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POISONING EFFECTS IN SPALLATION NEUTRON SOURCES.

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1 INTRODUCTION.

The practical design for a continuous spallation neutron source needs to take proper note of neutron economy. The critical region in and around the moderator should avoid the use of materials with a high thermal neutron capture cross-section, as far as this is practicable; for example, the excellent performance of heavy-water as a moderator stems largely from its very low capture cross-section and among many reasons why depleted Uranium is not used in the SINQ design is that the roughly factor-of-two gain of fast-neutron intensity is largely eroded by its high thermal capture probability. The actual choice of material is based on the absorption probability; the mechanical properties (strength of the material and the job it has to do) determine the thickness and it is this times the macroscopic cross-section which needs to be used in any comparison.

During operation, high-energy nucleon induced spallation reactions lead to production of nuclides which have a large thermal capture cross-section. A list of nuclides with σ_{cap} greater than 50b is given in Table I: The high-mass nuclides are spallation products of Lead & Bismuth, also the weak fission in these elements leads to the presence of nuclides in the medium mass range; the lower mass nuclides are possible products from structure materials.

Rather small quantities of material with cross-section in the kilo-barn range will lead to significant increase of absorption. The build-up rates of absorption in some materials considered for use in SINQ have been estimated and will be presented in the next section.

TABLE I

Nuclides with a thermal-neutron capture cross-section of > 50b.

	$t\frac{1}{2}$	σ_c	e		$t\frac{1}{2}$	σ_c	e		$t\frac{1}{2}$	σ_c	e
^{3}He	S	5327	1.8	^{6}Li	S	940	0.4	^{7}Be	53.29d	48000	19
^{10}B	S	3837	0.2	^{22}Na	2.602y	29000	3.4	^{39}Ar	269y	600	50
^{50}V	S	70	47.0	^{58}Co	9.1h	136000	7.3	^{58}Co	71.3d	1880	6.4
^{60}Co	10.48m	58	14	^{59}Ni	75ky	104	3.8	^{66}Cu	5.1m	135	7.4
^{74}Se	S	51.8	2.3	^{76}Se	S	85	8.2	^{83}Kr	S	200	15
^{87}Kr	76.3m	<600		^{103}Rh	S	150	3.3	^{104m}Rh	4.4m	800	13
^{105}Rh	35.5h	16000	9.4	^{109}Ag	S	91	3.3	^{110}Ag	249.9d	82	13
^{109}Cd	435d	650	15	^{113}Cd	S	19910	1.5	^{115}In	S	202	1.0
^{115}Sn	S	50	40	^{123}Te	S	406	7.4	^{125}I	60.14d	894	10
^{126}I	13d	5960		^{124}Xe	S	128		^{125}Xe	16.8h	<5600	
^{131}Xe	S	90	11	^{133}Xe	5.25d	190	47	^{135}Xe	9.1h	2650000	7.5
^{134}Cs	2.06y	140	8.6	^{138}La	S	172		^{143}Pr	13.58d	89	11
^{143}Nd	S	342	2.9	^{146}Pm	5.53y	8400	20	^{147}Pm	2.62y	181	3.9
^{148}Pm	5.37d	2000	50	^{148m}Pm	41.3d	22000	11	^{149}Pm	53.1h	1400	21
^{151}Pm	28h	<700		^{145}Sm	340d	110		^{147}Sm	S	64	7.8
^{149}Sm	S	41000	4.9	^{150}Sm	S	102	4.9	^{151}Sm	93y	15000	12
^{152}Sm	S	206	2.9	^{151}Eu	S	9200	11	^{152}Eu	13.3y	2300	43
^{153}Eu	S	390	7.7	^{154}Eu	8.8y	1500	27	^{155}Eu	4.96y	4040	3.1
^{152}Gd	S	1100	9.1	^{154}Gd	S	85	14	^{155}Gd	S	61000	0.8
^{157}Gd	S	254000	0.8	^{161}Gd	3.6m	31000	39	^{160}Tb	72.1d	525	19
^{160}Dy	S	61	9.8	^{161}Dy	S	585	5.1	^{162}Dy	S	180	11
^{163}Dy	S	130	7.7	^{164}Dy	S	2700	2.8	^{165}Dy	2.35h	3900	7.7
^{165}Ho	S	66.5	5.0	^{167}Er	S	670	4.5	^{171}Er	7.5h	280	11
^{169}Tm	S	103	2.9	^{170}Tm	128.6d	92	4.3	^{168}Yb	S	3470	2.9
^{171}Yb	S	50	8.0	^{174}Yb	S	65	7.7	^{176}Lu	S	2100	2.4
^{174}Hf	S	390	14.0	^{177}Hf	S	365	5.5	^{178}Hf	S	86	8.1
^{180}Ta	S	700	29.0	^{182}Ta	114.4d	8200	7	^{187}W	23.8h	64	16
^{185}Re	S	112	2.7	^{187}Re	S	74	5.4	^{184}Os	S	3000	5.0
^{187}Os	S	336	5.0	^{193}Os	30h	1540		^{191}Ir	S	924	5.7
^{192}Ir	74d	1100	36.0	^{193}Ir	S	112.5	6.7	^{197}Au	S	98.8	0.3
^{198}Au	2.7d	25800	4.7	^{196}Hg	S	3300	6.5	^{199}Hg	S	2000	50
^{200}Hg	S	<60		^{201}Hg	S	<61					

** For the half-life units, m= mins, h= hours, y= years, ky= 1000's of years and S= stable or very long.

e is the percentage error.

2 ESTIMATES FOR THE BUILD-UP OF ABSORPTION CROSS-SECTION.

The effect of thermal-neutron capture in the region of the target on the flux is illustrated in Fig 1: The thermal flux maps for the SINQ moderator with and without a 2cm thick Iron sleeve around the target (σ_{cap} circa 2b), show that in the latter case a flux depression of about 25% results.

The estimates for thermal-capture cross-section build-up are based on calculations using the HETC [1] code and, in the case of the target, with chain-yield analysis using the ORIHET [2] code. The materials have been selected from those which might be used for SINQ in the region of the moderator; the layout of this region of the source is sketched in Fig 2.

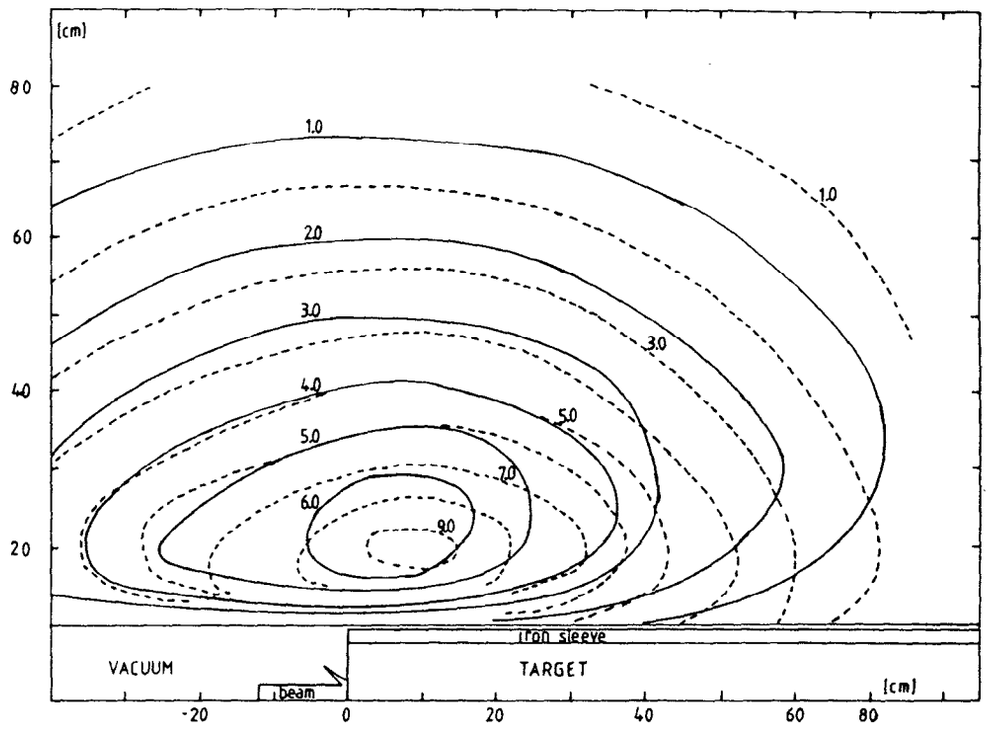

Fig 1: Flux maps for the SINQ moderator with (solid lines) and without (dashed lines) an absorbing Iron sleeve around the target. The diagram is cylindrically symmetric about the axis of the target. The flux intensities are in units of 10^{13} neutrons/(cm^2.sec) and correspond to 1mA at the target.

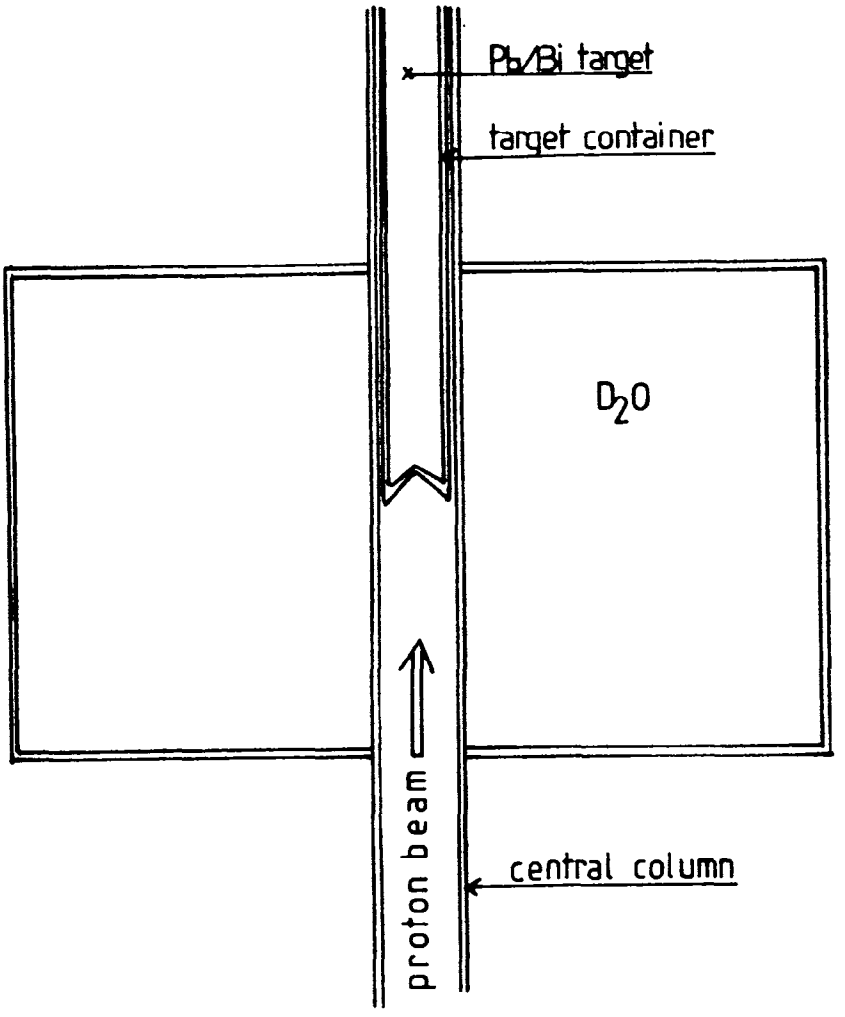

Fig 2: Sketch of the layout for SINQ in the region of the moderator. The principal material choices are for that of the target container and the central column; this latter is the barrier between the heavy-water and the proton channel vacuum.

2.1 Absorption cross-section for Lead-Bismuth under irradiation.

The Lead-Bismuth for the target has an unirradiated capture cross-section of 90mb. The nuclides produced include 57 giving a noticeable contribution to the increase of capture cross-section; the calculation includes fast-fission and chain-yield analysis. About 90% of the overall increase of capture cross-section comes from 9 nuclides and a further 11 contribute another 8%; these nuclides are listed in TABLE-II together with the calculated production rate, g, (grams after 10^7 seconds irradiation by a 1 mA 590 MeV proton beam), the thermal neutron capture cross-section for the product, σc, and the relative contribution to the increase (= g x σ_c/A, A is the mass number).

The increase of capture cross-section is given by:

$$\Delta\sigma_{abs} = \frac{20.8}{V} \sum_{n} \frac{9n \cdot \sigma_c}{An}$$

Where V is the total volume of the target and the sum is over the relative contribution for each of the nuclides produced. The numerical factor 20.8 is A/density for the Lead-Bismuth. The target is in the liquid state with motion from natural convection; it is assumed that the products will be uniformly dispersed throughout the total amount of material (0.25 m^3 in the current design).

TABLE-II

The high capture cross-section products in the SINQ target.

NUCLIDE	THERMAL CAPTURE CX (σ_c barns).	PRODUCTION AT 10^7 sec. and 1 mA (g grams).	$\frac{\sigma_c \, g}{A}$
^{199}Hg	2000	1.0	10.1
^{196}Hg	3080	.65	10.3
^{191}Ir	924	.45	2.2
^{184}Os	3000	.17	2.8
^{168}Yb	3470	.045	0.9
^{157}Gd	254000	.002	3.2
^{155}Gd	61000	.0059	2.3
^{113}Cd	19910	.021	3.7
^{58}Co	136000	.00064	1.5
^{201}Hg	<61	1.29	0.4
^{200}Hg	<60	1.20	0.4
^{197}Au	98.8	.825	0.4
^{187}Os	336	.313	0.6
^{177}Hf	365	.13	0.3
^{174}Hf	390	.085	0.2
^{167}Er	670	.049	0.2
^{149}Sm	41000	.0042	0.3
^{145}Sm	110	.00089	0.3
^{135}Xe	2650000	.0000089	0.2
^{104}Rh	800	.021	0.2

The increase of absorption cross-section after 10^7 seconds irradiation at 1 mA proton current is 3.4 mb and after one year irradiation at 3 mA (the lifetime and maximum current for SINQ targets), 30 mb or one third of the unirradiated cross-section value.

The extra volume of target material (included for technical reasons concerned with the cooling system) dilutes the produced nuclides about 50 times more than if its size were limited to that required by neutron production; were this factor not present, an expected increase of about 1.5 b would result, which in view of the results shown in Fig 1 would lead to a significant degradation of performance during operation.

2.2 Heavy-water moderator and structural materials.

The average high-energy neutron intensity leaving the first 30 cm of the target (the important region from the absorption point of view) is $5.5 \times 10^{-4}/(cm^2.sec.proton)$. The peak flux is about 30% higher. The build-up of absorption cross-section for material irradiated in this region is given by:

$$\frac{d\sigma_{abs}}{dt} = C \times \phi \quad \langle \sigma_p^i(E) \rangle \times \sigma^i_{abs}$$

Where $\langle \sigma_p^i(E) \rangle$ is the production cross-section for nuclide i averaged over the incident neutron spectrum, σ^i_{abs} is the absorption cross-section for product i and ϕ is the neutron flux. The value for $C \times \phi$ is 1.03×10^{-14} /(mb.sec) at 3 mA for the surface of the target. All results to be quoted will be for a proton beam-current of 3 mA at the target. The spectrum averaged production cross-sections have been computed by direct Monte-Carlo using the spectrum shown in Fig 3.

2.2.1 Heavy-water.

The build-up of absorption cross-section comes from the spallation products of oxygen and the results are summarised in Table-III.

TABLE-III

The build-up of absorbing nuclei in heavy-water.

Nuclide	$\langle \sigma_p^i(E) \rangle$ mb	σ_{abs} b	$\langle \sigma_p^i(E) \rangle \times \sigma_{abs}$ mbxb
^3He	0.8	5327	4262
^6Li	4.2	940	3948
^7Be	0.3	48000	14400
^{10}B	0.9	3837	3453

The absorption cross-section initially increases at the rate of 2.7×10^{-7} mb/sec. ^7Be has a half-life of 53.4 days and reaches an equilibrium absorption cross-section of about 1.0 mb and the build-up rate for stable nuclides is 1.2×10^{-7} mb/sec.

This will cause no problem for SINQ because of dilution through mixing into the large volume of heavy-water (about 8 m^3) and removal of some of these nuclides by the processing plant.

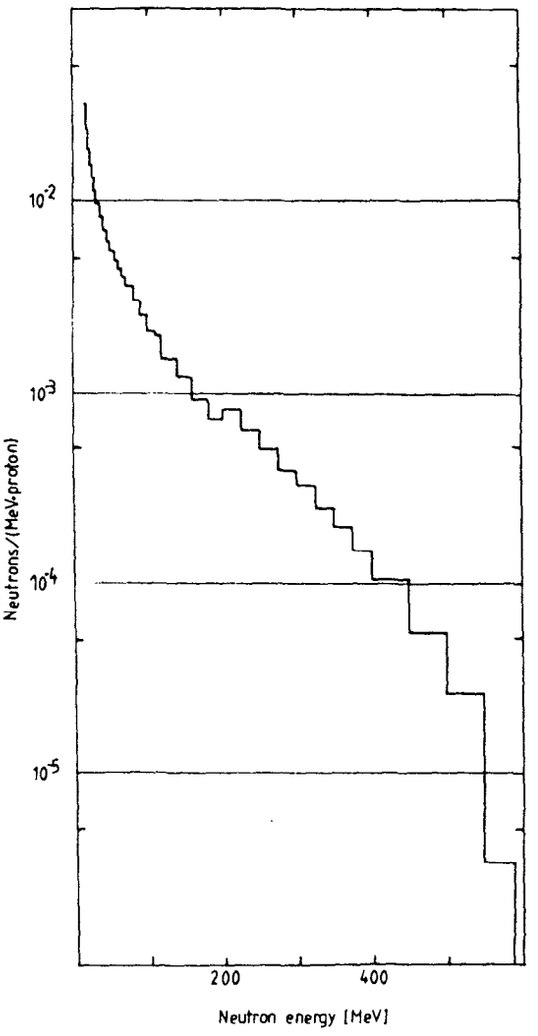

Fig 3: Energy spectrum for HE neutrons from the SINQ Pb/Bi target averaged over the length and all production angles. The spectrum has been calculated using HETC [1].

2.2.2 Structural materials.

The following materials are candidates for either the target container or the central column (see Fig 2).

(a) Carbon.

NUCLIDE	$\langle\sigma_p^i(E)\rangle$ mb	$\langle\sigma_p^i(E)\rangle \times \sigma_{abs}$ mb x b
^3He	1.8	9889
^6Li	20.0	18800
^7Be	3.9	187200
^{10}B	14.0	53718

The predicted initial increase rate of absorption cross-section is 9.2×10^{-7} mb/sec. Again, the ^7Be will come to equilibrium and reaches a value of 13 mb with the stable products giving an increase rate of 8.5×10^{-7} mb/sec. The absorption cross-section for carbon close to the target will rise from an initial 3.4 mb to about 43 mb at 1 year. There might be some slight reduction due to the ^3He diffusing out of the material. The increase of absorption cross-section does not exclude carbon as a target container material.

(b) Iron and Steel.

The production cross-sections of high-absorption cross-section nuclides for the principal constituents of stainless-steel are given in Table-IV.

TABLE IV

Spectrum average production cross-sections (mb) for high absorption cross-section nuclides in constituents of stainless-steel.

PRODUCT / MATERIAL*	^3He	^{50}V	^{36}Ar	^{39}Ar	^{58}Co	^{60}Co	^{59}Ni
Fe	1.3	1.6		0.1			
Ni	1.8	2.9			52.0	4.3	27.0
Cr	1.0	104.0	0.07	0.4			
Mo	0.7		<0.1				
S/S	1.2	28.0		0.12	6.5	0.5	3.4

* The cross-sections are averaged over natural isotopic mixtures and stainless-steel is taken to contain 68.5% Fe, 12.5% Ni, 16.5% Cr and 2.5% Mo.

For pure Iron, the unirradiated absorption cross-section will rise at a rate of 7.3×10^{-8} mb/sec; this has a small effect on the unirradiated absorption cross-section of about 2 b.

In stainless-steel, the ^{58}Co & ^{60}Co both reach equilibrium absorption cross-sections of 0.42 to 1.1 mb and 270pb respectively (^{58}Co has two states, 9.1 hr (0.136 Mb) and 71.3 days (1880 b) half-lives and the two values are for 100% of each state). The increase of absorption cross-section is predicted to be 8.7×10^{-8} mb/sec. At one year an increase of 3.2 mb in the absorption cross-section would result; this is small compared to the unirradiated value of 3.4 b.

(c) Aluminium.

The two high-absorption cross-section nuclides produced are ^{3}He and 2.6 year ^{22}Na. The spectrum averaged production cross-sections are 7 and 14 mb respectively. The ^{22}Na causes an increase of the absorption cross-section of 4.2×10^{-6} mb/sec at 1mA and reaches an equilibrium value of 490 mb. The increase from the ^{3}He is 5.4×10^{-8} mb/sec and gives a contribution at 1 year of about 17 mb. The unirradiated absorption cross-section of 230 mb will increase by a factor of about 3.

3 CONCLUSIONS AND DISCUSSION.

The increase of thermal neutron absorption cross-section, although noticeable, does not seem a major problem for SINQ operation. The use of a large volume liquid-metal target mitigates what could have been a significant degradation of performance as operation proceded. Similarly for the heavy-water, the large volume plus the processing avoids another source of performance reduction.

The carbon results are of more general interest as it is a possible candidate as a material to be used as the moderator. Unirradiated carbon is a good moderating material and has some attractions compared to heavy-water from a safety point of view as it is less easily dispersed. The increase of absorption cross-section causes the moderating ratio (average logarithmic energy loss times scattering cross-section divided by the absorption cross-section) to decrease by a factor of 5 during operation at positions close to the target but (obviously) more slowly at larger distances. This first look at the problem indicates a more careful study of the performance of a carbon moderator would be required before it could be accepted as an alternative to heavy-water.

On a more philosophical note, in 1834, Lenz deduced the law that in electromagnetism an induced current flows in a direction such that it opposes the change that produces it. This is a manifestation of a more general principle, that effects produced tend to oppose that which causes them and finds many examples in Physics and other systems: Seebeck and Peltier effects, the poisoning in reactors etc. We could translate the principle into the vernacular as "This looks wonderful, but what is the catch?".

What about spallation sources? They look wonderful, more proton current more neutron flux! What is the catch? Poisoning?

ACKNOWLEDGEMENT.

This paper is the result of the author annoying his group leader, Walter Fischer, with safety problems concerning the tritiation of the heavy-water moderator. His counter to these quibbles was to suggest the use of a Carbon moderator and was coupled with a request that such a moderator be examined. The author being somewhat reluctant, managed to generate this poisoning argument. WEF then asked "What about the target?" - hence this paper and my grateful thanks.

REFERENCES.

[1] Radiation Shielding Information Centre code package CCC-178, HETC.

[2] ORIHET is a modified version of the ORIGEN code, RSIC code package CCC-217.

M J Bell ORNL-4628 (1973)