INVESTIGATION INTO ANOMALOUS LEAD-CABLE RESPONSES IN VANADIUM SIR DETECTORS IN PT. LEPREAU AND GENTILLY-2

B. Sur, D.P. McAllindon and C.M. Bailey
Atomic Energy of Canada Ltd.

1.0 INTRODUCTION

Vanadium In-Core Flux Detectors (ICFDs) are used for flux mapping in CANDU 6 stations. The Inconel lead-cables (LCs) of these detectors produce a small (0.5% - 2%) but significant signal contribution which varies with the in-core length of the lead-cable. The ICFD signal must therefore be corrected for the lead-cable contribution to avoid biasing the flux map. Corrections for lead-cable contributions are made on the basis of a prediction of lead-cable relative sensitivity and its change with irradiation history due to the complex burn-up of the Inconel constituents. Lead-cables without any detectors are installed at the stations and monitored regularly to validate the lead-cable relative sensitivity prediction. A survey of the detector-less lead-cables at Pt. Lepreau and Gentilly-2 shows an unpredicted rapid increase and subsequent decrease in lead-cable relative sensitivity in the first few hundred full-power days after installation.

In this paper, the prediction of lead-cable relative sensitivity and its change due to burn-up effects are reviewed. The data from the stations is presented and modeled. Possible physical causes of the rapid variation in sensitivity are explored. Further work to resolve the issue of anomalous lead-cable sensitivity is discussed.

This work was funded by New Brunswick Power and Hydro Quebec under a joint contract with Atomic Energy of Canada Ltd.

2.0 LEAD-CABLE RELATIVE SENSITIVITY

In-core flux detectors and lead-cables consist of a metallic central core (the emitter), separated from an outer metallic sheath (the collector) by a layer of oxide insulation. In a neutron and gamma flux, electrons are ejected from, or collected on the emitter, generating a charge with respect to the (grounded) collector. When the emitter and collector are connected through an external circuit, a current is produced in that circuit. The magnitude of the current is proportional to the neutron and gamma flux.

Vanadium flux-mapping ICFDs have a pure ^{51}V emitter and produce a positive current in a thermal neutron flux. This current is due to the net transfer of electrons, produced by β-decays of 2.25 m (half-life) ^{52}V, from the emitter to the collector. Lead-cables used in CANDU reactors have both core-wires (0.24 mm diameter) and outer sheaths (1.12 mm O.D., 0.181 mm wall thickness) made of Inconel 600. The dimensions of the core-wire and sheath have been chosen so that there is a cancellation between the charge (electrons) transferred in to and out of the core-wire by neutron and γ-induced reactions in the elemental constituents of Inconel 600. Consequently, metre for metre, lead-cables are expected to produce only 0.05% as much positive current as Vanadium SIR detectors. Integrated over the length of the lead-cable in the core of a typical CANDU reactor, the net lead-cable contribution to the Vanadium detector current is expected to vary from approximately 0.5% to 2.5%.

The total flux experienced by lead-cables increases, in general, with their in-core lengths. Thus the lead-cable contribution, though small, induces a significant artificial tilt in the Vanadium ICFD-based flux map, and must be subtracted. The lead-cable relative sensitivity, α, is a convenient parameter to characterize the lead-cable contribution, assuming that the properties of all the lead-cables (for ~102 Vanadium detectors) are uniform.

Relative sensitivity is defined by the following relationships:

$$\alpha = \frac{S_{LC}}{S_D} = \left(\frac{i_{LC}}{\sum \phi_{LC}}\right)\left(\frac{\phi_D}{i_D}\right) = \left(\frac{i_{LC}}{i_T - i_{LC}}\right)\left(\frac{\phi_D}{\sum \phi_{LC}}\right) \qquad (1)$$

where: α is the relative sensitivity of the lead-cable,

S_{LC} is the absolute sensitivity of the lead-cable (output current per unit length per unit flux),

S_D is the absolute sensitivity of the detector,

i_{LC} is the measured lead-cable current,

i_D is the detector current,

i_T is the measured total detector + cable current,

ϕ_D is the detector flux, and

$\sum \phi_{LC}$ is the lead-cable flux, summed over the appropriate number of detector-length lead-cable segments.

The value of α changes with time (irradiation) since both i_{LC} and i_D change due to nuclear transmutation (burn-up). A first-order correction for the lead-cable contribution can be made for each Vanadium detector signal using the values of ϕ_{LC} inferred from the uncorrected flux map, and knowledge of α. Because ϕ_{LC} is different in general for each lead-cable segment, both the contribution of that segment, and the change in that contribution due to burn-up must be tracked individually. The variation in current from the detector is programmed into module *DLCSENSIT in the RFSP simulation code, which is used to calculate the mapped bundle and channel fluxes. For the calculation, the lead-cable is subdivided into n sections each one lattice pitch in length since the flux and burnup on each section is different.

2.1 Current Production Mechanisms in Inconel 600 Lead-Cables

Currents in in-core flux detectors and mineral-insulated cables are produced mainly by three types of interactions, namely (n, γ, e), (γ, e), and (n, β). These interactions can occur in the emitter, collector, or insulator, resulting in an electron being ejected from that material. Depending on where the electron is ejected from and deposited, a net positive charge, net negative charge, or no net charge is produced on the emitter. The probability of the interactions depends on the neutron and gamma cross-sections, density, and atomic number of the material, the energy spectrum of the neutron and gamma flux, and detector geometry.

The currents or sensitivities of mineral-insulated Inconel 600 lead-cables has been studied experimentally by Allan and Lynch [1], and theoretically (by Monte Carlo simulations) by Hall [2]. The major elemental constituents of Inconel 600 are nickel (74%), Chromium (15%) and Iron (9%). The response of Inconel 600 detectors and lead-cables and the time-variation of its components is complex because there are a large number of isotopes as well as potential

contaminants, notably manganese, that contribute to the total current. These currents are discussed briefly below.

(n, γ, e): The absorption of a neutron by an atom results in the immediate emission of capture gamma rays. The gamma rays can then produce energetic electrons (or positrons) in matter by Compton scattering, the photoelectric effect, or pair production. The (n, γ, e) mechanism produces current instantaneously in response to neutron flux and is therefore referred to as a "prompt" interaction. In an Inconel lead-cable the (n, γ, e) interaction produces a net positive current that varies approximately as the cube of the emitter diameter. All elements present in Inconel 600 contribute to this current. However, roughly 50% of the (n,γ) interaction rate in Inconel 600 is due to ^{58}Ni (~75% Ni in Inconel 600; 68% natural abundance of ^{58}Ni in Nickel; thermal neutron cross-section of Inconel, $\langle\sigma_{th}(\text{Inc.})\rangle = 4.1$ b, and $\sigma_{th}(^{58}\text{Ni}) = 4.6$ b). Neutron capture on ^{58}Ni produces ^{59}Ni ($t_{1/2} = 7.6 \times 10^4$ a, $\sigma_{th}(^{59}\text{Ni}) = 78$ b). Thus the isotope ^{59}Ni, whose initial abundance is zero, "burns in" with irradiation of Inconel 600, and increases the total (n,γ,e) current as a function of FPD. The dominant time constant for this increase in current is equal to the inverse of the neutron transmutation rate:

$$\tau(^{59}Ni) = \frac{1}{\phi \cdot \sigma(^{59}Ni)} = 593.5 \text{ effective FPD} \qquad (2)$$

where ϕ, the neutron flux has been taken to be 2.5×10^{18} m^{-2} s^{-1}. This effect causes S(n,γ,e) to increase with time.

(γ, e): External reactor gamma rays (as opposed to neutron capture gamma rays generated inside the detector) can also eject electrons; this is called the (γ, e) interaction. The probability of this interaction depends on the flux and energy of the external gamma rays, and the atomic number, Z, and density of the matter with which they interact. No transmutation of atoms occurs in the (γ, e) interaction. The (γ, e) mechanism produces current promptly in response to gamma flux. However, the gamma flux is itself composed of prompt and delayed fractions. In a generic CANDU core, about 70% of gamma flux is prompt. In the mineral-insulated LC, the (γ, e) interaction contributes a net negative current that varies approximately linearly with the mass of the sheath. Since nuclear transmutation due to neutron captures and subsequent β-decays can, at most, change Z for any isotope by 1, it is assumed that S(γ,e) does not change appreciably due to burn-up effects vs. irradiation time.

(n, β): An atom absorbs a neutron, becomes unstable and decays with a characteristic half-life by the emission of an energetic electron or β-particle. As a result of this process, the original atom transmutes into an element with a higher atomic number and atomic mass. This mechanism results in the production of current with a half-life characteristic of the unstable isotope. Vanadium, rhodium, and manganese are examples of elements that produce current by the (n, β) process. In the mineral-insulated lead-cables, ^{56}Mn ($t_{1/2} = 2.58$ h) and ^{65}Ni ($t_{1/2} = 2.52$ h) are the main isotopes contributing to a net negative (n,β) current. This negative current is apparently the residual effect in the emitter, of competing positive (electrons ejected) and negative (electrons deposited) charge deposition due to β-decays in the emitter and collector respectively [2]. The abundance of these short-lived isotopes, hence the (n,β) current, requires a few hours to reach equilibrium with the ambient neutron flux. This causes a difference between steady-state sensitivity, S, and sensitivity after a long reactor shut-down, S*.

2.2 Predicted Time-Variation of LC Sensitivity and Dynamic Response

The prediction for lead-cable sensitivity is based on an estimate by C.J. Allan. This prediction was made for the nominally 1.0 mm diameter prototype Bruce B lead-cables. The prediction for each current producing mechanism is shown in Table 1.

Table 1
Estimated Values of the Sensitivity of a Standard 1 mm Inconel 600 Lead-Cable as a Function of Irradiation Time in a Mean Flux of 2×10^{18} $m^{-2}s^{-1}$

Parameter	Value ($\times 10^{-27}$ $Am^{-1}/(m^{-2}s^{-1})$)			
	T = 0 a	T = 2 a	T = 4 a	T = 6 a
S(n, γ, e)	2.6	3.6	3.7	3.6
S(γ, e)	-0.4	-0.4	-0.4	-0.4
S(n. β)	-0.4 to -1.6	-0.4 to -1.4	-0.4 to -1.2	-0.4 to -1.1
S_T (avg.)	1.2	2.3	2.5	2.4
S_T*	2.3	3.3	3.4	3.3

where,

$S_T = S(n,\beta) + S(n,\gamma,e) + S(\gamma,e)$ = equilibrium sensitivity

and,

$S_T^* = S(n, \gamma, e) + 0.7S(\gamma, e)$ = sensitivity after startup following a long shutdown and before appreciable buildup of ^{56}Mn ($t_{1/2}$=2.58 h) and ^{65}Ni ($t_{1/2}$=2.52 h).

Allan states that the estimates of total sensitivity are *subject to a lot of uncertainty* because of a large uncertainty in the contribution of the (n,β) sensitivity. This uncertainty is caused primarily by the unknown amount of ^{55}Mn impurity in the Inconel 600 used for the experimental cables on which the above predictions are based.

The calculation of the (n,β) current is further complicated by the following effects:

- The β-spectrum extends over a wide energy range, from 0 to 2.1 MeV (^{65}Ni) or 2.8 MeV (^{56}Mn). The total electron energy spectra at very low energies, which also contains contributions from conversion electrons, low-energy gamma rays and atomic x-rays, has large uncertainties.

- Effects due to low-energy electrons become progressively more important as the dimensions of the detector (lead-cable) become smaller.

- The net current is due to a cancellation between positive and negative charge deposition from β-decays in the emitter and collector respectively. Thus, small errors in the calculation of either effect are amplified in the net result. Significant variations in the positive and negative currents can also occur from small variations in the dimensions or in the isotopic content, especially manganese, of the emitter or collector.

- The neutron flux at the emitter is slightly smaller (~99.2%) than that at the collector due to self-shielding. Again, because of the cancellation involved in the net (n,β) current, the slightly different burn-up rates can give rise to relatively large changes in the net current as a function of irradiation time.

The long-term variation in the (n,β) current is due to the burn-in or burn-up of the parents of the β-decaying nuclei, i.e., ^{64}Ni and ^{55}Mn. Since manganese is present as an impurity in Inconel 600, the initial current produced by this isotope can vary over a wide range. The current from this small impurity (specified to be < 0.3% for this application) is significant because (a) the only naturally occurring (n,β) current-producing isotope in Inconel 600, ^{64}Ni, has a very small initial abundance (~ 0.68% of isotopic Inconel 600) and (b) the thermal neutron cross-section of ^{55}Mn (13.3 b) is almost a factor of ten greater than that of ^{64}Ni (1.52 b). Both ^{55}Mn and ^{64}Ni are also burnt-in by neutron capture on preceding isotopes. The most important burn-in effect is presumed to be that of the former from consecutive neutron captures on ^{52}Cr, ^{53}Cr, ^{54}Cr to produce ^{55}Cr ($t_{1/2}$ = 3.5 m), and the subsequent β-decay of ^{55}Cr to ^{55}Mn. The largest interaction rates, and therefore the smallest time constant in this series of neutron captures, are those due to ^{53}Cr (σ_{th} = 18 b; τ = 2572 FPD) and ^{55}Mn (σ_{th} = 13.3 b, τ = 3481 FPD), where an effective flux of 2.5×10^{18} m^{-2} s^{-1} has been assumed. The calculation of the (n,β) current due to ^{56}Mn as a function of irradiation time [2] exhibits a change over a period of six to ten years as expected.

For the time variation of S(n,β) in Table 1, Allan has simply assumed that the negative current burns out with an effective cross-section that depends on the initial current. For the lower limit of |S(n,β)| at T=0 in Table 1, he assumes a burn-up cross-section of 2 b (close to σ_{th} of ^{64}Ni), and for the upper limit of |S(n,β)|, he assumes 10 b. This is used to produce the range of S(n,β) as a function of irradiation time.

Since the geometry of the lead-cables is now different from when this original prediction was done, the values of sensitivity should be scaled by the experimentally determined dependence of the current-producing mechanisms on detector dimensions. This leads to (in units of 10^{-27} Am^{-1}/(nm^{-2}s^{-1})):

$$S(n,\gamma,e) = 0.24^3/0.22^3 * S(n,\gamma,e) = 3.38$$

$$S(\gamma,e) = S(\gamma, e) * M_{sheathnew}/M_{sheathold} = -0.48$$

$$S(n,\beta) = \text{unknown}$$

Therefore, we expect that the total sensitivity at T=0 a has increased by 0.7 to 1.9 x 10^{-27} Am^{-1}/(nm^{-2}s^{-1}).

Thus, the total sensitivity is a balance between a positive (n, γ, e) contribution and negative (γ, e) and (n, β) contributions. Uncertainty in any of the numbers can cause large differences in the initial sensitivity, burn-up effects and dynamic response.

The ratios of the components affects the dynamic response of the lead-cable. The (n, γ, e) is a prompt component. The (γ, e) is approximately 70% prompt with the remainder being composed of many different time constants. The (n, β) is expected to be caused by ^{55}Mn and ^{64}Ni in the Inconel of the sheath and collector with a time constant of 3.7 h. The prompt fraction, F_p, is thus calculated to be:

$$F_p = \frac{S(n,\gamma,e) + 0.7S(\gamma,e)}{S_T} \qquad (3)$$

Using the original values in Table 1, the prompt fraction at zero irradiation is predicted to be 193%. Using the new values for the new size of lead-cable, the prompt fraction is predicted to be 163%.

3.0 MEASURED RESPONSES

The lead-cable contributions based on the above prediction are used to correct the vanadium flux map. Instead of depending entirely on the prediction of lead-cable relative sensitivity, α, the CANDU 6 stations at Pt. Lepreau and Gentilly-2 installed two "bare" or detector-less lead-cables each at the time of SIR detector replacement in 1992. The current produced by each installed lead-cable is measured periodically at both stations to track and validate the model of lead-cable contribution and its variation with fluence.

The initial absolute sensitivities and locations of these installed lead-cables are shown in Table 2. These values have been calculated for Pt. Lepreau in the Pt. Lepreau spreadsheet [4] and for Gentilly-2 by taking the product of the calculated α's and the expected vanadium detector sensitivity of 30×10^{-25} $Am^{-1}/(m^{-2}s^{-1})$. The values vary around the predicted value of 1.9×10^{-27} $Am^{-1}/(m^{-2}s^{-1})$. These values were taken about one month after the lead-cables had been installed, so the initial value of sensitivity may well have been smaller.

Table 2
Absolute Initial Sensitivities of Installed Lead-cables

Installed Lead-cable	Sensitivity [$Am^{-1}/(m^{-2}s^{-1})$]
VFD11 (Pt. Lepreau)	1.72×10^{-27}
VFD14 (Pt. Lepreau)	2.34×10^{-27}
VFD12 (Gentilly-2)	2.07×10^{-27}
VFD16 (Gentilly-2)	1.86×10^{-27}

The results of the tracking of installed lead-cable currents [3], [4] as a function of effective full-power days (FPD) in the reactor are shown in Figure 1 and Figure 2. Figure 1 shows the raw current produced by each lead-cable. Figure 2 shows the relative sensitivity calculated for each lead-cable along with the predicted relative sensitivities of the installed lead-cables (based on Table 1). Note that the predicted relative sensitivity is specific to the cable because each cable is modeled as n lattice-pitch length sections that contribute current and burnup at different rates depending on their position in core. The predictions for the lead-cables were provided by C. Newman [5].

It is clear from these figures that there is a discrepancy between the predicted and the measured sensitivity of all four lead-cables. All four lead-cables show a faster, higher increase in sensitivity than the prediction in the first 200 FPD of irradiation. The three lead-cables with generally the same response also have an unpredicted decrease in sensitivity in approximately the next 200 FPD. It is also clear that there is a significant range of variation between the four measured lead-cable sensitivities as a function of FPD. In particular, the installed lead-cable in

Pt. Lepreau VFD-11 does not exhibit the rapid decrease in sensitivity (following the initial increase) as seen by the other three installed LCs.

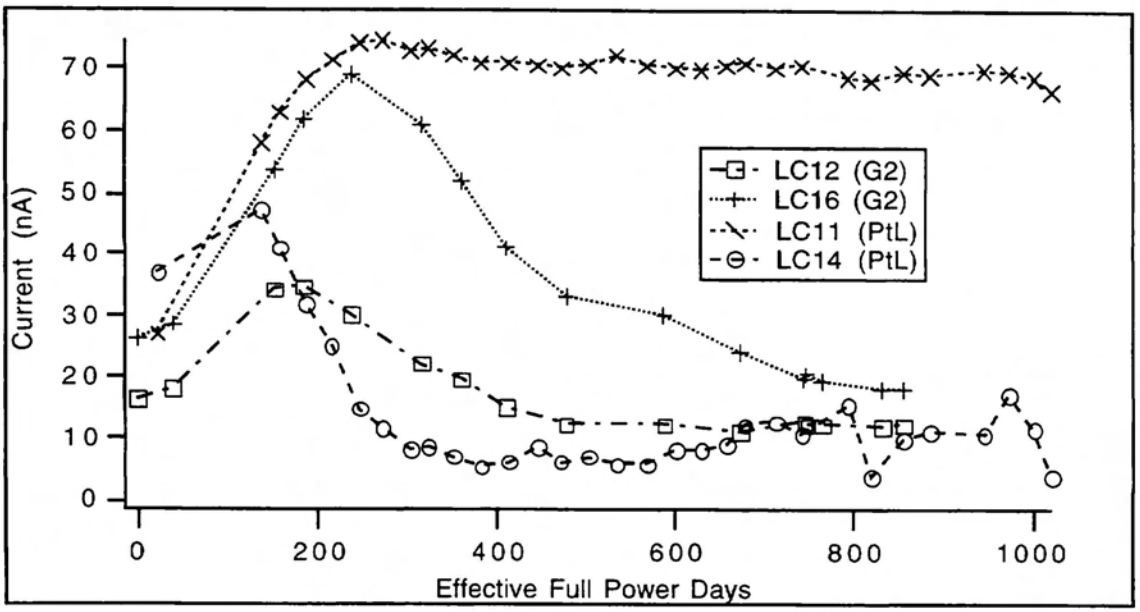

Figure 1: Measured Lead-cable Currents

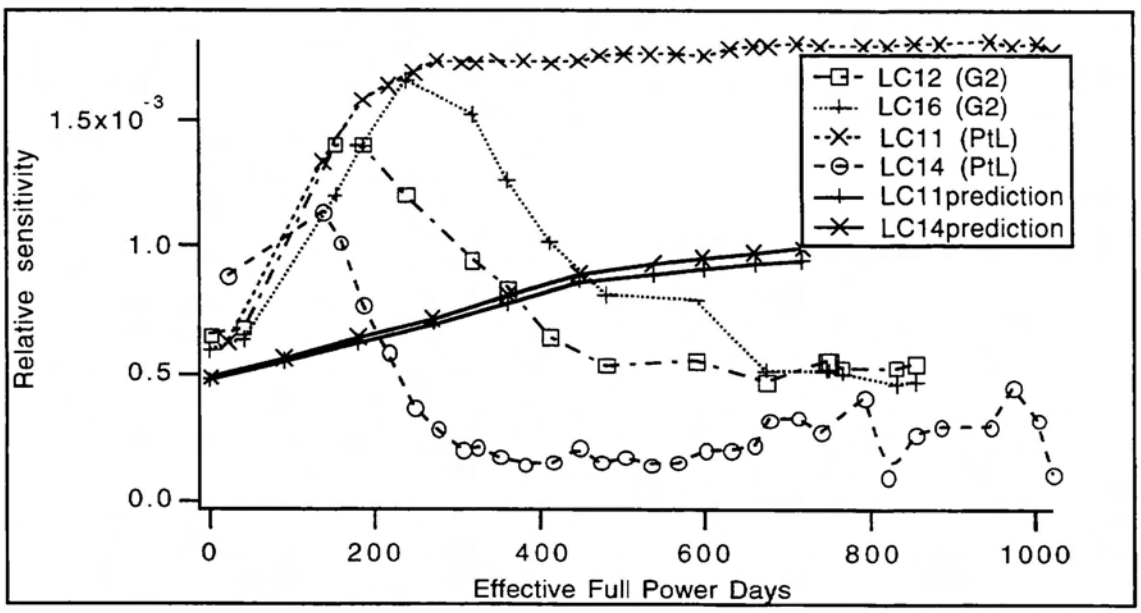

Figure 2: Comparison of Predicted and Measured Relative Sensitivities

The behavior of the other three lead-cable responses is also consistent with tilt differences (a) between measured and mapped detector fluxes, and (b) between mapped and heat-balance-

deduced fluxes as a function of irradiation in the first 400 days following detector replacement at both Pt. Lepreau and Gentilly-2 [6].

These observed discrepancies give rise to the following questions:

1) Are there cable-to-cable variations in the lead-cable constituent materials, especially those with low concentrations(e.g. ^{55}Mn)?
2) Are the installed lead-cables representative of the lead-cables in the 102 Vanadium flux-mapping detectors?
3) Should the predicted lead-cable sensitivity variation with irradiation (α vs. FPD) be revised?
4) Is there sufficient allowance for the uncertainty in the lead-cable contribution?

Thus the scope of the study funded by New Brunswick Power and Hydro Quebec was the following:

a) Review site operational data on lead-cable responses, both steady-state (Figure 1 and Figure 2) and after a shut-down.
b) Obtain and review material assays for the detectors and their cables, plus the installed lead-cables.
c) Develop models for material burnout andbuildup that could explain the observed behaviour.
d) Recommend new lead-cable α variation curve to be used by sites.

4.0 REVIEW OF DETECTOR AND LEAD-CABLE MATERIAL ASSAYS

The material test reports and specifications for the Vanadium detectors and installed lead-cables for both Pt. Lepreau and Gentilly-2 were reviewed. This information was contained in the history dockets provided by the detector manufacturer (Imaging and Sensing Technilogy Inc.), as part of the contract for the replacement SIR detectors It is apparent from the chemical analyses of the materials used to fabricate the Vanadium detector emitters, the Inconel lead-cable core-wires and the common Inconel sheaths, that all detectors and installed lead-cables for both stations were fabricated, respectively, from the same materials.

The following observations are made from the examination of these reports:

1. The initial concentration of manganese in the core wire (0.29 - 0.30%) is different from that in the sheath (0.19 - 0.20%). From these reported manganese concentrations, a calculation of the (n, β) sensitivity caused by ^{55}Mn in the lead-cables can be made . The calculated value is -0.05×10^{-27} $Am^{-1}/(nm^{-2}s^{-1})$. Since the original prediction of the (n,β) sensitivity caused by ^{55}Mn is unknown, we are uncertain how to adjust the prediction using this calculation.

2. The O.D. of installed lead-cable VFD-11 in Pt. Lepreau (1.10 mm) is smaller than the O.D. of the other three installed lead-cables (1.12 mm) by 1.8%. Assuming that the core-wire of this lead-cable is also smaller than those of the other three by a similar amount, then S(n,γ,e) for installed lead-cable VFD-11 in Pt. Lepreau (which varies as the cube of the core-wire diameter) will be smaller than that of the other three installed lead-cables by about 5.5%. It is difficult to distinguish this possible effect from the scatter of the initial sensitivity measurements shown in Table 2.

3. The O.D. of the detector lead-cables have a range of variation (maximum - minimum) of 3.7% in Gentilly-2 and 2.1% in Pt. Lepreau. There is no data available about the core-wire diameters. Again, assuming that the core-wire diameters have a similar range of variation, then the range of variation of S(n,γ,e) should be ~11% in Gentilly-2, and ~ 6% in Pt. Lepreau. The range of variation of the total sensitivity would be expected to be greater than this by a factor of 1.5 to 2 since there is a cancellation of S(n,γ,e) by the other two sensitivities (see section 2.2).

4. The material assays do not indicate the presence of any contaminant elements in the Inconel 600 (other than Manganese), at approximately the 0.1% concentration level and with large neutron cross-sections (with the possible exception of Cobalt).

5.0 MODEL DEVELOPMENT (FOR 100 FPD SENSITIVITY CHANGE)

The lead-cable may be viewed as a set of detectors, each one lattice pitch in length, connected in series. The total current generated by the lead-cable, $I_{LC}(total)$, is the sum of currents generated from all the n segments that constitute its length:

$$I_{LC}(total) = \sum_{i=1}^{n} I_{LC(i)} \qquad (4)$$

In each segment, i, the current is the sum of those produced by each mechanism:

$$I_{LC(i)} = I_i^{(n,\gamma,e)} + I_i^{(\gamma,e)} + I_i^{(n,\beta)} \qquad (5)$$

For each mechanism, x, the current in each segment is the sum of charge deposition per unit time due to interactions in the core (or emitter) and sheath (or collector):

$$I_i^x = {}^{core}I_i^x + {}^{sheath}I_i^x \qquad (6)$$

This statement is particularly germane to the (n,β) current, but may not be important for the other two mechanisms, since experimentally the (n,γ,e) current is proportional to the cube of the core diameter only, and the (γ,e) current depends linearly on the sheath mass.

The current in each segment due to a particular mechanism is the sum of currents due to each isotope, j, that participates in that mechanism:

$$I_i^x = \sum_j I_i^{x,j} \qquad (7)$$

and if there are N_i^j atoms of such an isotope in segment i at time t, then:

$$I_i^{x,j}(t) = A^{x,j} N_i^j(t) \cdot \sigma^j \cdot \phi_i(t) \qquad (8)$$

where:

σ^j is the thermal-neutron cross-section for (n,γ,e) and (n,β), and the γ-interaction cross-section for (γ,e);

$\phi_i(t)$ is the thermal-neutron flux for (n,γ,e) and (n,β), and the gamma flux for (γ,e) at segment i;

$A^{x,j}$ is a coefficient that relates current production to interaction rate for the particular interaction type, x, and the particular isotope, j.

Note that for (n,β) nuclei, such as ^{65}Ni and ^{56}Mn (with short half-lives), the current is actually proportional to the β-decay rate. But in the event that observations are made after a period of steady-state operation that is long compared to these half-lives, and if their neutron-capture rates are ignored, then these β-decay rates are equal to the neutron-capture rates of their parents. Thus the N_i in equation (8) refer to the abundances of the parents of the β-decaying nuclei which produce the (n,β) currents viz. ^{64}Ni and ^{55}Mn in the above-mentioned cases. Also, particularly in the case of (n,β), the coefficient A may be radically different for the core and sheath, with opposite signs.

The number of atoms, N_i^j(t) is governed by a set of coupled first-order differential equations:

$$\frac{d}{dt} N_i^j = N_i^{j-1} \sigma^{j-1} \phi_i + N_i^{j'} \lambda^{j'} - N_i^j (\sigma^j \phi_i + \lambda^j) \qquad (9)$$

The first two terms on the right hand side refer to production of N_i^j by neutron-induced transmutation of a preceding nucleus, (j-1) and by β-decay (or electron-capture) of an isobaric (long-lived) nucleus, $N_i^{j'}$, with decay rate $\lambda^{j'} = 1/\tau^{j'} = \ln 2 / t_{1/2}^{j'}$. Similarly the third term refers to the depletion or burn-out of nucleus j itself due to neutron induced transmutation and β-decay.

The determination of N_i^j requires solving simultaneously the equations for all the preceding nuclei that contribute to its production. This requires knowledge about their initial abundances, cross-sections and decay rates, and about the neutron flux as a function of time. If the neutron flux at each segment is assumed to be constant with time, then the neutron-induced transmutation terms in equation(9) can also be cast in terms of constant transmutation rates:

$$\lambda_i^j = \sigma^j \phi_i \qquad (10)$$

The transmutation time constant for a nucleus with 100 b cross-section in a neutron flux of 2.5 $\times 10^{18}$ m^{-2} s^{-1} is 463 d. Time constants can be obtained by inserting the values of cross-section and flux into the following equation:

$$\tau_i^j = \frac{1}{\lambda_i^j} = 463 \text{ FPD} \times \left(\frac{100 \text{ b}}{\sigma^j}\right) \times \left(\frac{2.5 \times 10^{18} \text{ m}^{-2} \text{ s}^{-1}}{\phi_i}\right) \qquad (11)$$

Equation (9) can now be solved for $N_i^j(t)$ by standard methods. The solution involves the sum of a series of exponential "delay" terms:

$$N_i^j(t) = \sum_{k,k'=0}^{j} B_i^{k,k'} \exp\left(-\lambda_i^{k,k'} \cdot t\right) \qquad (12)$$

where the index, k (k'), runs over all the nuclei whose transmutation (β-decay) contributes to the abundance of N_i^j . The coefficients, B_i^k, are complicated functions of initial abundances and transmutation or β-decay rates of all the nuclei in the transmutation chain.

Working back to equation (4) it is clear that the long-term variation in current from a detector or lead-cable is expected to also consist of a sum of exponential terms:

$$I_{LC}(total) = \sum_{core,sheath} \left(\sum_{i=1}^{n} \left[\sum_{x,j} A^{x,j} \lambda_i^j \sum_{k,k'=0}^{j} B_i^{k,k'} \exp\left(-\lambda_i^{k,k'} \cdot t\right) \right] \right) \qquad (13)$$

For an Inconel lead-cable, with many different segments i, and several current producing nuclei, j, there will be a large number of terms with different time-constants involved in such a sum. A rapid variation in current or sensitivity as seen in the data, requires terms with time constants close to the time-scale of variation. As shown in section 2, none of the transmutation chains considered in the prediction of lead-cable sensitivity have such short time constants.

To simplify the enormous complication embodied in equation (13) and to gain insight into the cause of the rapid variation of lead-cable sensitivity, the observed normalized sensitivity curve was modeled by the following simple equation:

$$y(t) = I(0) + k1\, e^{-t/\tau_1} + k2(1 - e^{-t/\tau_2}) \qquad (14)$$

The unitless quantity, y, represents the normalized relative sensitivity, i.e., the ratio of the sensitivity at time, t, to that at time zero. The value of y can be scaled by the actual initial relative sensitivity of the lead-cable to obtain α(t). k1 and k2 are amplitudes of two currents with burn-up or β-decay lifetimes given by τ_1 and τ_2 respectively. I(0) represents the initial sensitivity or current. The long-term variation in sensitivity due to $^{58,59}Ni$ and ^{55}Mn is not included in the above model, but may be incorporated either as a slow time variation in I(0) or with additional exponential-delay terms.

Figure 3 is similar to Figure 2 with the addition of two empirical curves based on equation (14). The curves labeled "model1" and "model2" use the parameters listed in Table 3.

Table 3
Parameters for the Empirical Model

Parameter	model1	model2
I(0)	6	6
k1	-5	-5
k2	-5.5	-1.5
τ_1	100 days	100 days
τ_2	200 days	200 days

These values for the parameters were chosen because the resulting curves seem to model the actual lead-cable sensitivities fairly well (although some further fine tuning may be possible, it won't help much at this preliminary stage). The only difference between the models is the value of k2. The relative values of k1 and k2 can be tuned to fit the varying lead-cable sensitivities.

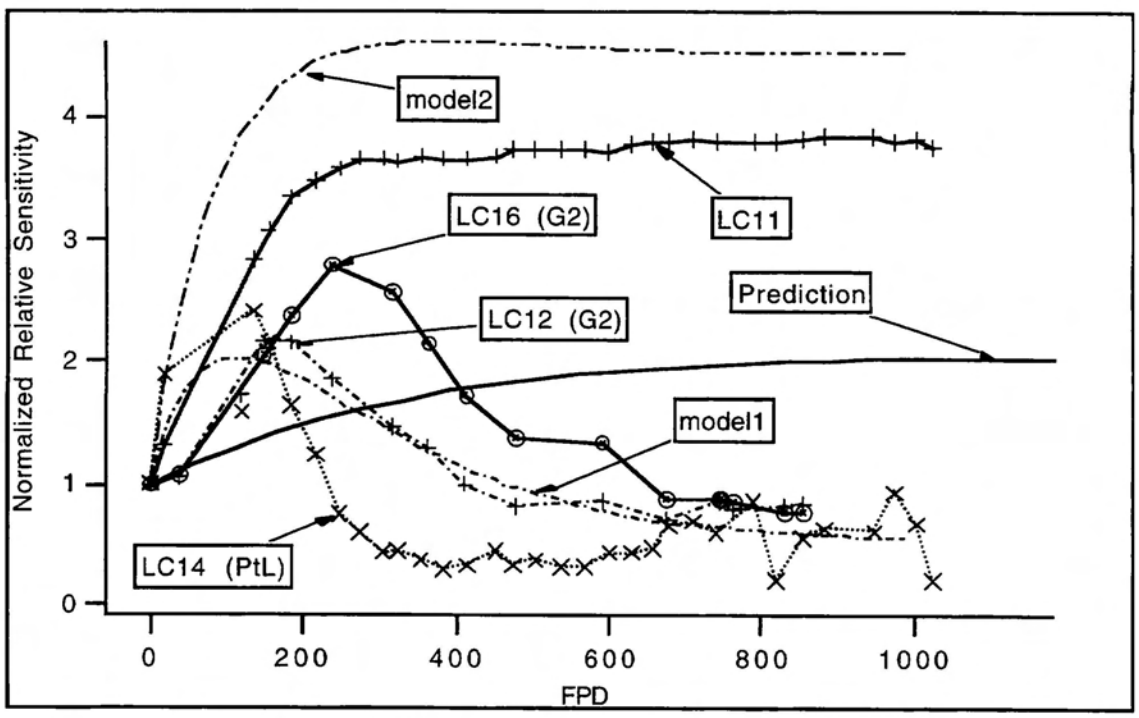

Figure 3: Measured Lead-Cable Normalized Relative Sensitivity and Model

In the case of nuclei that produce current by β-decay or electron capture, the time constants of interest, i.e., 75 to 250 d correspond to half-lives of 50 to 175 d. In the case of nuclei whose neutron-induced transmutation is involved, the time constants correspond to the product of neutron flux and capture cross-section of the component that is being transmuted. Assuming an average neutron flux of 2.4×10^{18} $m^{-2}s^{-1}$, the predicted capture cross-section for the k1

component in the above model is 330 b ±25%. The predicted capture cross-section for the k2 component is 170 b ±25%. The range of variation in amplitudes and time constants that can fit the data is ± 25%.

The simple model in equation (14) gives an idea of the time constants and amplitudes of currents that can cause the observed variation in lead-cable sensitivity. Several specific scenarios that can cause this behaviour are possible:

A) Single current: A single trace isotope, X, with 150-350 b cross-section and negligible current production (from, say, (n,γ,e)) is initially present in the lead-cable material. Its burn-up produces isotope, Y, which has either a high (n,β) cross-section or a 100-200 d half-life. The abundance of Y will thus increase and subsequently decrease in the required time-frame. This scenario requires the isotope Y to produce a net positive current by (n,β), hence it is likely present in the core-wire. The burn-up product of Y should also produce negligible current.

B) Opposite polarities burning out: There are two isotopes, X and Y, present initially which produce competing positive and negative currents, and thus a close-to-zero net signal. Both X and Y have transmutation cross-sections in the 150 - 350 b range. If the positive current producing isotope burns out at a smaller rate than the negative, then the modeled behaviour would be reproduced. Again, there is negligible current production from the resulting burn-up products.

C) Opposite polarities burning in: There are two isotopes, X and Y, present initially which themselves produce negligible currents. But their burn-up products, X' and Y', produce competing positive and negative currents. If the positive current burns in at a faster rate (shorter time constant) than the negative current, then the observed behaviour will be reproduced. In this scenario, either the parents, X ,Y, must have high cross-sections *or* their respective progeny, X', Y', must have high cross-sections or β-decay half-lives characteristic of the 100 - 200 FPD build-up lifetimes required in the model.

D) Burn-in and Burn-out: In this scenario, there is an initial negative current that burns out with ~100 FPD lifetime and a second negative current which burns-in asymptotically with a ~200 FPD life-time. The isotope, X, producing the initial negative current must have a high burn-up cross-section, while isotope, Y, producing the second negative current either has a ~200 day half-life or its *parent* has a high burn-up cross-section and negligible current production. It is possible (though unlikely) that X is the parent of Y.

In scenarios B, and C the positive and negative currents represented by X and Y may also be due to the same isotope which produces currents with opposite polarities, and different efficiencies and burn-up rates due to its presence in both the core and sheath.

The investigation of possible physical sources for the components and their time variation in the above model is outlined in the following sections. In this investigation, transmutation chains in both the known normal constituents of Inconel as well as possible (trace) contaminants were examined.

6.0 MODEL CANDIDATES

In the search for candidate nuclei or transmutation chains that give rise to the observed or modeled (but previously unpredicted) behaviour of lead-cable sensitivity versus time, the following general observations are made:

- The initial sensitivities of the lead-cables agree well with the prediction. Therefore it is likely that either (a) the rapid change in sensitivity is caused by a previously un-accounted series of transmutations of a normal Inconel constituent, or (b) the initial concentration of the (contaminant) candidate isotope in Inconel 600 is small.
- The (n,β) mechanism is a far more efficient current producer than either (n,γ,e) or (γ,e). Since the candidate nucleus likely has a small concentration, it probably contributes current via the (n,β) mechanism.
- There is no *a-priori* reason why the (n,β) current *must be negative* [1] as postulated in the case of ^{64}Ni and ^{56}Mn. The net current in these cases is calculated to be the result of cancellation between positive (from the core) and negative (from the sheath) currents and is < 10% of the magnitude of these individual (positive and negative) currents [2], with ~100% uncertainties.
- Low-energy electrons (from β-decays, internal conversion, interactions of nuclear de-excitation γ-rays and atomic de-excitation processes) and x-rays, with energies of the 1 to 500 keV have ranges (1 to 500 μm) comparable to the radial thicknesses of the lead-cable core, insulation and sheath (~ 0.2 mm each). Such low-energy electromagnetic radiation is therefore expected to play a much larger role in lead-cable current production, as opposed to the case of SIR detectors where the major current contribution is from the much thicker emitter.

For the reasons outlined above, both the polarity and the magnitude of (n,β) currents are very dependent on the energy spectrum of electrons, and on the dimensions of the lead-cable components. It is plausible that the anomalous lead-cable (Pt. Lepreau LC-11) response vs. time is due to the burn-up of the same constituents as the other three LCs, with differing absolute magnitudes of competing negative and positive currents because of its somewhat smaller diameter.

6.1 Candidates in Inconel 600 Constituents

An examination of the nuclear properties of the constituents of Inconel 600 reveals that there are two isotopes in the normal burn-up routes with half-lives, hence time constants, comparable to those required in the model. These isotopes are ^{51}Cr ($t_{1/2}$=27.7 d) and ^{59}Fe ($t_{1/2}$=44.5 d). They are produced in Inconel 600 via the following transmutation chains:

$$^{50}\text{Cr}\ (n,\gamma)\ ^{51}\text{Cr}\ (\text{Electron Capture})\ ^{51}\text{V}\ (n,\beta)\ \ldots$$

and

$$\ldots{}^{58}\text{Fe}\ (n,\gamma)\ ^{59}\text{Fe}\ (\beta^-)\ ^{59}\text{Co}\ (n,\gamma)\ \ldots$$

The "seed" nuclei for these transmutation chains have relatively low abundances and are not expected to contribute substantially to the overall (n,γ) current. ^{51}Cr decays by electron capture and produces intense low-energy (~ 5 keV) x-rays, and monoenergetic (320 keV) γ-rays and conversion electrons. ^{59}Fe produces low-energy β's and two intense γ-rays. ^{51}V and ^{59}Co are well-known current producers, used as emitter material in detectors.

The abundance of the nuclei in each of the above chains was calculated as a function of FPD, (assuming a flux of 2.5 x 10^{18} $m^{-2}s^{-1}$) by solving equation (9). This calculation was done by

starting (at 0 FPD) with only ^{50}Cr and ^{58}Fe, with abundances in Inconel 600 of 0.653% and 0.028% respectively. These initial abundances are the products of the respective isotopic abundances and their elemental concentrations in Inconel 600, taken to be nominally 15% for Chromium and 10% for Iron. The resulting abundances, as a function of FPD, were converted into absolute interaction (or decay) rates by multiplying them with the corresponding neutron interaction rate or decay rate. The coefficients for converting these interaction rates to currents are not known. Therefore, for comparison, these interaction rates were divided by the known interaction rate, at zero FPD, of a "standard" β-decaying component of Inconel, taken to be ^{64}Ni (1.5 b cross-section, 0.683% abundance in Inconel 600). The relative interaction rates for each of the isotopes in the above chains is plotted as a function of FPD in Figure 4 and Figure 5. It is seen that the decay rate of ^{51}Cr reaches a peak in about 100 FPD at a value of 9.5 times the initial interaction rate of ^{64}Ni. The decay rate of ^{59}Fe saturates in about 200 FPD, but its maximum value is only about 3.3% relative to ^{64}Ni. Note in this context that the interaction rate of 0.3% ^{55}Mn is about 5 times that of the standard ^{64}Ni interaction rate in Inconel 600.

As an illustration, the interaction rate of the last nuclide in the above chains is multiplied by an appropriate factor (indicated in the respective figures) and subtracted from that of ^{51}Cr and ^{59}Fe respectively. The resulting curves (in Figure 4 and Figure 5) show a time variation which is reminiscent of the observed variation in lead-cable currents. The underlying is that ^{51}Cr and ^{59}Fe decays produce positive currents that build in with irradiation time, and that ^{51}V and ^{59}Co produce negative currents (with larger current production "efficiency").

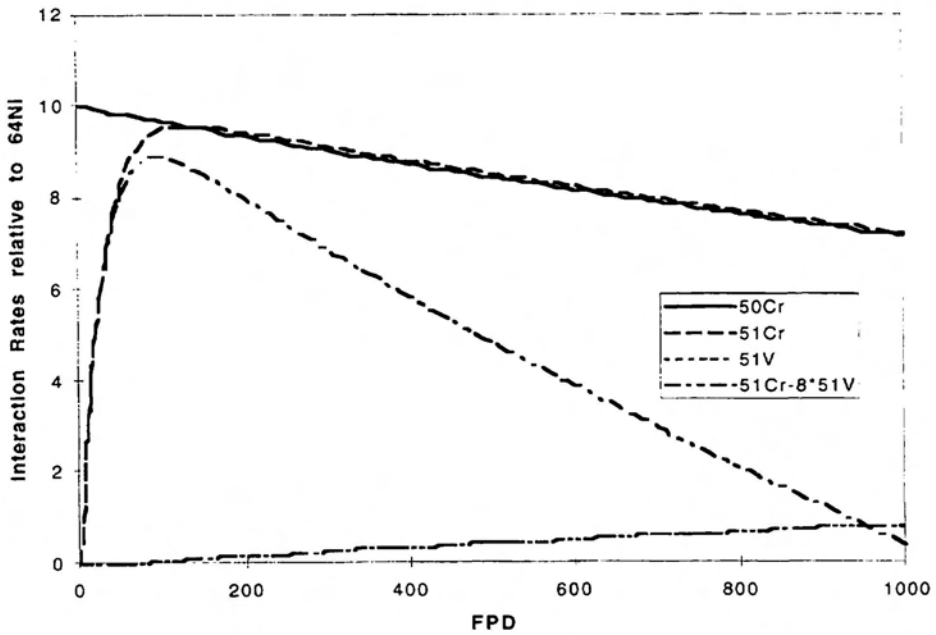

Figure 4: Interaction Rates of ^{50}Cr, ^{51}Cr and ^{51}V relative to ^{64}Ni vs. FPD in Inconel 600

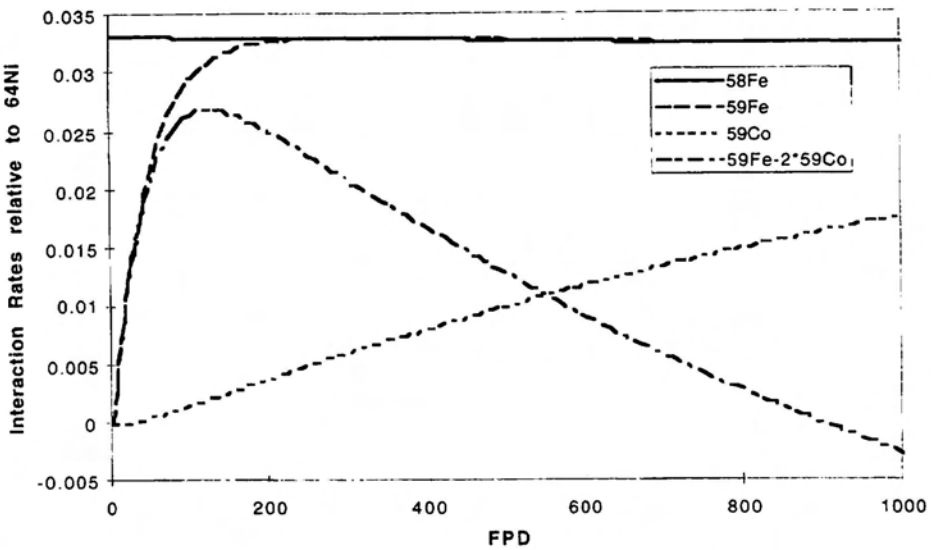

Figure 5: Interaction Rates of ^{58}Fe, ^{59}Fe and ^{59}Co relative to ^{64}Ni vs. FPD in Inconel 600

In principle, there are a large number of possibilities for adding and subtracting currents due to the isotopes in these (and other) transmutation chains. There are expected to be both positive and negative contributions of different magnitudes from the interactions of each of these isotopes in the core and the sheath. There should also be current contributions due to each isotope with slightly different neutron transmutation life-times because of differences in flux between the core and sheath (~ 0.8% difference) and along the length of the lead-cable due to varying proximity to fuel channels and large-scale flux variations in the core itself (10% - 50%).

Other potentially interesting transmutation chains in Inconel 600 are:

$^{58}Ni\ (n,\gamma)\ ^{59}Ni\ (n,p)\ ^{59}Co$ ... followed by

$^{59}Co\ (n,\gamma)\ ^{60}Co\ (\beta)$, $(n,\gamma)\ ^{61}Co\ (\beta)$

and

$^{54}Fe\ (n,\gamma)\ ^{55}Fe$ (EC and (n,p)) $^{55}Mn\ (n,\beta)$

The issue of current contributions and burn-out rates due to these and other potential isotopes can be evaluated in detail by numerical methods. The evolution with irradiation time of abundances and interaction rates is relatively easy to calculate by solving the coupled differential equations represented by equation (9). The only reliable method of calculating the (n,β) current contributions in Inconel 600 lead-cables (and detectors) is to perform Monte Carlo calculations.

The possible currents due to the isotopes mentioned at the beginning of this section, i.e. ^{51}Cr and ^{59}Fe, represent essentially delayed components in the dynamic response of the installed lead-cables. If they are the cause of the 100-200 FPD rise and fall in lead-cable current following reactor start-up, then they will also be present after a shut-down, and an appropriate decay in this current should be observable in the event of a long reactor shut-down of the order of a month or more. The pattern of rapid rise and then fall in current should also be reproduced when the reactor becomes operational after such a long shut-down. The effect will be modified

because the initial abundances at the present time will not be the same as at the start of life of these lead-cables.

It should also be possible, in principle, to discern the effects of such postulated currents in normal run-down tests. The lead-cable currents should show an offset after the "normal" delayed currents (mainly 2.5 h half-life) have decayed away. However, because of the very small absolute magnitude of the delayed current (~ 1 - 10 nA), it may be difficult to distinguish such an offset from an amplifier zero offset.

6.2 Candidate Contaminants

Current-producing interactions, with burn-up lifetimes in the 100-200 FPD, range can also be caused by isotopes with nuclear transmutation cross-sections in the 150-350 b range, as outlined in section 5. No such candidates exist in the normal constituents of Inconel 600, or at the 0.1% (1000 ppm) contamination levels in the assayed elemental constituents of these lead-cables (Section 3). However, an (n,β) current-producing contaminant with such high cross-sections needs to be present at only 100 ppm levels to compete with the standard (n,β) interaction rate of ^{64}Ni in Inconel 600. A contaminant that produces current by (n,γ), even with such high cross-sections, would need to be present at the 1-10% level to compete with the (n,γ) interaction rate from ^{58}Ni or ^{59}Ni. Several scenarios can be constructed based on the postulated current producing properties of such contaminants and their burn-up chains:

- A single contaminant or a combination of contaminants with either equal concentrations in the core and sheath, or with different concentrations, initially produces a negligible (n,γ,e) current. The rapid transmutation of this high-cross-section contaminant produces a daughter, or combination of daughter products that results in a net positive (n,β) current burning in with a time-scale of 100-200 FPD. The subsequent decay or transmutation of the (n,β) producing daughter, leads to the observed decrease in positive current in 200-300 FPD.
- A combination of contaminants, or different isotopes of an elemental contaminant or a combination of a contaminant and a normal Inconel 600 constituent produces a negative and a positive (n,β) current that burn out at different rates. The net result of this combination is the rise and fall in current observed in the installed lead-cables.
- Although it is unlikely that a contaminant can produce a (n,γ,e) current which would compete with the normal Inconel components, this possibility cannot, *a priori*, be ruled out.

An examination of the Chart of the Nuclides provides no single candidate nucleus that would satisfy the criterion for both the rise and fall seen in the lead-cable sensitivity (with the possible exception of ^{109}Ag). Thus we have to look for two contaminants whose burn-ups separately contribute to the fast rise and subsequent decay in sensitivity. Candidate isotopes have been identified and are listed in Table 4.

There are several other elements and isotopes with neutron cross-sections in the 100 - 400 b range. Most notable as candidates among these are Au and Hg. However no obvious scenario consistent with the observed rise and fall in sensitivity could be constructed based on these contaminants.

If the rapid change in sensitivity observed in the installed lead-cables is caused by the presence of high-cross-section trace contaminants, then these contaminants have likely burnt-out in the ~1500 FPD that have elapsed since detector installation. Since (n,β) currents cause a delayed dynamic response, it may be possible to discern the effects of such contaminants by analyzing the dynamic response of the Vanadium detectors and installed lead-cables following a fast

power run-down or trip. The object would be to look for time constants of these contaminants which are not present in normal Inconel detector responses. The burn-out can potentially also be probed by comparing dynamic responses from the newly installed detectors and lead-cables to those at present.

A possible source of trace contamination on the lead-cable sheath is residual lubricant left over from the manufacturing process. The lubricant is used in conjunction with a die to draw out the lead-cables to their required diameter and length.

Table 4

Candidate Contaminant Isotopes for rapid change in LC sensitivity.

Isotope	Capture cross-section (b) thermal, resonance integral	half-life of daughter	Concentration required to compete with LC currents
^{59}Co	37 , 74	10.5 m, 5.3 y	1% by weight
(^{60}Co build up from ^{59}Co)	2 + 4	1.65 h	(build up from ^{59}Co)
^{115}In	194 , 3200	2.2 s, 14.1 s, 54.2 m, 1.2 m	100-200 ppm
^{103}Rh	145,1100	42.3 s, 4.36 m	100-200 ppm
^{109}Ag (48% abundance)	87,1380	25 s, 250 d	~ 100 ppm.
^{110}Ag ($t_{1/2}$=250 d)	80	1.1 m, 7.5 d, .. 3.1 h	(build up from ^{109}Ag)
$^{(156,...164)}$Dy	<940 , 1600>	1.26 m, 2.33 h	20 - 100 ppm
$^{(162,...170)}$Er	<160 , 740>	2.3 s, 7.5 h + (γ,e)	1000 ppm
^{169}Tm	105, 170		~ 200 ppm
^{191,193}Ir	420, 282	19.3 h + (γ,e)	50 - 100 ppm

Contaminants at the 100 ppm level can be easily identified by chemical and isotopic analyses techniques, such as Inductively Coupled Mass Spectroscopy (ICPMS). For the proposed high cross-section contaminants, it should also be relatively simple to perform Neutron Activation Analysis (NAA). It is recommended that such analyses be performed (with very small samples) on a representative selection of the spare lead-cables, made of the original detector materials that exist at the stations. The confirmation or refutation of the presence of any candidate contaminant will greatly simplify the picture with respect to burn-out effects in lead-cable sensitivity.

6.3 Evaluation of Models By Dynamic Response Analysis

Data on lead-cable and vanadium detector trip responses from Pt. Lepreau and Gentilly-2 have been acquired. The July 1992 trip in Pt. Lepreau took place immediately after the replacement with new vanadium SIR detectors and "installed" lead-cables. Thus, by comparing the dynamic response of these detectors and lead-cables between July 1992 and April 1995, it may be possible to identify changes corresponding to the burn-in and burn-out of current producing contaminants in the material of these detectors. These data were therefore examined visually for all 102 Vanadium detectors in Pt. Lepreau and for the four installed lead-cables in Gentilly-2

and Pt. Lepreau. An attempt was made to analyze the amplitudes and time constants present in the response of representative lead-cables and detectors from both stations.

In summary, it can be stated that there are no definite indications of any time-constants that are not expected to be present in Inconel components, and that can therefore be associated with possible contaminants. However, dynamic response data from the installed lead-cables when they were new (~ 1992) is not available. Since high-cross-section contaminants may have burnt-out by 1995, the absence of such time constants cannot itself be taken to be an indication of the initial lack of trace contaminants.

The data on the installed lead-cables does have offsets at long times (~1 d) after shut-down, indicating the presence of currents produced by relatively long-lived (hence low decay-energy) radioactive isotopes. As mentioned previously, two such isotopes are ^{51}Cr and ^{59}Fe. The dynamic response of the "anomalous" lead-cable (Pt. Lepreau VFD-11) has delayed-response terms with approximately the same time constants, but radically different polarities than the other three installed lead-cables. This is an indication of the significant dependence of (n,β) currents on dimensions for such small diameter "detectors".

7.0 CONCLUSIONS AND RECOMMENDATIONS

Because there are many possibilities for isotopes and transmutation chains that can potentially explain the observed anomalous behaviour of the lead-cable sensitivities, no definite conclusion about the cause of this variation can be drawn at present.

A crucial test that will narrow the range of candidate possibilities is the confirmation or elimination of any high-cross-section contaminants that may be present in the un-irradiated lead-cables. It should be relatively straightforward to assay a lead-cable sample from the original spare detectors at the 100 ppm level by mass spectroscopy or neutron-activation analysis.

Another useful test is to probe for the presence of long-lived delayed currents (from normal Inconel constituents) in the event of a long reactor shut-down or subsequent start-up. A third diagnostic tool would be the analysis of dynamic response of the installed lead-cables when they were relatively new, and comparison to present trip data.

A statistical analysis of data from all the vanadium detectors at Pt. Lepreau [7], [8] suggests that the α for all 102 detectors may be approximated by a constant value, which is related but not equal to the value assumed in the prediction. However, this analysis does not directly yield the value of α. Additional statistical analyses have been suggested to track the change in α from the date of installation to the present time [9].

A model of the type represented by equation (14), with parameter values as shown for "model1" in Table 3 is a good representation of the lead-cable sensitivity variation with time. Such a model can be refined, if required, by doing a detailed fit to the sensitivity as a function of actual fluence, averaged over length, experienced by the individual installed lead-cables. The uncertainty in the lead-cable sensitivity is $\pm$ 30%. This uncertainty is gauged by (a) the scatter of the data from the three "representative" installed lead-cables (Figure 3), (b) the range of dimensions of the lead-cables (section 4.0) and (c) additional statistical analyses of lead-cable contributions as outlined above.

If it is assumed that the 100 - 300 FPD rise and fall in lead-cable sensitivity, caused possibly by high cross-section contaminants, is over for all or most of the detectors then the data shows that a present constant value of $\alpha = 0.036\%$ is appropriate to use for flux mapping applications at

Pt. Lepreau and Gentilly-2 [9]. Such an assumption fits both the data from the installed lead-cables and comparisons of flux tilts from flux-mapping, flux-measurements, and heat-balances.

The lead-cable sensitivity and dynamic response variation with time are important issues for both conventional flux-mapping and for proposed on-line flux-mapping schemes using dynamic compensation of Vanadium detectors. Thus the resolution of this issue warrants further study. Possible approaches are (a) statistical analyses of Vanadium detector responses and comparison with RFSP-calculated sensitivities (b) Monte Carlo calculations of current contributions in Inconel combined with knowledge at the parts per million level of Inconel constituents and (c) experimental determination of the separate current contributions by measurements on installed lead-cables in a power or research reactor.

REFERENCES

[1] C.J. ALLAN and G.F. LYNCH, "Radiation Induced Currents in Mineral-insulated Cables", IEEE Transactions on Nuclear Science, Vol NS-27, No. 1, 1980 February.

[2] D.S HALL, "Theroretical Study of the Effects of Manganese on the Sensitivity and Prompt Response of Bruce B SIR Detectors", CRNL-2592, 1986.

[3] D. MINH NGUYEN, private communication, 1994 Le 17 aout.

[4] R.W. SANCTON, "Recommended Initial Lead-Cable Sensitivities", PLGS internal report PIR 93-010, 1993 March 5; D. COLEMAN, private communication, 1996 April.

[5] C. NEWMAN, private communication, 1996 July 17.

[6] C.M. BAILEY et al., TTR-545, Addendum, Vol 3 Appendix D1, 1995.

[7] J. HANDBURY, see paper in these proceedings; also private communications 1996 August, 1996 September, 1996 December.

[8] C. M. BAILEY, private communication, 1996 August 28.

[9] C.M. BAILEY, private communication, 1997 Februaury.