Thorium Energy Conference THEC13
October 27 - 31, 2013, Globe of Science and Innovation, CERN, Geneva, Switzerland

Neutronic analysis and transmutation performance of Th-based Molten Salt Fuels

S. -I. Bak[1], J. S. Chai[1], J. P. Curbelo[2], S. W. Hong[1], E. J. In[1], Yacine Kadi[1,3], Claudio Tenreiro[1,4], Y. Zhou[1]

[1]Department of Energy Science, SungKyunKwan University, Suwon 440-746, Korea,
[2]Higher Institute of Technologies and Applied Sciences, InSTEC, Havana, Cuba,
[3]Engineering Department, European Organization for Nuclear Research, CERN, CH-1211 Geneva 23, Switzerland,
[4]Faculty of Engineering, University of Talca, St 720-747, Talca, Chile

Introduction

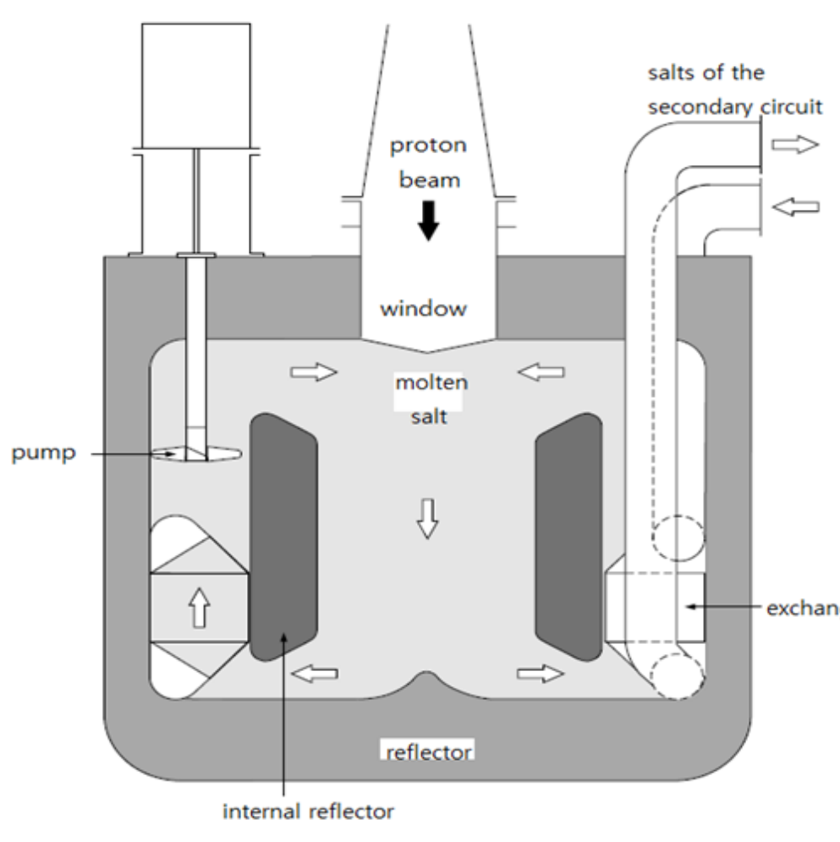
- Molten Salt Reactor (MSR)
  - MSR operates at near atmospheric pressure.
  - It runs at temperature higher than water cooled reactors.
  - It is online processing.
  - Molten Salt acts both as the primary coolant and as the fuel itself.

- We consider subcritical MSR with Accelerator Driven System
- Studies of Li-Be and Na based fuels have done in the past but little attention has been given to the Pb based fuel.
- Thorium fuel and thorium fuel cycles are attractive for the long-term nuclear energy production with low radioactive waste.

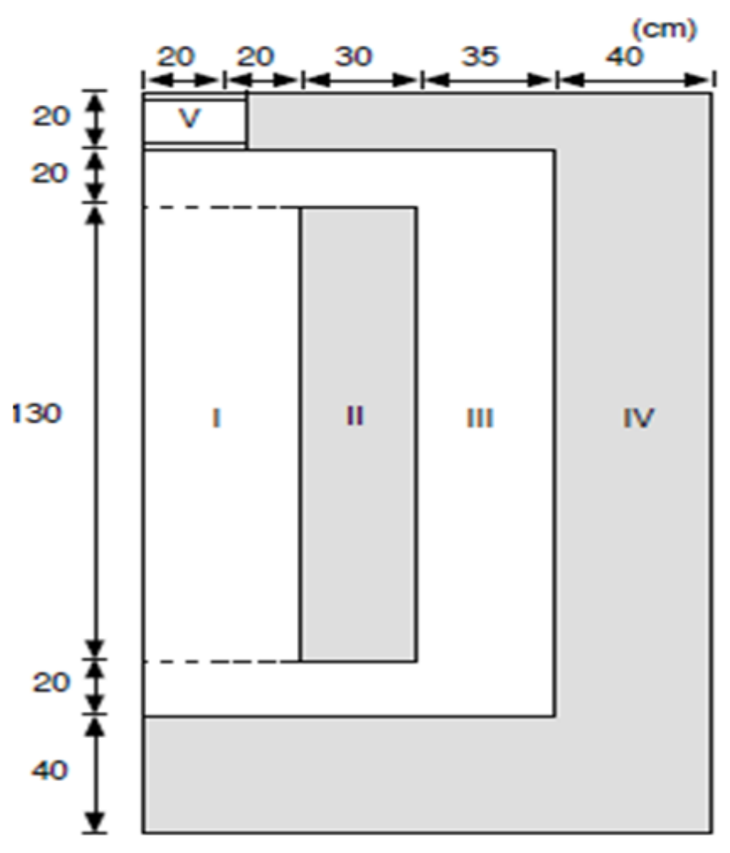
Main motivation is to estimate

- neutron multiplication factor (K_{eff}),
- breeding or conversion ($^{232}Th \rightarrow ^{233}U$) potential,
- elimination potential of Minor Actinides,
- safety characteristic (temperature coefficient)
  - for Li-Be Fluoride, Na Chloride and Pb Chloride
  - with or w/o Th and ^{233}U and additionally Minor Actinides and Pu

Geometry of Accelerator Driven Molten Salt Target System



JAERI's molten salt ADS concept
Y.Kate, et al. JAERI,
Meeting on Accelerator Based Transmutation, PSI, 1992



Simplified geometry for the simulation.
Zone I : Primary salt
Zone II : Hastelloy-N
Zone III : Primary Salt / Structural materials / Secondary Salt (5:2:4 in volume)
Zone IV : Hastelloy-N
Zone V : Helium

Fuels considered in this work

- Li-Be Fluoride
- Na Chloride
- Pb (II) Chloride
- These fuels are based on the works by ORNL, JAERI, etc.

Fuel	Composition
Li-Be Fuel 1	64% ^{7}LiF, 18% BeF_2, 18% (Th,^{233}U, Pu, MA) Fluoride
Li-Be Fuel 2	73% ^{7}LiF, 15% BeF_2, 12% (Th,^{233}U, Pu, MA) Fluoride
Na- Fuel	64% NaCl, 36% (Th,^{233}U, Pu, MA) Chloride
Pb- Fuel	64% $PbCl_2$, 36% (Th,^{233}U, Pu, MA) Chloride

Monte Carlo Simulation

We used two calculation methods:

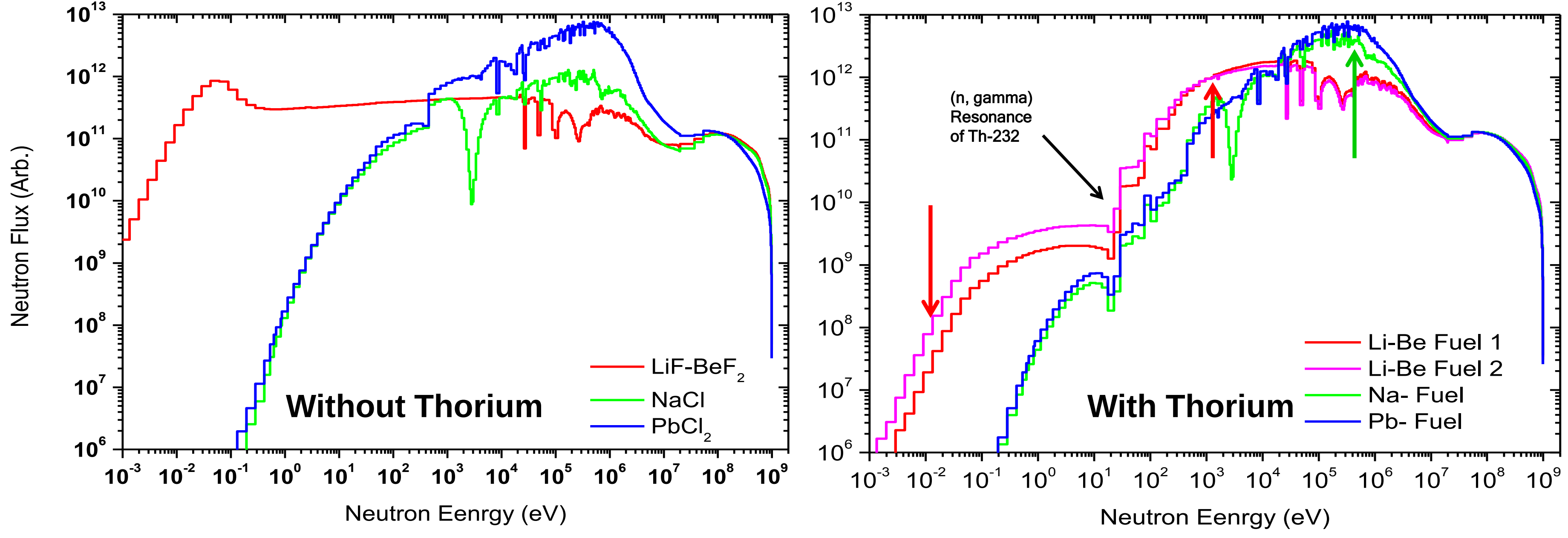
- Energy Amplifier Monte Carlo (EA-MC): assuming D+D fusion neutron source
- FLUKA: Correct simulation of spallation process induced by 1 GeV protons impinging directly on the molten salt fuel

	EA-MC	FLUKA
Neutron Source	• Neutron from D+D fusion (E_n ~ 2.4 MeV) • Power 10 MW-thermal	• Neutrons generated by 1 GeV, 1mA proton beam
Calculated Quantities	• K_{eff} • Neutron flux • Contribution from different reaction channels	• Distribution of particles (neutron, photon, charged particles) in space and in energy • Neutronics • Number of secondary neutrons per incident proton
Nuclear data	• JAR-95 @ 800, 900, 973K point data analysis	• ENDF/B-VII @ 296K 260 group data analysis

Results

1. Comparison of neutron spectra in molten salts with and without Thorium

Assuming 1 GeV 1mA proton beam impinges directly on the fuel



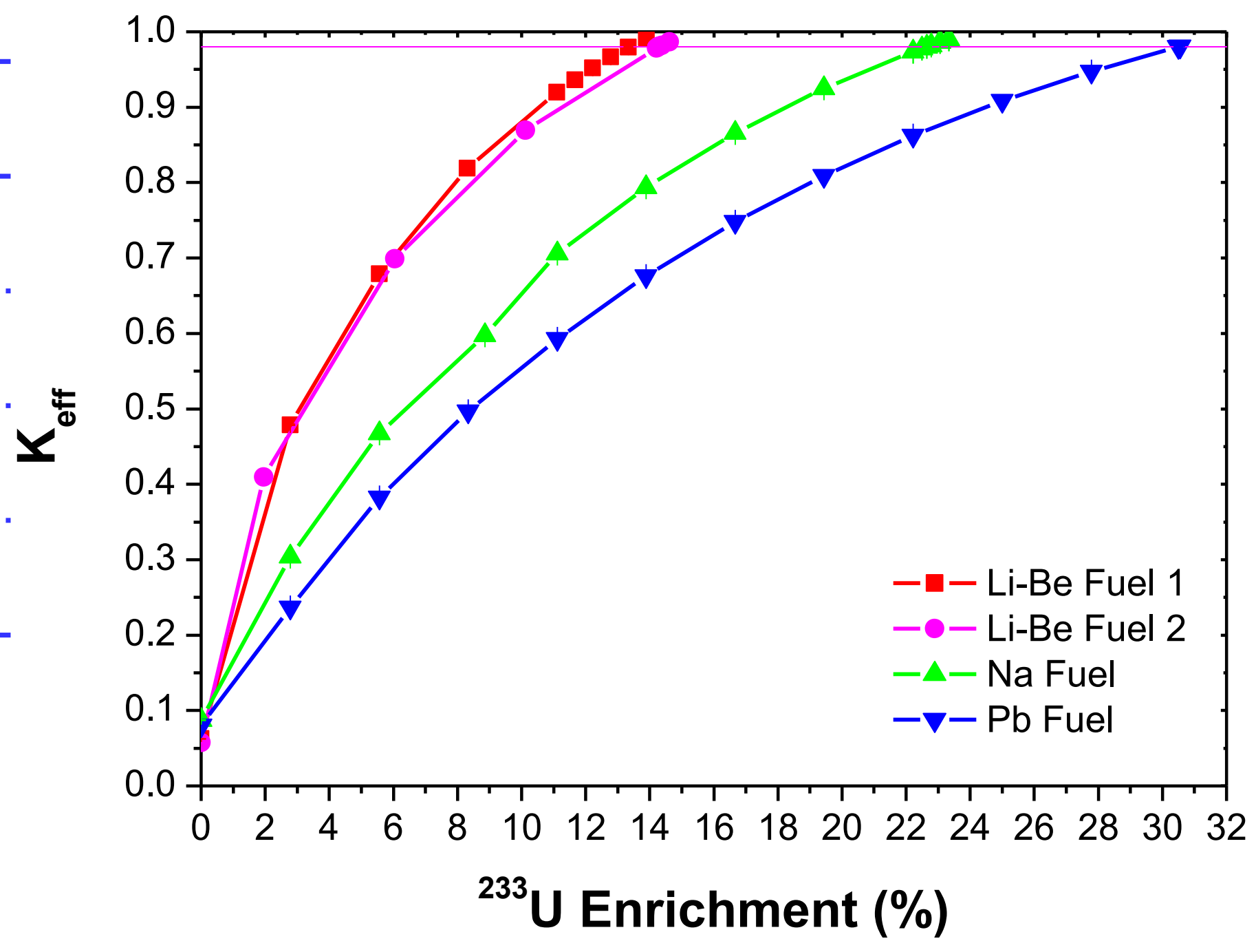
- $LiF+BeF_2$ can produce thermal neutron spectrum
- Neutron spectrum of $PbCl_2$ is similar to that of NaCl, because neutron absorption on ^{35}Cl isotopes is dominant in thermal energy region
- Fuels of different atomic mass have different moderating power
- Thorium makes neutron spectrum harder and captures neutrons in the region from thermal to resonance energies

2. K_{eff} and Breeding Ratios for Thorium-^{233}Uranium Fuels

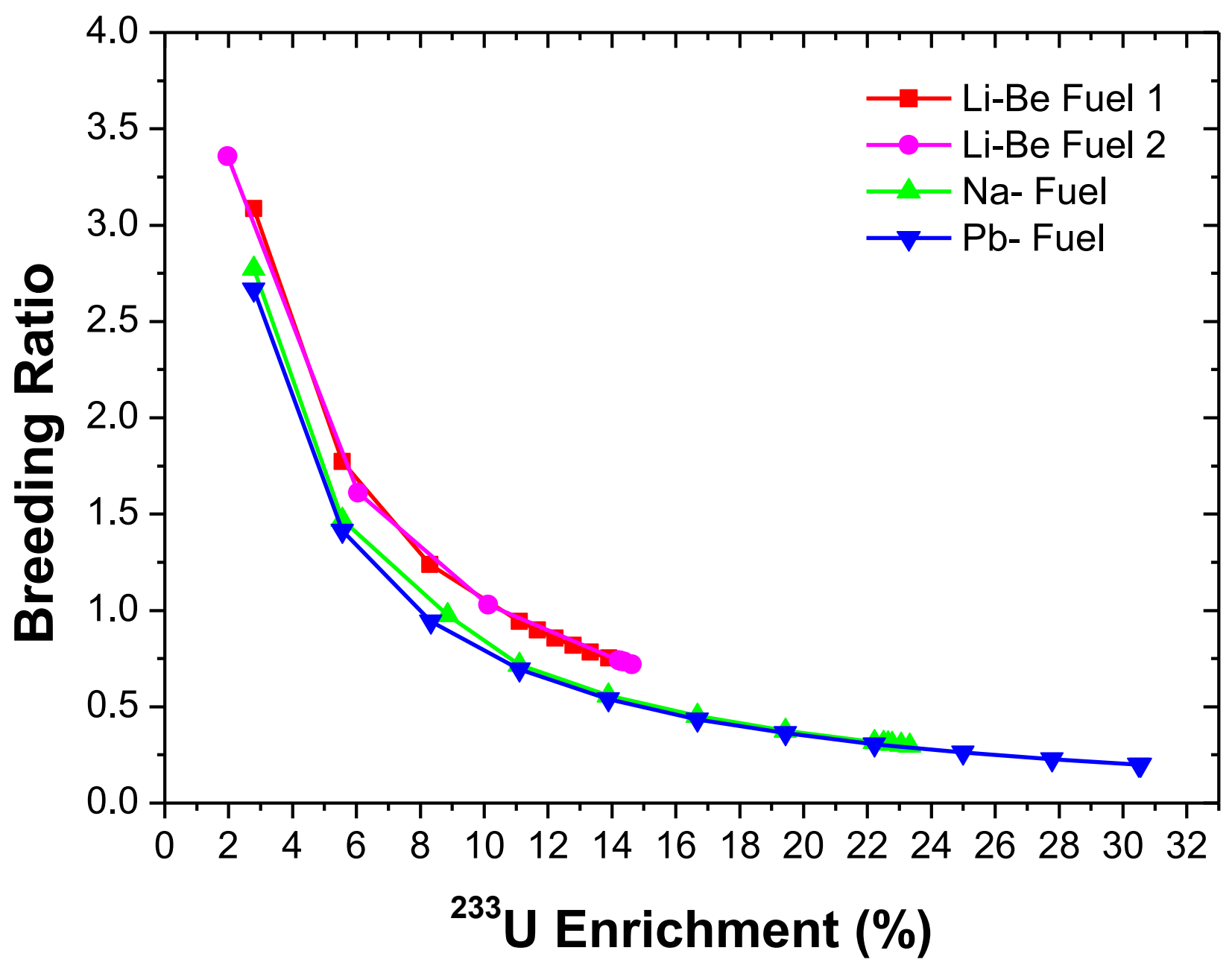
Calculation of K_{eff} with different Uranium enrichment

Fuel (% in moles, at K_{eff} ~0.98)	Enrichment* (U-Th %)	Neutron leakage (%)
$^{7}LiF + BeF_2 + ^{232}ThF_4 + ^{233}UF_4$ 64-18-15.6-2.4	13.3	0.54
$^{7}LiF + BeF_2 + ^{232}ThF_4 + ^{233}UF_4$ 72.76-15-10.493-1.747	14.3	0.53
$NaCl + ^{232}ThCl_3 + ^{233}UCl_3$ 64-27.85-8.15	22.6	2.14
$PbCl_2 + ^{232}ThCl_3 + ^{233}UCl_3$ 64-25.02-10.98	30.5	2.57

$$* \text{ Enrichment} = \frac{^{233}U\ (mole\ \%)}{^{233}U(mole\ \%) + ^{232}Th\ (mole\ \%)} \times 100\ \%$$



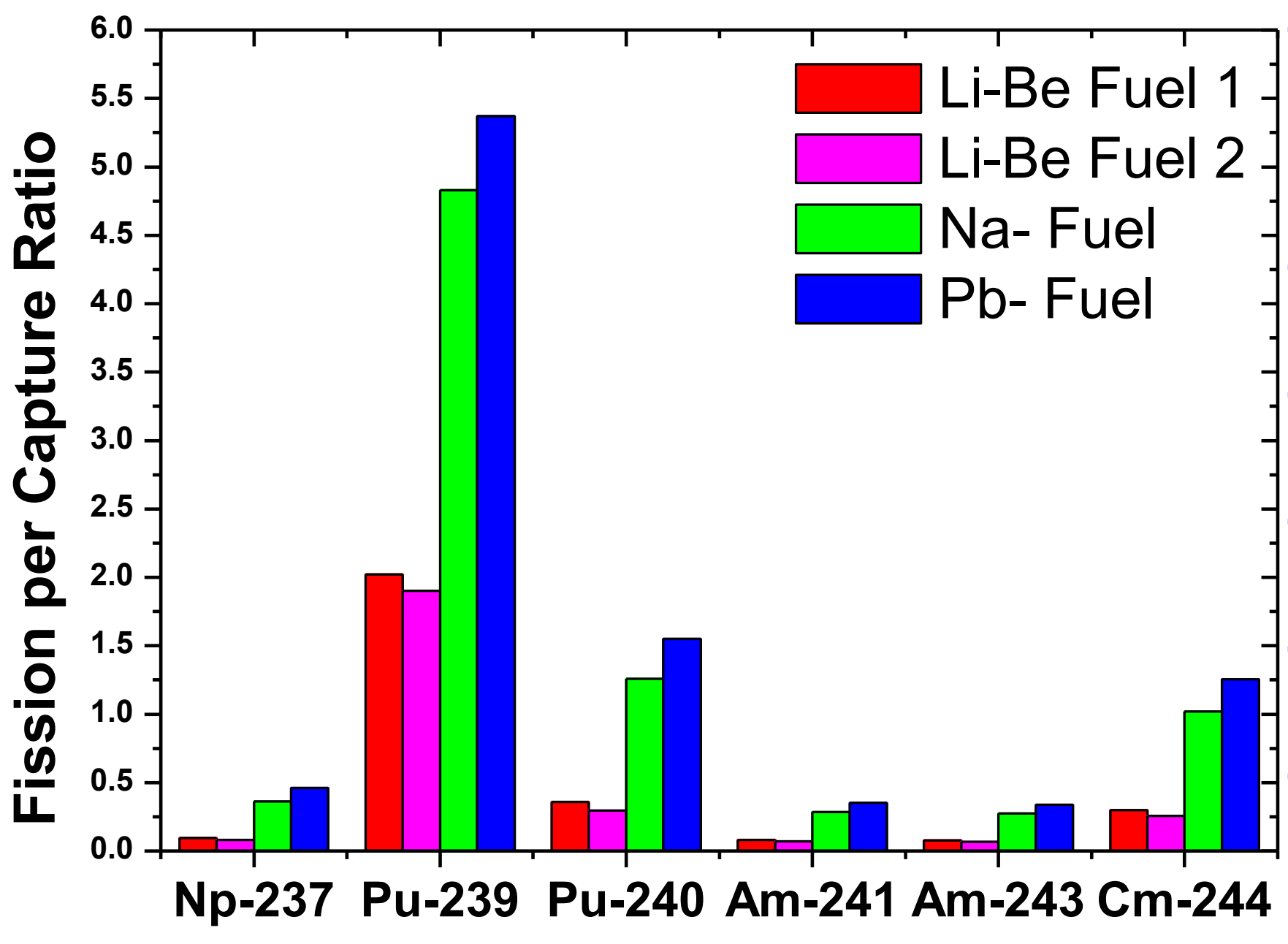
Breeding (Conversion) Ratio



$$\text{Breeding Ratio} = \frac{neutron\ capture\ on\ ^{232}Th}{Amount\ of\ spending\ fuel}$$

- $LiF+BeF_2$ based salts require much less enrichment than NaCl or $PbCl_2$ salts because of neutron spectra (thermal/epithermal with $LiF+BeF_2$ vs fast with NaCl or $PbCl_2$).
- This is also reflected in the rate of neutron leakage: More leakage of fast neutrons.
- Better breeding ratio is achieved for thermal/epithermal ($LiF+BeF_2$) systems

3. Thorium + Plutonium + Minor Actinides @ K_{eff} ~ 0.98



Fuel (% in moles)	Breeding (conversion) Ratio	Estimated Transmutation of MA* (%/MW/year)
$^{7}LiF + BeF_2 + ThF_4 + (Pu+MA)F_3$ 64% + 18% + 12.6% + 5.4%	0.41	0.0032
$^{7}LiF + BeF_2 + ThF_4 + (Pu+MA)F_3$ 72.76% + 15% + 8.06% +4.18%	0.35	0.0031
$NaCl + ThCl_3 + (Pu+MA)Cl_3$ 64% + 24.49% + 11.51%	0.29	0.0055
$PbCl_2 + ThCl_3 + (Pu+MA)Cl_3$ 64% + 21.37% + 14.63%	0.19	0.0068

- MA : $^{237}Np + ^{241}Am + ^{243}Am + ^{244}Cm$ (12.5 : 50 : 25 : 12.5)
- $^{239}Pu : ^{240}Pu = 80:20$
- Pu : MA = 10:1

- Above several hundred keV, fission cross section of Minor Actinides is higher than capture cross section
- Fast neutron system is preferred for the transmutation of Minor Actinides.

4. Temperature Coefficients for different fuels

	Temperature Coefficient (pcm/K)	
	$^{232}Th+^{233}U$	$^{232}Th+Pu+MA$
Li-Be Fuel 1	-2.78	-1.15
Li-Be Fuel 2	-3.09	-1.1
Na- Fuel	-0.3	-0.26
Pb- Fuel	-0.31	-0.11

$$\alpha_t = \frac{d\rho}{dT} = \frac{1}{k^2}\frac{dk}{dT}, \text{ where } \rho = \frac{k-1}{k}$$

- Temperature coefficient is one of the most important parameters of the nuclear reactor safety.
- All the values of the different fuels are negative and the Li-Be Fuels have bigger feedback values than Na- and Pb- fuels.

Conclusion

- AD-MSR is interesting: Molten Salt acts as fuel, coolant and target.
- Thermal/epithermal Li-Be fluoride molten salt is better than Na- or Pb (II) chloride for a pure Th-U fuel.
- Chloride fuels have disadvantage due to higher (n,γ) cross section from thermal to resonance energies.
- Fast neutron system is preferred for the transmutation of Minor Actinides.

Relative comparison of MSFs

- For less ^{233}U enrichment	$LiF+BeF_2$ > NaCl > $PbCl_2$
- For better Breeding (Conversion) Ratio	$LiF+BeF_2$ > NaCl ~ $PbCl_2$
- For more Spallation neutrons	$PbCl_2$ > NaCl > $LiF+BeF_2$
- For more Transmutation of Minor Actinides	$PbCl_2$ > NaCl > $LiF+BeF_2$
- For safer Temperature Coefficient	$LiF+BeF_2$ > NaCl ~ $PbCl_2$