Surrogate reactions and their applications

Larry Phair Lawrence Berkeley National Laboratory

The Surrogate Method



Outline:

- I. "New" surrogate methods: ratios
- II. Measurement details
- III. Examples: new cross sections, ²³⁷U(n,f)
- IV. Benchmarking
 - (a,a'f) (d,pf)
 - (³He,af) (³He,tf)
- V. Theory (or lack thereof)
- VI. Applications

- Surrogate reaction collaborators
- L.A. Bernstein, <u>D.L. Bleuel</u>, J.T.
 Burke, L. Ahle, J. Church, J.
 Escher, F.S. Dietrich, <u>S.L. Lesher</u>,
 K. Moody, J. Punyon, <u>N. Scielzo</u>,
 M. Wiedeking



L. Phair, M.S. Basunia, P. Fallon, R.M. Clark, M. Cromaz, M.A. Delaplanque-Stephens, I.Y. Lee, A.O. Macchiavelli, M.A. McMahan, E. Rodriguez-Vieitez, <u>S. Sinha,</u> F.S. Stephens, <u>J.D. Gibelin</u>



- <u>B.F. Lyles</u>, L.G. Moretto, E.B. Norman, P. Lake
- Yale H. Ai, <u>C. Plettner,</u>



C.W. Beausang, B. Crider, <u>J.M.</u>
 <u>Allmond</u>, B. Darakchieva, M.
 Evtimova

PNNL – J. Cagianno, J. Ressler ₃ Rutgers – <u>R. Hatarik</u>, J. Cizewski,

The Surrogate Method



Surrogate Reaction "Flavors"

J.D. Cramer, H.C. Britt, Nucl. Sci. Eng. **41**, 177 (1970) H.C. Britt, J.B. Wilhelmy, *ibid.* **72**, 222 (1979)



Surrogate Reaction "scorecard"



Example: ²³⁷U(*n*,*f*) via surrogate external <u>ratio</u>

Spirit of the method

$$\sigma_{n,f}(238) = \sigma_n^0(238)P_f(238)$$









292 mg/cm² target with no backing



Example of light particle, fission and g Spectra



The surrogate ratio method has been applied to the ²³⁷U(n,x) cross sections



Uranium Reaction Network

- Understanding the destruction of ²³⁷U is important for a number of applications
- Activity associated with a 10 mg target: 810 Curies!
- An earlier attempt to measure ²³⁷U(*n*,*f*) used an "unconventional" neutron source (Bomb)



Example: ²³⁷U(*n*,*f*) via surrogate external <u>ratio</u>

Spirit of the method

$$\sigma_{n,f}(238) = \sigma_n^0(238)P_f(238)$$





Burke et al, Phys. Rev. C 73, 054604 (2006)

Testing the surrogate method(s)

- $(\mathbf{M}, \mathbf{M}')$ for (n, f), external ratio
- (*d*,*pf*) for (*n*,*f*)
- (³He, *f*) for (*n*,*f*)
- (³He,*tf*) for (*n*,*f*)
- •

Benchmarking the external ratio method - $^{234}U(\mathbb{W},\mathbb{W}'f)/^{236}U(\mathbb{W},\mathbb{W}'f)$ vs. $^{233}U(n,f)/^{235}U(n,f)$



Ratios work even when we are not in the Weisskopf-Ewing limit

Recent LANL work allows us to compare our ²³⁸U(³He,*tf*)²³⁸Np reaction to a direct ²³⁷Np(n,f) measurement

PHYSICAL REVIEW C 75, 034610 (2007) 2.8 This work 237 Np(n, f Neutron induced fission cross section of ²³⁷Np from 100 keV to 200 MeV Cross section (barn) Ref. 23 2.6 ENDF/B-MI.0 F. Tovesson^{*} and T. S. Hill JENDL 3.3 2.4 Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA (Received 16 November 2006; published 20 March 2007) 22 2.0 1.8 First test of the absolute 1.6 This work (237Np) / ENDF/B-VILO (235U) Ref. 24 surrogate method at high 237 Np(n,f) / 235 U(n,f) 1.4 ENDF/B-ML0 E_x Ratio 1.0 0.8 10 12 17 19 11 13 14 15 16 18 Neutron Energy (MeV) 111111 Basunia et al. NIM B 267, 1899 (2009) BERKELEY

20



Internal Ratio Result for ²³⁵U(n,g)



ENDF: solid

STARS/LIBERACE: open ²³⁵U(d,p)

J. M Allmond et al. Phys. Rev. C 79, 054610 (2009)

Examining the effects J^π differences (0⁺ vs. 7/2⁻): ²³⁶U(n,f) from ²³⁸U(³He, ⊠'f)/²³⁵U(³He, ⊠'f) (external ratio and absolute)



Ph.D. thesis project Bethany Lyles

Failed because of contaminants in the target

Absolute





FIG. 12. The 236 U(n, f) cross section determined using the SRM relative to the 233 U(n, f) cross section as a function of equivalent neutron energy. The solid line is the ENDF/B-VII library evaluation for this cross section.

The External Ratio approach works for (n,f) for suitable spin distributions

J. Escher & F.S. Dietrich, PRC 74 054601 (2006)





Ratios are relatively insensitive to spin differences between the actual and surrogate reactions

Collaboration publications

Experiment	Principle Investigator	Publication
²³⁵ U(d,pg) and (d,pf) for (n,g) and (n,f)	J. M. Allmond	Phys. Rev. C 79 , 054610 (2009)
^{234,236} U(a,a' f) for (n,f)	S. R. Lesher	Phys. Rev. C 79, 044609 (2009)
²³⁸ U(³ He,tf) for (n,f)	M. S. Basunia	Nucl. Inst. & Meth. B 267 , 1899 (2009)
^{235,238} U(³ He,af) for (n,f)	B. F. Lyles	Phys. Rev. C. 76, 014606 (2007)
²³⁸ U(a,a' f) for ²³⁷ U(n,f)	J. T. Burke	Phys. Rev. C 73, 054604 (2006)
²³² Th(³ He, ³ He'f) and ²³² Th(³ He,af) for ^{230,231} Th(n,f)	B. L. Goldblum	To be submitted to Phys. Rev. C
^{171,173} Yb(d,pg) for (n,g)	R. Hatarik	To be submitted to Phys. Rev. C

Applications

- Stockpile stewardship
- Reactor designs
- Nuclear astrophysics

s-process

Example: Th fuel cycle

M. Petit et al. / Nuclear Physics A 735 (2004) 345-371

Neutron energy (MeV)



²³⁰Th(n, f) Cross Section Using the SRM



FIG. 2: The ²³⁰Th(n, f) cross section in the equivalent neutron energy range of 0 to 25 MeV. The error bars on the surrogate data represent both the statistical and systematic uncertainty. For comparison, evaluated and directly-measured ²³⁰Th(n, f) cross section data are shown.

231 Th(n, f) Cross Section Using the SRM

²³²Th(³He,³He'f) 1 0.9 231Th(n,f) Cross Section (barns) 0.8 ģ 0.7 0.6 0.5 0.4 0.3 0.2 0.1 0 2 8 0 4 6 10 Equivalent Neutron Energy (MeV)

FIG. 4: The ²³¹Th(n, f) cross section extracted using the SRM, relative to the evaluated ²³⁵U(n, f) cross section obtained from ENDF/B-VII, as a function of equivalent neutron energy is given by the open squares. The error bars represent both the statistical and systematic uncertainty. For comparison, the evaluated ²³¹Th(n, f) cross section from ROSFOND is denoted by the solid line.

²³⁹Np(n,f) and ²³⁹Pu breeding in reactors

	E 3.3	236.0514	E 2.7	238.05302	E 1.8	240.055519	E .767	242.058829	243.061382
	Am234 2.3 m ε	Am235 15 m ε?	Αm236 4 m	Am237 ^{5/ (−)} 1.22 h	Am238 1+ 1.63 h	Am239 (5/)- 11.9 h ε	Am240 ⁽³⁻⁾ 2.12 d	Am241 5/- 432.7 a α 5.4857, 5.4430,	5- Am242 **- 141 a 16.52 **
95	α 6.46 ?	500	500	γ 280.2,… α 6.04 ω	β ⁺ , ω γ 962.8, 918.7, 561.0,… α 5.94 νω	$\begin{array}{l} \gamma \; 277.6, \; 228.2, \cdots \\ \alpha \; 5.774 \; (\omega), \; 5.734, \cdots \\ \gamma \; 49.3 \; \omega \end{array}$	γ 987.7, 888.8,… α 5.378 νω,…	γ 59.5, 26.3-955 SF νω σγ (5E1+55E1), (19E1+13E2) σf 3.2, 14	0.0.0.0000 y 49.20 SF vo a, -17E2. 262 262 E-meter
	E 4.2	E 2.6	E 3.3	E 1.5	E 2.3	E .803	E 1.38	241.056823 🏷	18E2 E-78
	20.9 m	8.8 h	25.3 m	2.87 a	Pu237 7/- 45.2 d	Pu238	Pu239 1/+	Pu240	Pu241 ===
94	E 1/ 235 3 534 7	ε α 6.200, 6.149,…	ε ν 49.2. 756.4	α 5.7675, 5.7209,	ε, γ 59.5,··· α 5.344(ω),···	α 5.4992, 5.4565, γ 43.5 ω (e),	α 5.156, 5.144, 5.105,… γ 51.6 e=, 30.1=1057.3ω	α 5.1683, 5.1237,… γ 45.2ω (e),	β- 0208 α 4.897 α. 4.852
0-	α 6.30 ω		α 5.85 ω	SF ννω σε 16E1_1E3	γ 280.4(νω), 289.9, 320.8,…	99.9 (e), SF νω	SF ννω σγ 271, 20E1	104.2(e), SF νω	7 145.5 mml 700.00 0,-361, 1650
	E 2.1	E .39	E 1.14	236.046048 🛇	े ^न 24E2 E .22	of 18, ~33 238.049553 ∅	$\sigma_{\alpha} < 0.4 \text{ mb}$ 239.052157 \bigcirc	of 0.05, 2.4 240.053807	au < 02 me = 101mi
	Np232 ⁽⁴⁺⁾ 14.7 m	Np233 ^(5/+) 36.2 m	Np234 ⁽⁰⁺⁾ 4.4 d	Np235 5/+ 1.085 a	^{1 (-)} Np236 ⁽⁶⁻⁾ 22.5 h 1.55E5 a	Np237 5/+ 2.14E6 a	Np238 2+ 2.117 d	Np239 5/+ 2.355 d	1 (+) Np240 === 7.22 m 1.552 m
93	ε γ 326.8, 819.2, 866.8, 864.3,…	ε γ 312.0, 298.9, 546.5,…	ε β+.79 ω γ 1558.7, 1527.2,	ε α 5.021 (νω), 5.004,…	$\mathcal{E}, \beta^ \mathcal{E}, \gamma 160.3, \cdots$ 54, $\beta^-, \gamma 44.6$ $\gamma 642.3, e^-, \cdots$	α 4.788, 4.771,… γ 29.4, 86.5,… σ _γ 15Ε1, 65Ε1	β ⁻ .263, 1.248,… γ 984.5, 1028.5,…	β ⁻ .438, .341, γ 106.1, 277.6, 228.2,	β-2.18, β-35 1.60,
	F 0.0	α 5.53 ? ω	δ _f 9E2	γ 25.6—188.8 νω σ _γ (15E1+?)	of 2.7E3, E+ 0.9	σf 0.02, 7	σf 21E2, 9E2	∂ _γ (3E1+3E1)	597.4, HILLIH
		E 1.0	E 1.81	E .124	7E2 E- 0.5	237.048167 🔿	E 1.292	E .722	H EZZE
	4.2 d	69.8 a	1.592E5 a		26 m AcU	U236	0237 1/+	U238	U239 5**
92	ε γ 25.65, 84.2,…	α 5.3203, 5.2635, γ 57.8 ω (e),	α 4.824, 4.783,… γ 42.5 ω, 97.1, 54.7,…	2.46E5 a α 4.776, 4.725,···	IT ~76.8 eV (ω), e 7.04E8 a	α 4.494, 4.445,…	β24, .25,···	4.47E9 a	β- 1.21, 1.28,
	α 5.456 ω , γ 68.33, 53.23,	129.1,··· SF <i>ω</i>	SF νω σ _γ 46, 14E1	γ 53.2 (e), 120.9,··· SF νω	α 4.398, 4.366,… γ 185.72, 143.76,…	γ 49.4 ω (e), 112.8, SF νω	σ _γ 4E2, 12E2	γ 49.6 ω (e), SF νω	Gy 22
	∂f 3E2	σ _γ 73, 28E1 σ f 75, 38E1	$\sigma_{\rm f}$ 531, 76E1 $\overline{\sigma}_{\alpha}$ < 0.3 mb	σ_{γ} 100, 7E2 $\sigma_{\rm f} < 5$ mb, 7	SF νω σ _γ 99, 14E1 σ _f 585, ~275	σ _γ 5.1, 36E1 σ _f 0.04, 4	σf < 0.35	α _γ 2.68, 277 σf - 5μb 1.3mb	^o f 15
	E .382	232.03/146 O	P 233.039628		$\sigma_{\alpha} < .1 \text{ mb } 235.043923$	$236.045562 \bigcirc$	E .519	D ₂ 238.050783	E 1.260
	17.4 d	Pazor	1.31 d	26.967 d		24.4 m	9.1 m	8.7 m	2.3 m
01	ε, γ 952.0,… β=.51	100 3.28E4 a	β^{-} .31, .29, y 969 3 894 3	$\beta^{-}.256, .15, \cdots$	1.17 m 6.69 h β^- 2.29, β^- .48, .65,	β^{-} 1.41, v 30 1 - 658 9	β^{-} 2.0, 3.1,	β ⁻ 1.2, 1.5, 2.25	β-1.7, 1.2, 22-
31	$\gamma 314.8 \omega, \cdots$ $\alpha 4.766 = 5.345 v\omega$	α 5.013, 4.950, 5.029,… γ 27.4, 300.1,… α. 20E1 5E2	8 ? 8 552 352	σ_{γ} (21+20), (46E1+44E1)	$\gamma 1001.0, \dots \gamma 10$	/ 0001 00000	1762.7,	γ 853.6, 865.0,	448, 681,-
	ôf 15E2 E +1.3010 E564	of 0.020, 0.05 231.035879 ♥	of 7E2 E 1.34	$\hat{\sigma}_{f < 0.1} E.570$	IT<10 ^γ 73.9 ω, = E 2.195	E 1.4	SF νω E 2.9	529.3, 540.7, E 2.3	E35
		140		1/12	<u> </u>	111		146	
		ITU		142		1		140	

Next proposed experiment at 88" Cyclotron



The surrogate ratio method can also be applied to other areas: Generation-IV reactor design

Target	Surrogate Reactions	Ratio Reactions	Reactor Type*
²³⁸ Pu	²³⁹ Pu(₩,₩')	²³⁵ U(₩,₩')	LFR,SFR
²³⁹ Pu	²³⁹ Pu(d,p), ²⁴⁰ Pu(⊮,⊮')	²³⁵ U(d,p), ²³⁶ U(₩,₩')	GFR,LFR,SFR,EFR
²⁴⁰ Pu	²⁴⁰ Pu(d,p), ²⁴² Pu(³ He,⊠)	²³⁶ U(d,p), ^{236,238} U(³ He,))	GFR,LFR,SFR,EFR
²⁴¹ Pu	²⁴² Pu(₩,₩')	²³⁶ U(₩,₩')	GFR,LFR,SFR,EFR
²⁴¹ Am	²⁴³ Am(³ He,⊠), ²⁴⁰ Pu(³ He,d)	²³⁵ U(³ He, ເເ), <i>none yet</i>	GFR,LFR,SFR
^{242m} Am	²⁴² Pu(³ He,t), ²⁴³ Am(⊮,⊮')	²³⁸ U(³ He,t), ²³⁵ U(₩,₩')	LFR,SFR
²⁴³ Am	²⁴³ Am(d,p), ²⁴² Pu(³ He,d)	²³⁹ Pu(d,p), <i>none yet</i>	SFR
²⁴² Cm	²⁴³ Am(³ He,t)	²³⁸ U(³ He,t)	EFR
²⁴³ Cm	²⁴⁵ Cm(³ He,⊠), ²⁴³ Am(³ He,t)	²³⁵ U(³ He, ₩), ²³⁸ U(³ He, <i>t</i>)	GFR,EFR
²⁴⁴ Cm	²⁴⁵ Cm(₩,₩'), ²⁴³ Am(³ He,d)	²³⁵ U(₩,₩'), none yet	GFR,LFR,SFR,EFR
²⁴⁵ Cm	²⁴⁷ Cm(³ He,𝔄), ²⁴⁵ Cm(d,p)	²³⁵ U(³ He, M), ²³⁵ U(d Richmon	F R

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*from Aliberti et al.,

Surrogates for nuclear astrophysics: the s-process (slow neutron capture)

<u>s-process</u>: slow neutron capture moves along valley of stability with branch points where
 decay competes with capture.



Figure 1 The s-process pathway from ¹⁶⁹Tm to ¹⁷³Yb from [2]. Branch-point nuclei are outlined in red.

HEDP-induced population of low-lying states in sprocess branch point nuclei could affect nucleosynthesis



Equilibrium population of the excited state is $\approx 36\%$

s-process, surrogate measurement "issues"

- Difficult targets
- Low energy neutrons, high intensity
- Dense plasma environment
 - Screening
 - Population of low lying excited states
 - Affects 🕅 decay lifetimes
 - Affects (n,) cross sections (Stellar enhancement factors)

Stellar enhancement factors (SEF)



- No data exists for (n, i) cross sections on two different states of the same nucleus at stellar sprocess energies (tens keV)
- Surrogate reactions will allow us to address the J mismatch issue

Important s-process branch points

Branch		1 st Exc. State	$1^{\underline{st}}$ Exc.	STARS/LIBERACE
Point	Ground State J^{π}	E_x (keV)	State J^{π}	reaction
⁷⁹ Se	7/2+	95.77	1/2-	⁷⁶ Ge(⁶ Li,d) ⁸⁰ Se [*]
⁸⁵ Kr	9/2+	304.871	1/2-	87 Rb(d, {}^{3}He) 86 Kr*
¹⁴⁷ Pm	7/2+	91.1	5/2+	149 Sm(d, 3 He) 148 Pm *
¹⁵¹ Sm	5/2-	4.821	3/2-	152 Sm(α , α ') 152 Sm [*]
¹⁶³ Ho	7/2-	100.03	9/2-	165 Ho(³ He, α) ¹⁶⁴ Ho [*]
¹⁷⁰ Tm	1.	38.7139	2-	171 Yb(d, ³ He) ¹⁷¹ Tm [*]
¹⁷¹ Tm	1/2+	5.0361	3/2+	172 Yb(d, ³ He) ¹⁷² Tm [*]
¹⁷⁹ Ta	7/2+	30.7	9/2+	181 Ta(3 He, α) 180 Ta *
²⁰⁴ T1	2-	414.1	4-	$^{205}\text{Tl}(\alpha,\alpha)^{205}\text{Tl}^*$
²⁰⁵ Pb	5/2-	703.3	7/2-	207 Pb(3 He, α) 206 Pb
¹⁸⁵ W	3/2-	23.547	1/2-	186 W(α, α') 186 W

Table I: Important s-process branch point nuclei from [1]. Potential STARS/LIBERACE reactions are in the right-most column.

Surrogate measurements will only take you so far.

Some speculative ideas

- Regarding the possible complementary measurements for the surrogate method:
 - Average cross section
 measurements at NIF

The National Ignition Facility (NIF): A new kind of nuclear laboratory



NIF is designed to implode D-T (or other) pellets to achieve thermonuclear fusion Standard ignition configuration: 192 beams, 1.8MJ in 3 in 1 ight



Up to 300 shots/year with $\approx 15\%$ dedicated for basic science (Ride-along also possible)

What do the NIF neutron spectra look like (Modeling courtesy of C. Cerjan)



Stellar reaction cross section measurements are enhanced by $(M_{areal} M^2 \text{ at NIF compared to accelerator-based experiments})$

Assumptions

- 1 mm diameter initial pellet size with density≈0.1 g/cm³ Compression to 30 µm diameter
- No fuel loaded. 50/50 mix of A and B



Consider the reaction A(B,X)Y $N_Y = \sigma_{(b,x)} (N_A N_B / area)$ for $\sigma_{(b,x)} = 0.01 pb (10^{-38} cm^2)$ $N_Y = 10^{-38} cm^2 (10^{20} \cdot 10^{20} / 4\pi (30 \mu m)^2) \approx 10^6 +$



Conclusions

- Neutron-induced reactions are of interest for a variety of applied and basic science reasons
 - s process
 - Reactor designs
 - Stockpile stewardship
- Surrogate reaction often the only way to estimate these cross sections
- NIF represents unique opportunities and challenges