angular distributions of (n, el) have been revised.



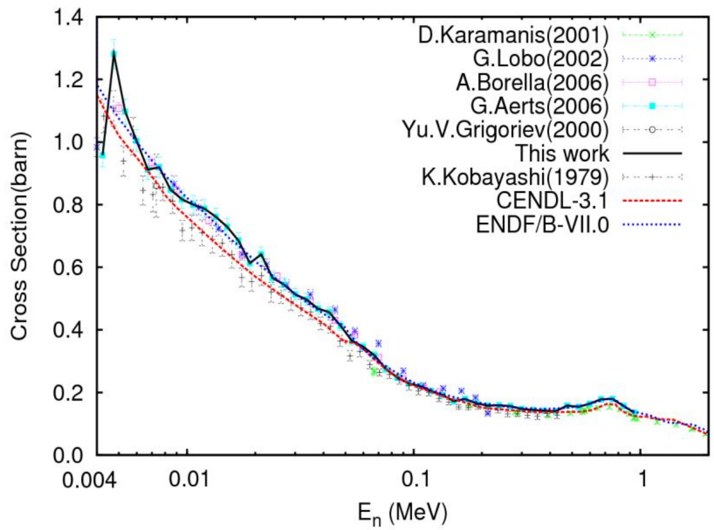
Comparison of ^{233}U(n,inl) cross sections



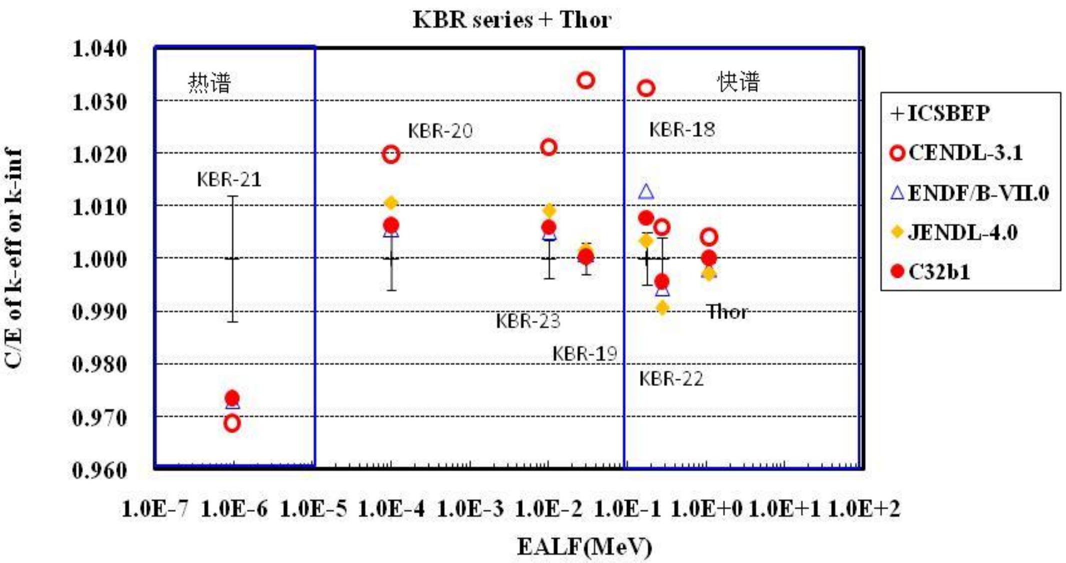
Comparison of C/E value for UMF system

(2) Re-evaluation of neutron reaction data for ^{232}Th

- The C/E values for KBR experiment with epithermal and fast spectrum are seriously over predicted by CENDL-3.1.
- ^{232}Th(n,γ) XS from 4keV to1MeV have been re-evaluated based on G. Aerts' work.



Comparison of ^{232}Th(n,γ) cross section evaluation with CENDL-3.1、ENDF/B-VII.0 and experimental data

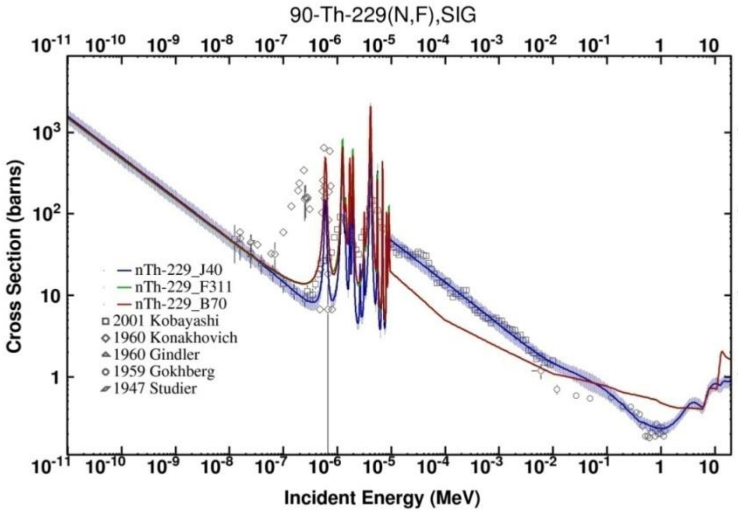


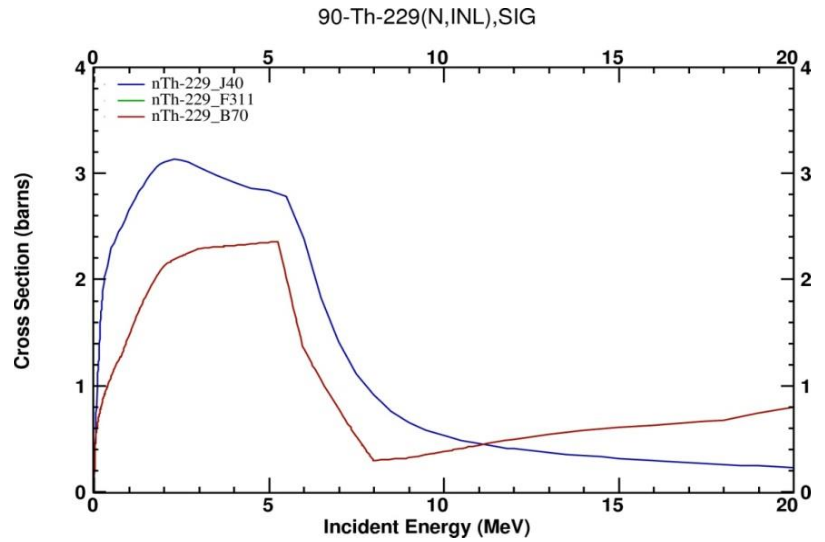
Comparison of C/E values for KBR and Thor

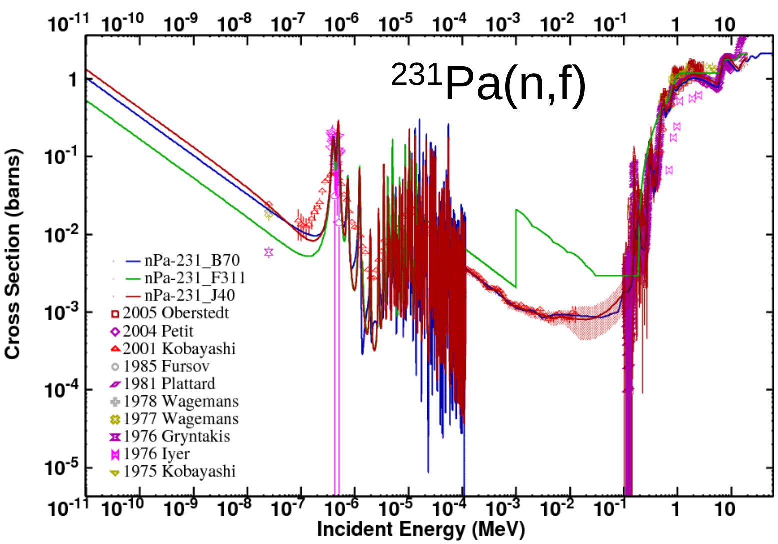
(3) Recommendation of ^{233,234}Th、^{231,232,233}Pa and ^{232,234,236}U

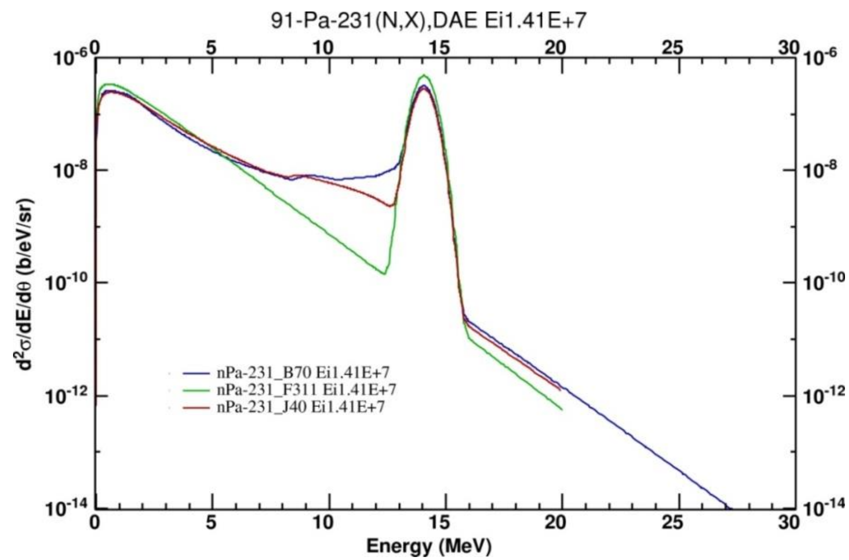
- **The nuclear reaction data evaluations for these isotopes have been done in ENDF/B-VII.0, JENDL-3.3, -4.0 and JEFF-3.1.1.**
- **Experimental data are quite limit for some nuclides.**
- **Better evaluation was selected by comparison among libraries.**
  - Theoretical calculation is reasonable;
  - Have good agreement with experimental data if available.
- **Recommendations are listed in following table.**

Evaluation Library	*Materials*
ENDF/B-VII.0	^{231,233}Pa、^{232,234,236}U
JENDL-4.0	^{227,228,229,230,231,233,234}Th、^{232}Pa


90-Th-229(N,F),SIG
Cross Section (barns)
Incident Energy (MeV)
nTh-229_J40
nTh-229_F311
nTh-229_B70
2001 Kobayashi
1960 Konakhovich
1960 Gindler
1959 Gokhberg
1947 Studier


90-Th-229(N,INL),SIG
Cross Section (barns)
Incident Energy (MeV)
nTh-229_J40
nTh-229_F311
nTh-229_B70


231Pa(n,f)
Cross Section (barns)
Incident Energy (MeV)
nPa-231_B70
nPa-231_F311
nPa-231_J40
2005 Oberstedt
2004 Petit
2001 Kobayashi
1985 Fursov
1981 Plattard
1978 Wagemans
1977 Wagemans
1976 Gryntakis
1976 Iyer
1975 Kobayashi

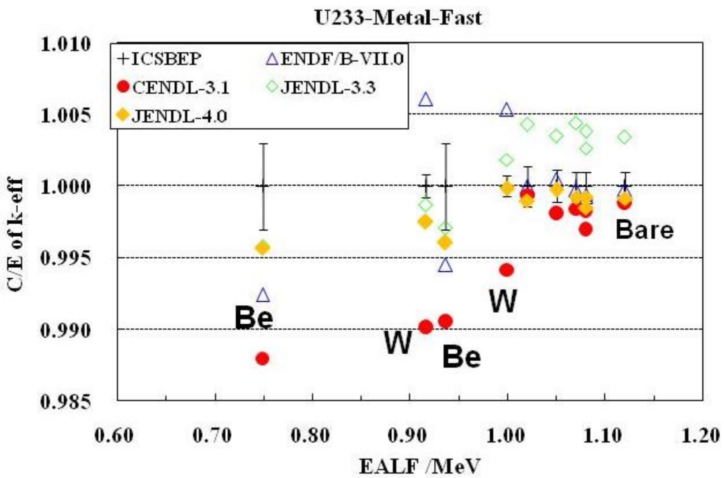

91-Pa-231(N,X),DAE Ei1.41E+7
d2σ/dE/dθ (b/eV/sr)
Energy (MeV)
nPa-231_B70 Ei1.41E+7
nPa-231_F311 Ei1.41E+7
nPa-231_J40 Ei1.41E+7

5. Nuclear Data Validation for ^{233}U and ^{232}Th

5.1 Validation of the evaluations before 2010

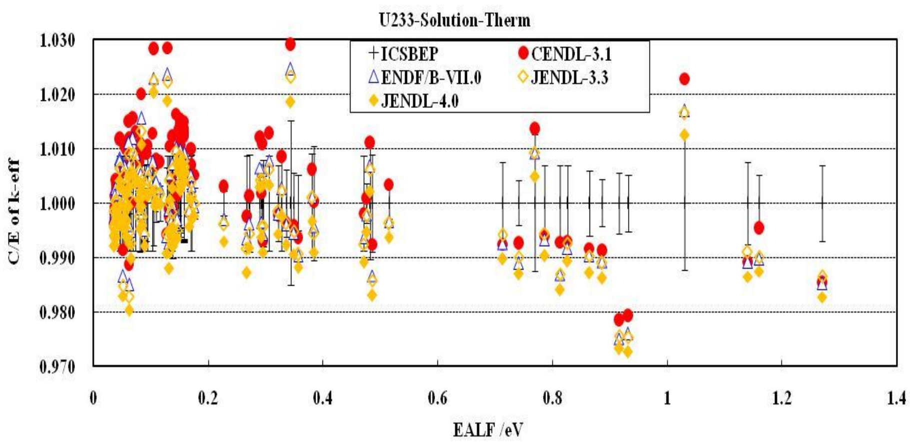
- **159 cases for ^{233}U and 55 cases for ^{232}Th were selected from ICSBEP2006 to test the evaluations in CENDL-3.1, ENDF/B-VII.0, JENDL-3.3 and -4.0.**
  - These benchmarks contains the fast, intermediate and thermal spectra systems.
  - The criticality calculations were done by MCNP5.
  - And all ACE files used were generated by NJOY99 with the same processing parameters.
  - Thermal scattering data comes from sab2002.

^{233}U Cases

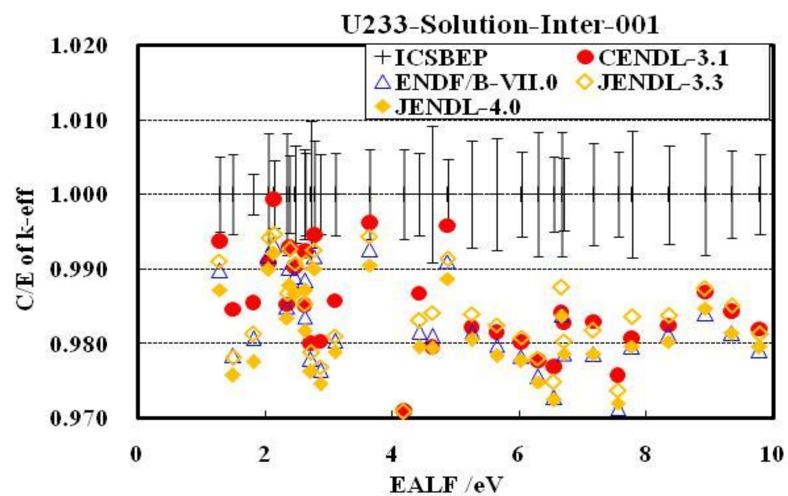


C/E values of k_{eff} for UMF system

- **Under prediction of k_{eff} for UMF system is caused by too low ^{233}U(n, inl) XS and too high (n,el) XS;**
- **The resolved resonance parameters are poor，serious under estimation of k_{eff} depend on the spectrum index was observed in results of all libraries.**



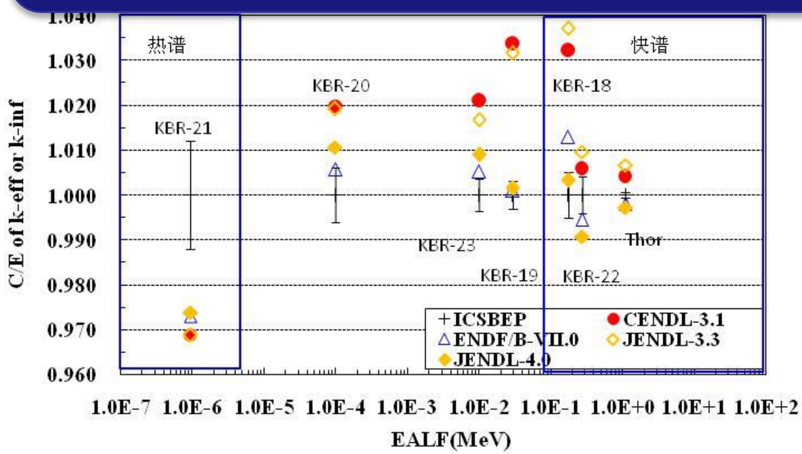
C/E values of k_{eff} for UST system



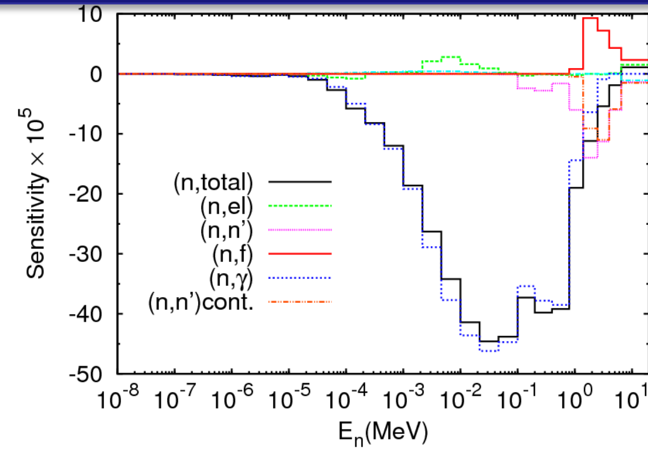
Comparison of C/E values for USI system

^{232}Th Cases

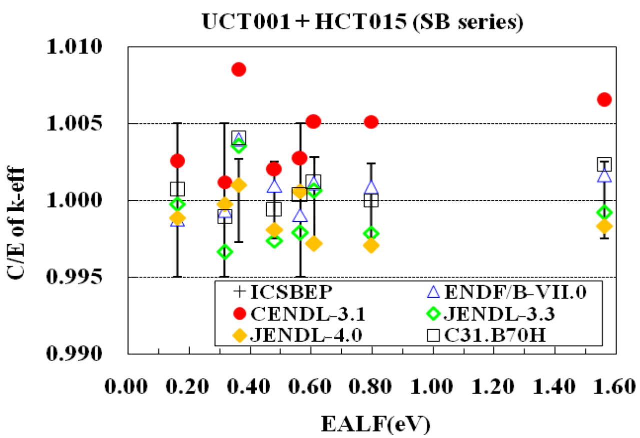
- ^{232}Th(n,γ)XS of CENDL-3.1 in 4keV～1MeV region is too low;
- Resolved resonance XS of ^{232}Th(n,γ) may not large enough.



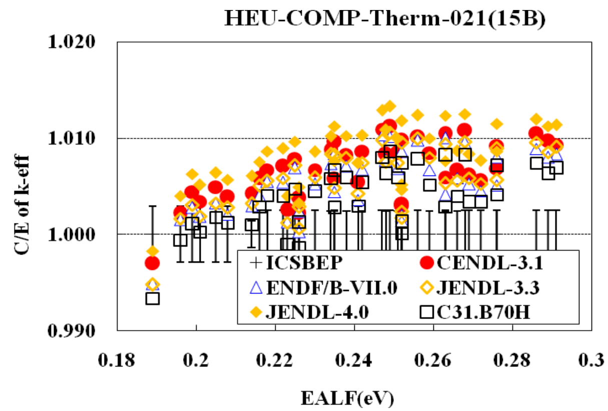
C/E values of k_{eff} for KBR and Thor



Sensitivities of k_{eff} to ^{232}Th XS (KBR-19)



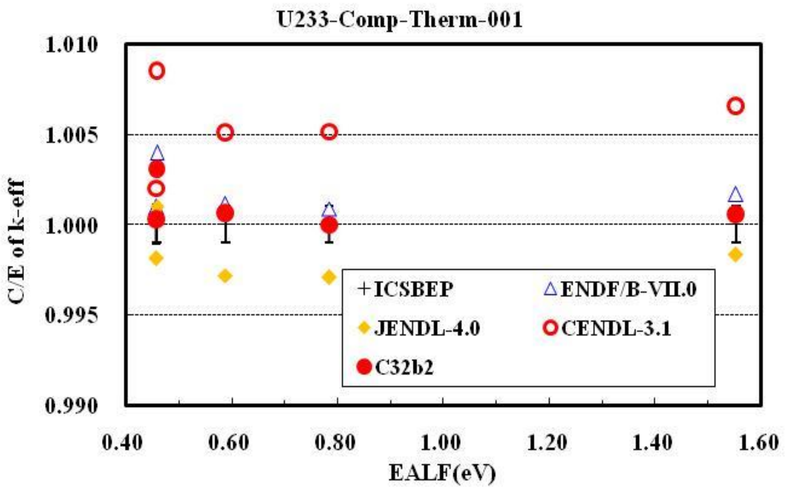
C/E values of k_{eff} for SB series



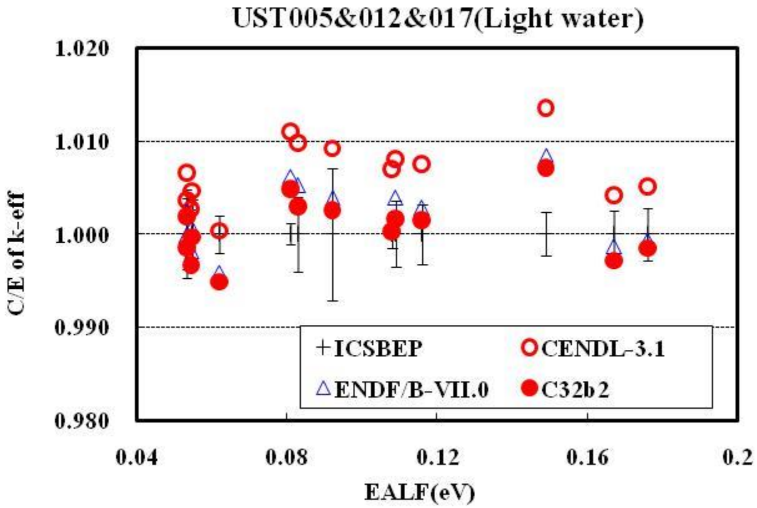
C/E values of k_{eff} for HCT021(15B)

5.2 Validation of the new CENDL evaluations

- **The new evaluations for ^{232}Th and ^{233}U accomplished in this work were tested with the same benchmark set.**
- **For C32b2, only ^{232}Th, ^{233}U and ^{1}H were replaced with new evaluations.**
- **Improvements are obtained.**



UCT system



Water reflected ^{233}U nitrate solution system

6. Brief Introduction of CENDL-TMSR

- **A hybrid library was recommended based on CENDL-3.1, -3.2beta、ENDF/B-VII.0、JENDL-4.0、JEFF-3.1.1 and IAEA/ADS-2.0**

	Materials	No.
Light nuclides (Z=<10)	^{1,2,3}H、^{3,4}He、^{6,7}Li、^{9}Be、^{10,11}B、^{12}C、^{14,15}N、^{16,17}O、^{19}F	16
Structure Material (11=<Z=<83)	^{22,23}Na、^{24,25,26}Mg、^{27}Al、^{28,29,30}Si、^{31}P、^{0,32,33,34,36}S、^{35,37}Cl、^{36,38,40}Ar、^{39,40,41}K、^{40,42,43,44,46,48}Ca、^{45}Sc、^{46,47,48,49,50}Ti、^{0}V、^{50,52,53,54}Cr、^{55}Mn、^{54,56,57,58}Fe、^{59}Co、^{58,60,61,62,64}Ni、^{0,63,65}Cu、^{0}Zn、^{175,176}Lu、^{174,176,177,178,179,180}Hf、^{181,182}Ta、^{180,182,183,184,186}W、^{185,187}Re、^{191,193}Ir、^{197}Au、^{196,198,199,200,201,202,204}Hg、^{204,206,207,208}Pb、^{209}Bi	86
Fission Products (31≤Z≤68)	^{69,71}Ga、^{70,72,73,74,76}Ge、^{74,75,77,79}As、^{74,76,77,78,79,80,82}Se、^{79,81}Br、^{78,80,82,83,84,85,86}Kr、^{85,86,87}Rb、^{84,86,87,88,89,90}Sr、^{89,90,91}Y、^{90,91,92,93,94,95,96}Zr、^{93,94,95}Nb、^{92,94,95,96,97,98,99,100}Mo、^{99}Tc、^{96,98,99,100,101,102,103,104,105,106}Ru、^{103,105}Rh、^{102,104,105,106,107,108,110}Pd、^{107,109,110m,111}Ag、^{106,108,110,111,112,113,114,115m,116}Cd、^{113,115}In、^{112,113,114,115,116,117,118,119,120,122,123,124,125,126}Sn、^{121,123,124,125,126}Sb、^{120,122,123,124,125,126,127m,128,129m,130,132}Te、^{127,129,130,131,135}I、^{123,124,126,128,129,130,131,132,133,134,135,136}Xe、^{133,134,135,136,137}Cs、^{130,132,133,134,135,136,137,138,140}Ba、^{138,139,140}La、^{136,138,139,140,141,142,143,144}Ce、^{141,142,143}Pr、^{142,143,144,145,146,147,148,150}Nd、^{147,148,148m,149,151}Pm、^{144,147,148,149,150,151,152,153,154}Sm、^{151,152,153,154,155,156,157}Eu、^{152,153,154,155,156,157,158,160}Gd、^{159,160}Tb、^{156,158,160,161,162,163,164}Dy、^{165,166m}Ho、^{162,164,166,167,168,170}Er	224
Actinides (Z≥84)	^{223,224,225,226}Ra、^{225,226,227}Ac、^{227,228,229,230,231,232,233,234}Th、^{230,231,232,233}Pa、^{232,233,234,235,236,237,238,239,240,241}U、^{235,236,237,238,239}Np、^{236,237,238,239,240,241,242,243,244,246}Pu、^{240,241,242,242m,243,244,244m}Am、^{240,241,242,243,244,245,246,247,248,249,250}Cm、^{249,250}Bk、^{249,250,251,252,253,254}Cf、^{253,254,255}Es、^{255}Fm	74
Total		400

7. Summary

- **To improve the nuclear data related to Th-U fuel cycle, some key data have been measured or re-evaluated:**
  - nFYs for 14MeV n + ^{232}Th were measured with uncertainties < 5% around two peak of the mass distribution.
  - Systematics formula of thermal (n,f) XS was obtained.
  - Nuclear decay data and structure data for A=232 and 234 were updated.
  - Fission product yields for n+ ^{232}Th were recommended based on combination of ENDF/B-VII.0 and JEFF-3.1.1.
  - Nuclear reaction data for ^{232}Th and ^{233}U in CENDL were revised and improved based on the feedbacks from benchmark testing.

- The benchmarking results show the resonance parameters for ^{233}U and ^{232}Th still need to be improved.
- A hybrid general purpose library, CENDL-TMSR, which contains 400 nuclides is recommended.

- **Research work need to be done**
  - Improve nFY for ^{232}Th and ^{233}U;
  - Establish reliable theory or systematics for thermal capture;
  - Improve neutron reaction data, especially resonance parameters for ^{233}U and ^{232}Th;
  - Too many others can not be fully listed here.
- **Are the current nuclear data libraries good enough to design a Th reactor, even get a license?**

Thank you for your attention !