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NEW CROSS SECTION NEEDS FOR ZIRCONIUM HYDRIDE
SNAP REACTORS*

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ABSTRACT

A zirconium hydride SNAP reactor consists of hydrided rods of U-Zr alloy surrounded by rotatable beryllium reflector drums. The current direction of SNAP reactor designs is toward reliable long-term (1 to 5 years) operation and additional safety features. These designs may require poison-backings on the reflector drums (for some applications) plus a variety of new burnable poisons.

Candidate poison-backings include europium and other rare earths as well as B₄C. Both absorption and scattering cross sections are important, particularly in the kev range. Candidate burnable poisons include Gd-157, Sm-149, Cd-113, Gd-155, Hf-177, Ir-191, Eu-151, Dy-162, Sm-152, and Dy-163. The absorption cross sections below 10 ev are particularly important.

One of the desired safety features in future zirconium hydride reactors is subcriticality in water in the event of a launch abort. This might be achieved through spectral shift if the burnable poison has a low-lying (below 0.1 ev) resonance. A large number of stable or long-lived isotopes which have not been measured for low-lying resonances are indicated.

INTRODUCTION

A zirconium hydride SNAP reactor consists of hydrided rods of U-Zr alloy surrounded by rotatable beryllium reflector drums. The current direction of SNAP reactor designs is towards reliable long-term (1 to 5 years) operation and additional safety features. Designs for both "unmanned" (involving the presence of instruments, etc.) and "manned" (involving the presence of humans) missions are being made.

Additional emphasis is placed on some of the conventional SNAP material cross section components, and the cross section needs for a wide variety of new isotopes is indicated.

BURNABLE PREPOISONS

Multi-year operation of a SNAP reactor requires the use of burnable prepoisons to match the reactivity loss rate. Reactivity is lost by hydrogen moderator loss from the fuel, by fuel burnup, and by fission product

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poisoning. The mismatch between the reactivity loss rate and the pre-poison burnout rate imposes increased control drum requirements, thereby limiting some SNAP reactor applications. Therefore, it is important to have as wide a variety of prepoisons to choose from as possible, and to know their effective absorption cross-sections well. The absorption cross sections below 10 ev are particularly important to the thermal SNAP reactor.

Prediction of the prepoison burnout rate is subject to several uncertainties:

- 1) cross-section uncertainties in the parent isotope
- 2) cross-section uncertainties in the daughter product (cross-sections of appreciable magnitude for the daughter products will cause a slower effective rate of poison burnout. An example of this is Eu-151. Two-thirds of the Eu-151 captures lead to a relatively stable metastate of Eu-152 which has about the same cross-section.)

All stable and nonstable isotopes were examined on the basis of available thermal cross-section and resonance integral information. (The CINDA indices were very useful here.) Isotopes in the gaseous and liquid state and those which produce gases were eliminated. Table I lists stable and radioactive isotopes which could not be evaluated because insufficient cross-section information exists. Table II lists those which could be evaluated and evidenced potential, in order of effective cross-section. Included also are estimates of isotope cost. The economically feasible isotopes (and important daughters) of interest for up to five-year SNAP reactor lifetime applications are given in Table III.

All the others in Table II are useful in combination with a fast burning low-resonance absorber (for intrinsic subcriticality). They may also prove useful by themselves if additional cross-section measurements and/or further analysis should increase their effective cross-section.

INTRINSIC WATER SUBCRITICALITY (IWS)

The problems associated with preventing accidental criticality in a SNAP reactor are more severe than for an earthbound reactor. Over-the-road transport and potential launch abort over the ocean could accidentally result in infinite water reflector and water flooding of the core.

As SNAP reactors are undermoderated, the addition of water to the reactor core results in a reactivity increase due to reduced neutron leakage and to softening of the neutron energy spectrum (Figure 1). (The spectral shift effectively transports neutrons to energies where the U-235 η is higher.) It appears that a method of eliminating this core reactivity increase and thereby achieving subcriticality in water ("intrinsic" water subcriticality, IWS) would be to add an absorber which has a resonance prominent in a water-moderated spectrum, but not in the normal operational ZrH-moderated spectrum. From Figure 1, one observes that for the spectrum to shift into a low-lying resonance, the resonance energy should lie below 0.1 ev.

Figure 2 plots for each isotope the lowest resonance energy recorded in the current BNL-325. Excluding the inert gas Xe-135, only four isotopes were found with thermal resonances at 0.1 ev or lower:

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<u>Isotope</u>	<u>E₀(ev)</u>
Sm-149	0.0976
Gd-155	0.0268
Gd-157	0.0314
Lu-176	0.104

Sm-149 and Lu-176 are marginally close to 0.1 ev, and therefore less attractive. Figure 3 illustrates the Gd-155 and Gd-157 cross-sections compared to a 1/v absorber. Of these two, Gd-157 burns out too fast for most reactor applications, posing impossible reactivity control requirements. Thus Gd-155 is the reference IWS absorber.

The Gd-155 worth lies predominantly in thermal absorption. The uncertainty in this worth is about 5% based on the most recent evaluations (Table IV). This uncertainty is not important at this time.

Gd-155 also burns out too fast for some reactor applications. Thus a wider variety of low-resonance absorbers featuring lower cross-sections is desired. Such isotopes might be found among the unmeasured long-lived radioisotopes, and stable isotopes obscured by many sister element isotopes (e. g., the seven stable isotopes of Yb). An attempt was made to find additional candidates by correlating lowest-resonance energies with atomic mass number, A (Figure 2). The lowest-lying resonances were found in the even-odd and odd-even isotopes. A literature search for isotopes located at the curve low points yielded Eu-155 with a resonance around 0.1 ev. However, Eu-155 is too radioactive to be useful. Hopefully, additional studies and new cross-section measurements will uncover more suitable substitutes which would increase the number of burnable poisons which are also useful for IWS.

REFLECTOR CONTROL DRUM OPTIMIZATION

Some manned mission applications may require complete (4π) shielding of the reactor. In order to reduce the total system weight, it is desirable to change from void-backed reflector drums (control by neutron leakage) to poison-backed reflector drums (control by neutron absorption). For the reflective material BeO is equivalent to Be in reactivity worth, and preferred for other engineering reasons. For the poison material, a Eu_2O_3 -metal matrix appears to be the preferred poison backing. Other poisons considered were boron compounds, tantalum, and other rare earths.

The relative importance of the Be and Eu cross section components upon this critical parameter, control drum worth, is explored by means of first-order perturbation theory in Tables V and VI. The components of highest worth require the most accuracy.

Another "reaction" of importance here is the zirconium transport cross section, between 500 ev and 10 Mev. Table VII shows that its first order worth is 26% $\Delta k/k$. (Zirconium constitutes 35 atom % of the core). Uncertainties here produce uncertainties in core leakage and, thereby, in drum control.

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TABLE I

STABLE AND NEAR STABLE ISOTOPES OF POTENTIAL USE
AS BURNABLE POISONS: NO AVAILABLE NUCLEAR DATA

Radioactive Isotopes	t 1/2 (yr)
Al-26m	7.4×10^5
Si-32	700
Ca-41	7.7×10^4
Ti-44	47
Fe-60	10^5
Ni-59	10^4
Ni-63	92
Se-79m	10^4
Mb-91m	long
Mo-93	10^4
Tc-97	10^6
Tc-98m	10^6
Sn-126	10^5
Ba-133m	7.2
La-137	10^4
La-138	stable
Pm-145	18
Sm-146	10^8
Gd-148	85
Gd-150	10^5
Gd-154	stable
Tb-157	150
Tb-158m	150
Dy-154	10^6
Ho-163	10^3
Yb-170	stable
Yb-171	stable
Yb-172	stable
Yb-173	stable
Hf-182	10^6
Os-186	stable
Os-187	stable
Os-188	stable
Pt-193	<500
Pb-202	10^5
Pb-205	10^7
Pb-210	22
Bi-208	10^5
Po-209	10^3
Th-229	7300

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TABLE II
EVALUATED BURNABLE POISONS

Isotope	SNAP Spectrum Weighted Cross Section (b)	Cost			t 1/2 (yr)
		\$/mg	Isotope Form	\$/mg Oxide	
Pd-107	13	NA*			10 ⁶
Gd-156	13	0.9	oxide		S†
Ac-227	28	NA			21.2
Nb-94	30	NA			10,000
Hf-179	30	1.55	oxide		S
In-113	49	12.0	oxide		S
Eu-154	49	NA			16
Hf-176	51	6.0	oxide		S
Dy-160	66	2.4	oxide		S
Ir-193	79	20.00	Ir	~\$3	S
Eu-153	84	1.00	oxide		S
Dy-164	85	0.25	oxide		S
Au-197	91	elem. §			S
Hf-178	92	0.50	oxide		S
Pa-231	94	5.6	oxide		32,480
Rh-103	99	elem.			S
Lu-176	100	43.00	oxide		S
Dy-161	108	1.50	oxide		S
Ag-109	109	0.60	Ag		S
Re-185	112	1.60	Re		S
Dy-163	113	1.40	oxide		S
Sm-152	173	0.35	oxide		S
Dy-162	192	0.15	oxide		S
In-115	197	0.45	oxide		S
Yb-168	384	32.0	oxide		S
Te-123	390	20	Te	~\$20	S
Eu-151	443	1.0	oxide		S
Ir-191	454	30.0	Ir	~\$4	S
Hf-177	462	1.5	oxide		S
Sm-151	526	NA			90y
Gd-155	908	1.4	oxide		S
Gd-113	1142	0.90	oxide		S
Sm-149	1719	0.70	oxide		S
Gd-157	3239	0.6 to 1.2	oxide		S

*NA: not commercially available

†S: stable

§elem.: 100% natural abundance in element, costs should be small compared to a separated isotope

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TABLE III
PERTINENT PREPOISON CROSS-SECTIONS FOR UP TO
FIVE-YEAR LIFE SNAP REACTORS

Parent Isotope	Important Daughter Isotopes
Gd-157	None; the parent isotope's cross-section is very high
Sm-149	Ibid
Cd-113	Ibid
Gd-155	Ibid
Hf-177	Hf-178, Hf-179
Ir-191	Ir-192, Ir-193, Pt-192
Eu-151	Eu-152 to Eu-156, Gd-152, Gd-154 to Gd-156
In-115	In-116, Sn-116
Dy-162	Dy-163
Sm-152	Sm-153, Eu-154
Dy-163	Dy-164, Dy-165, Ho-165

TABLE IV
GADOLINUM-155 2200 M/S ABSORPTION
VALUES REPORTED

Date	Cross Section (1000b)	Reference
9-60	60.6 ± 0.5	N.S. and E8, 183
1-60	56.2 ± 1.0	BNL-325 (1960)
7-59	88.7	Phys. Rev. 115, 424
58	66 ± 2	A/Conf. 15 P/11
7-58	61 ± 5	BNL-325 (1958)
57	87.2	Phys. Rev. 105, 196
50	78	Phys. Rev. 77, 634

TABLE V

BERYLLIUM RELATIVE CONTRIBUTIONS ($\% \Delta k/k$) TO CONTROL
DRUM WORTH (FIRST-ORDER PERTURBATION OF
TRANSPORT-THEORY CALCULATIONS)

Group	Lower Group Energy	σ_a	σ_{Tr}	σ_{g-g+1}
1	3.0 Mev	-0.0011	+0.0097	-0.0005
2	1.4 Mev	-0.0027	+0.0329	-0.0008
3	0.9 Mev	0.0000	+0.0128	-0.0058
4	0.4 Mev	0.0000	+0.0003	-0.0016
5	0.1 Mev	0.0000	-0.0017	-0.0094
6	17 kev	0.0000	-0.0053	-0.0017
7	3 kev	0.0000	+0.0014	-0.0093
8	550 ev	0.0000	+0.0002	-0.0074
9	100 ev	0.0000	+0.0013	-0.0041
10	30 ev	0.0000	+0.0042	-0.0041
11	10 ev	0.0000	+0.0130	-0.0017
12	3 ev	0.0000	+0.0223	-0.0004
13	1 ev	0.0000	+0.0216	+0.0003
14	0.4 ev	0.0000	+0.0234	+0.0040
15	0.1 ev	0.0000	+0.0662	-0.0005
16	0.009 ev	0.0000	+0.0469	
Total		-0.0038	+0.7456	-0.0431

TABLE VI

EUROPIUM RELATIVE CONTRIBUTIONS ($\% \Delta k/k$) TO CONTROL
DRUM WORTH (FIRST-ORDER PERTURBATION OF
TRANSPORT-THEORY CALCULATIONS)

Group	Lower Group Energy	σ_a	σ_{Tr}	σ_{g-g+1}
1	3.0 Mev	0.000	0.000	0.000
2	1.4 Mev	0.000	0.000	0.000
3	0.9 Mev	0.000	0.000	0.000
4	0.4 Mev	0.000	0.000	0.000
5	0.1 Mev	0.002	0.000	0.000
6	17 kev	+0.002	0.000	0.000
7	3 kev	+0.010	0.000	0.000
8	550 ev	+0.031	0.000	0.000
9	100 ev	+0.097	0.000	0.000
10	30 ev	+0.193	-0.005	0.000
11	10 ev	+0.310	-0.024	0.000
12	3 ev	+0.711	-0.091	0.000
13	1 ev	+0.762	-0.110	0.000
14	0.4 ev	+2.263	-0.469	0.000
15	0.1 ev	+2.217	-0.707	0.000
16	0.009 ev	+3.215	-0.722	
Total		+9.811	-2.128	-0.001

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TABLE VII
 REACTIVITY WORTH ($\% \Delta k/k$) OF ZIRCONIUM CROSS SECTION
 COMPONENTS (FIRST-ORDER PERTURBATION OF
 TRANSPORT-THEORY CALCULATIONS)

Group	Lower Group Energy	σ_a	σ_{Tr}	$\sigma_{g \rightarrow g+1}$
1	3.0 Mev	-0.009	<u>3.11</u>	-0.058
2	1.4 Mev	-0.030	<u>4.93</u>	-0.035
3	0.9 Mev	-0.026	<u>2.87</u>	-0.032
4	0.4 Mev	-0.062	<u>3.42</u>	-0.808
5	0.1 Mev	-0.083	<u>2.35</u>	-0.174
6	17 kev	-0.069	<u>1.57</u>	-0.014
7	3 kev	-0.036	<u>1.50</u>	-0.007
8	550 ev	-0.032	<u>3.45</u>	-0.004
9	100 ev	-0.475	0.42	-0.004
10	30 ev	-0.017	0.28	-0.006
11	10 ev	-0.014	0.41	-0.007
12	3 ev	-0.013	0.45	-0.006
13	1 ev	-0.015	0.41	-0.006
14	0.4 ev	-0.017	0.34	-0.008
15	0.1 ev	-0.046	0.61	-0.002
16	0.009 ev	-0.062	0.35	
Total		-1.007	26.46	-1.170

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Figure 1

Comparison of Thermal Neutron Spectra

Figure 2

AN EMPIRICAL STUDY OF THE LOWEST KNOWN RESONANCE ENERGIES

Figure 3
Total Gd-155 and -157 Cross
Section Times the Square
Root of Energy

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