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ATOMIC ENERGY OF CANADA LIMITED

L'ÉNERGIE ATOMIQUE DU CANADA LIMITÉE

THE USE OF THERMOCOUPLES WHICH TRANSMUTE DURING SERVICE IN NUCLEAR REACTORS

Utilisation de thermocouples subissant une transmutation en cours de service dans les réacteurs nucléaires

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Résumé

Certaines expériences courantes relatives au combustible nucléaire faites à Chalk River requièrent l'emploi de thermocouples pour mesurer les hautes températures allant jusqu'à 2200°C dans les conditions de fonctionnement des réacteurs.

En consultant la littérature spécialisée on s'est aperçu que les effets électriques transitoires et la transmutation des alliages employés dans les thermocouples peuvent causer des erreurs de mesure de température pouvant aller jusqu'à ±1% et ±30%, respectivement. Cependant, toute erreur due à des effets électriques transitoires peut être corrigée en effectuant des mesures de température immédiatement après l'arrêt du réacteur. De plus, on s'est rendu compte que les effets de transmutation peuvent être corrigés en calibrant les thermocouples en tungstène-rhénium pour hautes températures en fonction d'un thermocouple en chromel-alumel dans un environnement assez froid.

En ayant recours à ces techniques on peut s'attendre dans le meilleur cas, à un pourcentage d'erreur de ±2% en ce qui concerne la mesure des températures.

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ABSTRACT

Some current nuclear fuel experiments at CRNL require the use of thermocouples to measure temperatures of up to 2200°C under reactor operating conditions.

A literature search has shown that transient electrical effects and transmutation of the thermocouple alloys can cause temperature measurement errors of up to ±1% and ±30%, respectively. However, the error due to transient electrical effects can be corrected by making temperature measurements immediately following reactor shutdown. Furthermore it has been shown that transmutation effects can be corrected for by calibrating the high temperature tungsten-rhenium thermocouples against a chromel-alumel thermocouple in a cooler part of the experiment.

The use of these techniques is expected to reduce temperature measurement errors to ±2% in the best case.

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1. INTRODUCTION

Some current experiments at CRNL(1) require the measurement of temperatures of up to 2200°C under reactor operating conditions. Thermocouples are usually used for this purpose because:

- they may be made small enough not to appreciably affect the experimental results;
- thermocouple materials which will survive the operating temperatures are available; and,
- thermocouple technology is well understood(2).

The X-2 loop blowdown experimental fuel element consists of enriched uranium oxide fuel pellets encased in a Zircaloy sheath (Figure 1). The element is mounted in a flow tube and the whole assembly is supported by a hanger bar in the X-2 loop in the reactor where the thermal neutron flux is 0.9×10^{14} $n \cdot cm^{-2} \cdot s^{-1}$. To obtain temperature information, thermocouples are located at the centreline and the periphery of the fuel, in the Zircaloy sheath, flow tube and coolant. The fuel and sheath thermocouples are tungsten/5% rhenium-tungsten/26% rhenium (tungsten-rhenium) and the coolant and flow tube thermocouples are chromel-alumel. Tungsten-rhenium is used in the fuel because fuel temperatures will exceed 1700°C, which is above the maximum operating temperatures of both chromel-alumel (1100°C) and platinum-rhodium (1700°C). All the thermocouples are metal sheathed and mineral insulated. In normal reactor operation, the fuel centre is at 1800°C-2200°C, the fuel periphery is at 800°C-900°C, and the sheath and coolant are at about 300°C. During blowdown the fuel periphery temperature can increase to about 1400°C, and the sheath to 1300°C.

The thermocouples are all 1.0 to 1.3 mm diameter and installed in good thermal contact with the measurement points to minimize temperature measurement errors. The target accuracy for temperature measurements over the life of the experiment is ±1%.

In the past, blowdown experiments have been operated in reactor for two weeks, giving a total dose of 1.1×10^{20} nvt. However some future experiments are planned to continue for

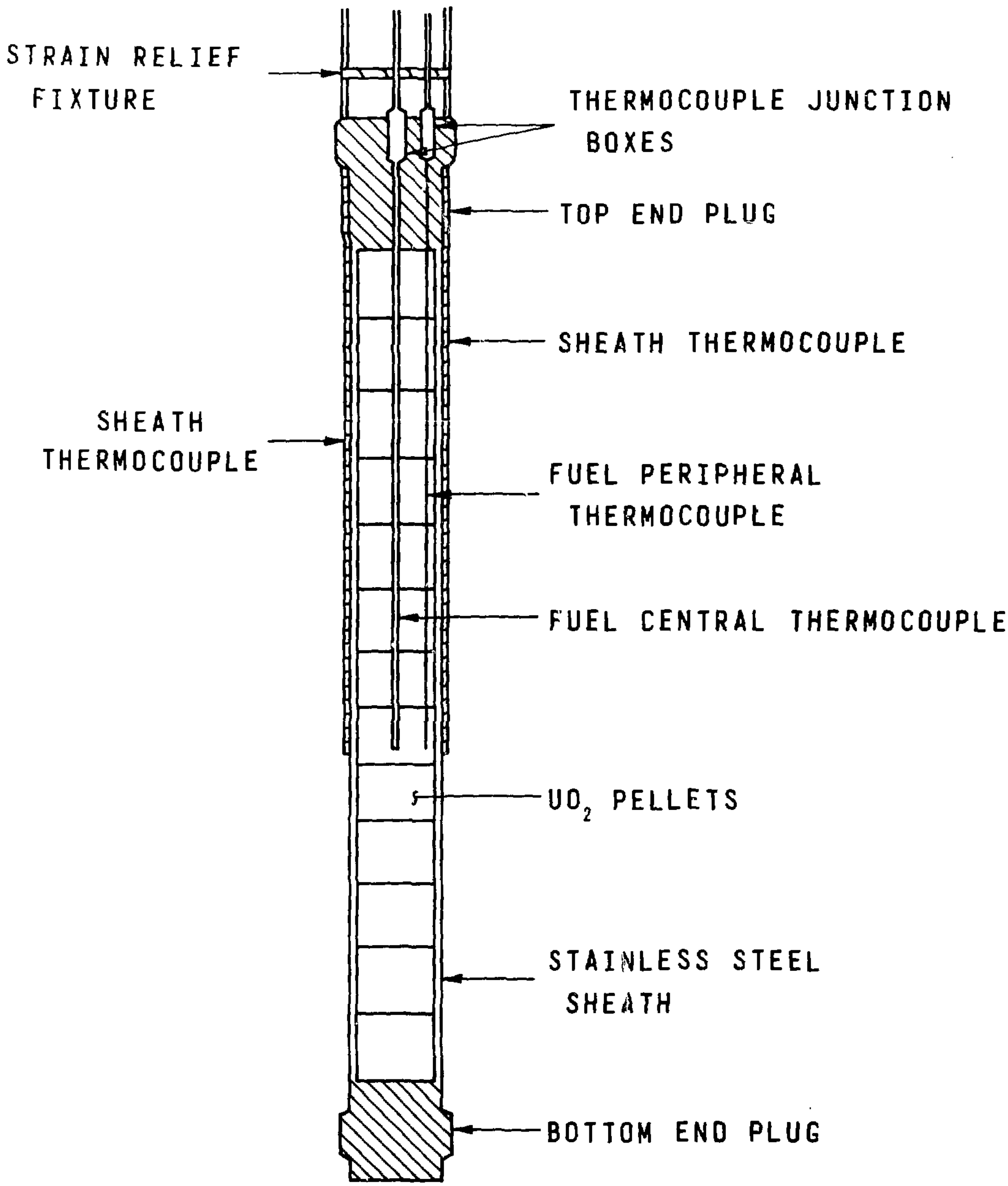

FIGURE 1

SCHEMATIC DRAWING OF A TYPICAL CRNL INSTRUMENTED FUEL ELEMENT periods of up to three months for a total dose of 7×10^{20} nvt. As it has been reported (3) that changes in thermocouple calibration occur as a result of such high doses, it was decided to carry out a study of the effects of irradiation on the thermocouple materials of interest for CRNL experiments and, if possible, to suggest methods to compensate for any changes that might take place.

The study was restricted to a literature search on the effects of radiation on thermocouple properties. Although the search was not intended to be all inclusive, each result quoted has been confirmed by at least one independent reference.

2. PROPERTIES OF THERMOCOUPLES

2.1 The Generation of Thermoelectric Voltages

Thermoelectric voltages are generated as a result of temperature gradients in any electrical conductor. The potential (dV) generated in an element of conductor supporting a temperature gradient dT is

$$dV = k_i dT,$$

where k_i is the thermoelectric (or Seebeck) constant for the material.

Thus, the thermoelectric voltage, V', generated in a wire where T_1 and T_2 are the temperatures of the ends, is

$$V' = \int_{T_1}^{T_2} k_i dT = k_i(T_2 - T_1) \qquad (1)$$

provided k_i is a constant over the length of the wire and is not temperature-dependent.

Since a potential difference cannot be measured without another conductor, a thermocouple is formed by connecting another wire of different composition and hence different k to the first. Then, the measured thermoelectric voltage V is given by

$$V = (k_1 - k_2)(T_2 - T_1)$$

and $$(T_2 - T_1) = V/(k_1 - k_2) \qquad (2)$$

where k_1 and k_2 are the thermoelectric constants for the two legs of the thermocouple.

Equation 2 shows that a thermocouple can be used to measure the temperature of the junction between the two wires, provided that:

- the cold junction temperature is known,
- k_1 and k_2 are known constants and do not vary along the lengths of the wires, and
- the wires do not change chemically or physically under the operating conditions.

However, although a thermocouple can be used to determine the temperature of its hot junction, it must be emphasized that the thermocouple voltage is generated by the temperature gradients along the wires and not at the hot junction itself.

2.2 Calibration of Thermocouples

It is usual to calibrate thermocouples by immersing the hot junction in a liquid or furnace hot zone at various known temperatures and comparing the resulting thermoelectric voltage with published data(2). However, unless the thermoelectric constant is truly uniform along the length of the thermocouple, a calibration thus determined is only valid for temperature gradients exactly the same as those existing during the calibration. A better calibration method is to slowly immerse the whole length of the thermocouple in the heated liquid while observing the thermocouple voltage. The calibration temperature gradient is thus passed along the whole length of the thermocouple and any non-uniformities in the thermocouple wires will be indicated by changes in the thermocouple output voltage. If voltage changes are within those specified for the thermocouple wires then the thermocouples may be used with confidence in temperature gradients which are not the same as those existing in the calibration apparatus.

2.3 Accuracy of Thermocouples

Thermocouple wires are usually supplied to ANSI* Standard C96.1, which gives the following best accuracies for the thermocouple alloys commonly used in nuclear experiments(4).

*ANSI: American National Standards Institute, 1430 Broadway, New York, NY 10018.

Material	Temperature Range (°C)	Accuracy
Copper-Constantan	-60 to 100	±1°C
	100 to 370	±0.375%
Chromel-Alumel	0 to 280	±1°C
	280 to 1260	±0.375%
Platinum-Platinum/Rhodium	0 to 540	±1°C
	540 to 1480	±0.25%

Tungsten-Rhenium alloys are not specified in ANSI C96.1, but the manufacturers claim errors not exceeding ±4°C or ±1%, [whichever is the greater(4)].

In situations where the cold junction is a long way from the thermocouple, and the thermocouple is made from expensive alloys, it is customary to use low cost compensating wires to connect the thermocouple to the cold junction. Compensating wires are made from special alloys having thermoelectric constants very similar to those of their corresponding thermocouple wires over a restricted temperature range, usually 0 - 250°C. These are connected to the thermocouple at a point where the temperature is below 250°C and to the cold junction. For a thermocouple/compensating wire combination, it can be shown that

$$\Delta T = (1 - A)(T_2 - T_1) \qquad (3)$$

where ΔT is the difference between the true and measured thermocouple temperatures, T_1 is the cold junction temperature, T_2 is the temperature of the compensating wire to thermocouple junction, and

$$A = \frac{(k_1' - k_2')}{(k_1 - k_2)}$$

where k_1' and k_2' are the respective thermoelectric constants of the compensating wires. Equation 3 shows that any error due to the compensating wires may be minimized by selecting the compensating wires to match the thermocouple wires as closely as possible and by minimizing the temperature gradient along the compensating wires.

In practice, the maximum errors due to compensating wires are stated to be ±1°C for copper-constantan, chromel-alumel and platinum-platinum/rhodium between 0°C and 250°C (4) and ±5°C for tungsten-rhenium compensating wires between 0°C and 450°C (5).

3. NEUTRON IRRADIATION OF THERMOCOUPLES

3.1 Irradiation Effects

When thermocouples or thermocouple materials are irradiated with neutrons in a reactor environment, four effects have been observed(3):

- gamma heating,
- structural changes in the thermocouple alloys,
- transient electrical effects, and
- transmutation effects.

All of these influence thermocouple temperature measurements, but are considered separately, since there is no published report of any interaction between them.

3.2 Gamma Heating

The gamma rays associated with nuclear fission interact with most materials to produce heat. In thermocouples heat would be produced in the vicinity of the hot junction, causing a higher temperature to be indicated.

Clearly the error produced by gamma heating will be strongly dependent on the relative thermal masses of the thermocouple and the object whose temperature is to be measured and on the heat transfer between them(3). In nuclear experiments the thermocouples must be made as small as possible to minimize disturbance of the neutron flux, and considerable effort is made to minimize thermal resistance between the thermocouples and the fuel or its sheath. In these conditions gamma heating errors of 2° to 4°C have been observed (6); these errors are small in relation to other sources of error and can safely be neglected.

3.3 Structural Changes in Thermocouple Alloys

In the nuclear fuel environment structural changes in thermoelectric alloys arise primarily from interactions between neutrons and atoms in the alloys. These cause changes in the thermoelectric voltage due to changes in the mean free path for electrons; however, calculation shows that the maximum error arising from extensively damaged material is ±2%(3). Furthermore, it is expected that this damage would be annealed out fairly rapidly at temperatures above 500°C. So, although structural radiation damage could produce errors in thermocouple voltages, in practice the errors will be small under the conditions attained in nuclear fuel experiments.

3.4 Transient Electrical Effects

The interaction of neutrons and gamma rays with conductors can produce free electrons which generate a spurious voltage in a thermocouple. This effect would be expected to change with changes in the radiation intensity.

Transient temperature measurement errors of up to ±1% have been reported for chromel-alumel and tungsten-rhenium thermocouples (7,8); however, other workers have reported errors of up to ±0.2% (9,10,11). A possible explanation for this discrepancy is that transient effects are dependent on the experimental conditions.

In any case, in nuclear fuel experiments, transient errors can be relatively easily corrected for by making measurements of temperature changes immediately following a reactor shutdown. Provided that the reactor cooling rate is relatively slow, the transient temperature error can be extracted from a plot of temperature versus time.

3.5 Transmutation Effects

Transmutation effects, produced in thermocouple materials by neutrons, result in compositional changes in thermocouple alloys. These changes, which can be quite substantial, generally cause changes in the thermocouple calibration. Errors of up to ±15% for platinum/platinum-rhodium(12,13) and ±30% for tungsten-rhenium(14,15) have been reported for neutron irradiations of 10^{21} nvt. However, chromel-alumel does not change appreciably during neutron irradiations because stable isotopes of the constituents are produced(16) and errors of only ±1% or less have been reported(6,12) [chromel-alumel can only be used for maximum temperatures of 1100°C, of course]. Since blowdown experiments can be irradiated to doses of 0.7×10^{21} nvt, temperature errors of up to ±30% can be expected for the tungsten-rhenium fuel thermocouples.

Clearly errors of this magnitude cannot be neglected and should be corrected for. However, since an irradiated thermocouple system normally consists of a section of unirradiated thermocouple joined to a section of irradiated thermocouple whose composition is changing as the experiment proceeds, an exact correction cannot be achieved.

4. CORRECTION FOR TRANSMUTATION IN THERMOCOUPLES

4.1 Errors Arising from Transmutation

An irradiated thermocouple may be considered in the same way as a thermocouple connected to compensating cable. If it is assumed that transition between the irradiated and unirradiated thermocouples is a point at temperature T_2, then

$$V' = (k_1 - k_2)(T_2 - T_1) + (k_1' - k_2')(T_3 - T_2)$$

and $$V = (k_1 - k_2)(T_3 - T_1)$$

where V' is the voltage of the irradiated/unirradiated thermocouple combination, V is the voltage of an unirradiated thermocouple in the same environment, T_1 and T_3 are the cold and hot junction temperatures, and where k_1' and k_2' are the thermocouple constants of the irradiated thermocouple and k_1 and k_2 those of the unirradiated thermocouple.

Then, the error in assuming an irradiated thermocouple is unirradiated is (in emf units)

$$\Delta V = V - V' = (k_1 - k_2)(1 - B)(T_3 - T_2)$$

and the error in temperature units is

$$\Delta T = \frac{\Delta V}{(k_1 - k_2)} = (1 - B)(T_3 - T_2) \qquad (4)$$

where

$$B = \frac{(k_1' - k_2')}{(k_1 - k_2)}$$

In this case the factor B is related to the amount of transmutation, which depends on the neutron fluence. Equation 4 shows that the observed temperature measurement error ΔT is proportional both to the transmutation as defined by B and to the temperature gradient in the irradiated thermocouple.

Furthermore, although B can be determined experimentally, the temperature gradient can be expected to change with changes in reactor power, and in different reactors. Thus it is impossible to measure an error in one experiment and expect to apply this to other experiments where the temperature gradient may be different. Nevertheless it is possible to determine B and T_2 for specific experimental conditions. Then, as stated above, since

$$V' = (k_1 - k_2)(T_2 - T_1) + B(k_1 - k_2)(T_2 - T_3),$$

$$T' - T_1 = \frac{V'}{(k_1 - k_2)} = (T_2 - T_1) + B(T_3 - T_2)$$

$$T' = T_2 + B(T_3 - T_2)$$

and

$$T_3 = \frac{T' - T_2(1 - B)}{B}$$

where T' is the temperature indicated by the transmuted thermocouple and T_3 is the temperature which needs to be measured. This method of calculation uses the factor B to correct the thermocouple constants at T' and thus compensates for any temperature dependence of the thermocouple constants.

Two possible techniques for measuring B are

- to measure the resistivity of the thermocouple wires and use this value to determine the correction factor (B) of the thermocouple, or

- to calibrate the transmuted thermocouple against one that does not transmute.

4.2 Correction By Resistivity Measurements

This technique, which was suggested for tungsten-rhenium thermocouples (13), makes use of the fact that tungsten transmutes to rhenium and that the resistivity of tungsten-rhenium alloys changes markedly with rhenium content. Thus the rhenium content of the tungsten or tungsten-rhenium alloy can be determined accurately as irradiation proceeds and the thermocouple constant derived from calibration curves of all the possible tungsten-rhenium alloys.

The method has a major advantage that the transmutation effect of each thermocouple in an experiment can be corrected for. Its disadvantages are that two extra wires must be connected to each thermocouple to permit the resistivity measurement to be made, and that a wide range of thermocouple alloys must be prepared and calibrated.

4.3 In-situ Calibration of Thermocouples

Chromel-alumel thermocouples are not much affected by transmutation effects, but cannot be used above 1100°C. However, if a chromel-alumel and a tungsten-rhenium thermocouple are both used to determine the temperature of a point in the neutron flux where the temperature is 1100°C or less, then the transmutation factor B can be determined from equation 4 if the transition point temperature T_2 is known (Figure 2). Then this calculated value of B can be used to correct the readings of other tungsten-rhenium thermocouples in the same nuclear environment. This calibration method will be valid provided that:

- the radiation environments for the low temperature thermocouples and the high temperature thermocouples are the same, and

- the factor B for the tungsten-rhenium does not vary with temperature.

The major advantage of this technique is its simplicity; the major disadvantage is that in practice it might be difficult to ensure that the high and low temperature thermocouples are in the same radiation environment.

4.4 Measurement of the Transition Temperature

In the analysis described in paragraph 4.1, it was assumed that the transition between the irradiated and unirradiated sections of the thermocouple took place at a single point along the thermocouple. In practice, this will not be the case and the amount of transmutation will vary gradually as the neutron flux changes along the length of the thermocouple (Figure 2). However, the transition point can be taken as the position along a thermocouple where the neutron flux is half its maximum value and where the thermocouple compositions are midway between those of the irradiated and unirradiated alloys, provided the transmutation falls off linearly within the transition zone. The transition point temperature can then be measured by installing a chromel-alumel thermocouple at this point.

However, if the flux distribution changes with temperature or time, the position of the transition point will not be stable and it will not be possible to determine the transition point temperature accurately. In this case, a mean position must be selected, with a resultant loss of accuracy.

Uncertainty over the position and temperature of the transition point can be overcome by the use of high temperature lead wires in the transition zone (Figure 3). The effect of this is to construct another thermocouple junction in the high neutron flux area of the reactor but at a point where the temperature is less than 800°C. Then the temperature of this junction can be measured using a non-transmuting thermocouple such as chromel-alumel. Temperature at other points may be determined using high temperature thermocouples such as tungsten-rhenium with high temperature lead wires and

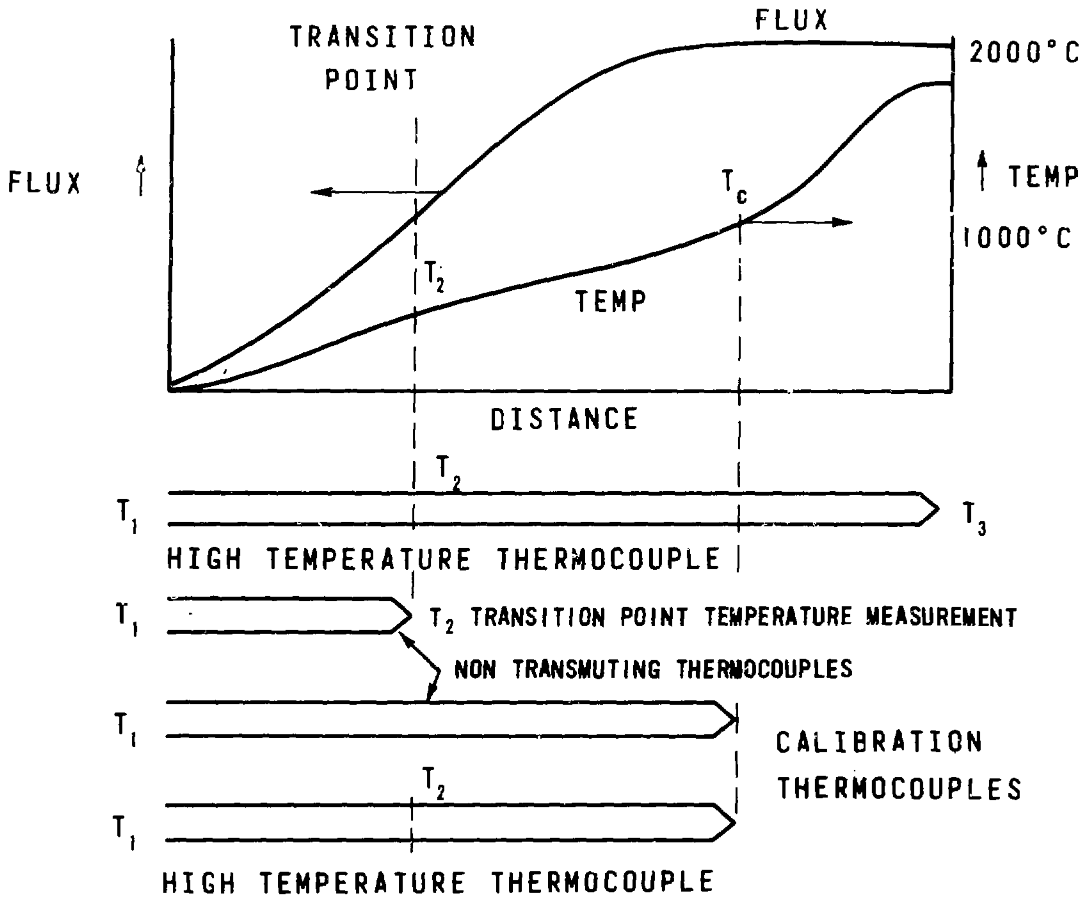

FIGURE 2

ARRANGEMENT OF THERMOCOUPLES FOR CORRECTION OF TRANSMUTATION ERRORS

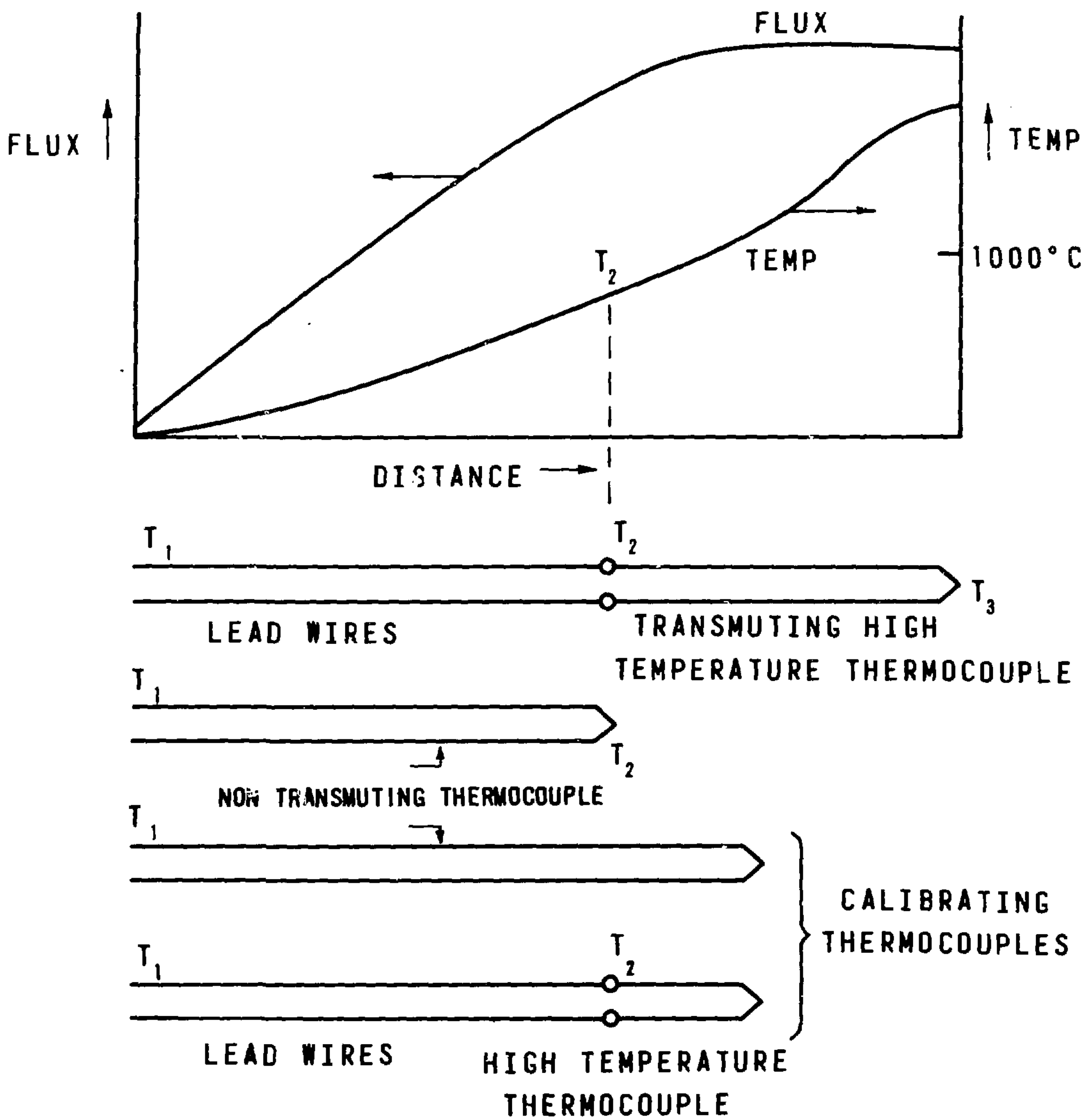

FIGURE 3

ARRANGEMENT OF COMPOUND THERMOCOUPLES FOR CORRECTION OF TRANSMUTATION ERRORS correcting for the transmutation occurring as the experiment proceeds. The lead wires can be made of chromel-alumel thermocouple cable, chromel or alumel, or any high temperature metal, but whatever material is used, it must be prepared to thermocouple standards to avoid the introduction of spurious thermoelectric voltages. Then from thermocouple theory (Figure 2):

$$V = (kL_1 - kL_2)(T_2 - T_1) + B(k_1 - k_2)(T_3 - T_2) \qquad (5)$$

where V is the net thermocouple voltage, kL_1 and kL_2 are the thermoelectric constants of the lead wires, B is the transmutation factor, and k_1 and k_2 the thermocouple constants of the high temperature thermocouple.

If the lead wires are both made of the same material, then equation (5) becomes

$$V = B(k_1 - k_2)(T_3 - T_2)$$

and the result is independent of any transmutation of the lead wires.

4.5 The Use of Compensating Cables

If thermocouple compensating cables extend into the irradiation zone, then equation (4) does not apply. In this case it can be shown that

$$\Delta T = (1 - A)(T_C - T_2) + (1 - B)(T_3 - T_2) \qquad (7)$$

where T_C is the temperature of the junction between the compensating cable and the thermocouple wires, and

$$A = \frac{(c_1' - c_2')}{(c_1 - c_2)}$$

where c_1' and c_2' are the thermocouple constants of the irradiated compensating cable and c_1 and c_2 those of the unirradiated compensating cable.

When $T_C = T_2$, equation (5) reduces to equation (4) and no error is introduced by the compensating cable. If $T_C \neq T_2$, an additional error is introduced which can only be corrected for by measuring T_C with a chromel-alumel thermocouple and by making a calibration measurement at two values of T_3 so as to allow the determination of both A and B. However, this is a rather cumbersome procedure and if compensating cable must be used, its junction should be maintained at the transition zone temperature.

5. DISCUSSION

The literature search has shown that of the four possible radiation effects on thermocouples, gamma heating and short-range order effects introduce small errors in thermocouple readings. However, electrical charge effects and transmutation of the thermocouple alloys have been shown to cause errors of between ±1 and ±30%.

Electrical charge effects are directly produced by radiation and disappear when irradiation ceases and so can be corrected for in nuclear fuel experiments by making measurements immediately before and after reactor shut-down. However, transmutation converts platinum-rhodium or tungsten-rhenium thermocouples into different alloy systems having different thermoelectric constants. Published results(12,13) show that tungsten-rhenium introduces a temperature measurement error of ±30% after a radiation dose of 10^{21} nvt. Since a two-week irradiation of a blowdown experiment is equivalent to 1×10^{20} nvt, it is apparent that the expected temperature measurement error of ±1% cannot possibly be achieved unless a correction for transmutation effects is used.

A simple analysis of thermocouples has shown that a correction for transmutation effects is possible by in-situ calibration against an untransmuted thermocouple. Since the untransmuted thermocouple cannot withstand high fuel temperatures, the calibration must be done at a lower temperature and it must be assumed that the thermoelectric coefficients measured are temperature-independent. In calibrating a transmuted thermocouple, it is necessary either to

- measure the temperature of the transition point between the transmuted and untransmuted thermocouples, or

- construct the high temperature thermocouples with high temperature lead wires to a point in the neutron flux where the temperature is about 800°C and can be measured by a chromel-alumel thermocouple.

Of these two methods, the most accurate is that of using a high temperature thermocouple with high temperature lead wires since this effectively eliminates errors introduced by temperature gradients in the neutron flux gradient or changes in the flux gradient as the experiment proceeds. However, the introduction of high temperature joints in the thermocouple could introduce reliability problems which cannot be tolerated. In this case the transition zone temperature method would have to be used.

Since the accuracy of the correction depends on the accuracy of the extra temperature measurements required and on the degree of transmutation of the high temperature thermocouples, it is impossible to calculate the potential accuracy of this method of correction for transmutation. An upper limit is set by the maximum errors of uncorrected thermocouples and a lower one by the ±1% accuracy of untransmuted tungsten-rhenium thermocouples. However, it would seem reasonable to expect that errors could be reduced to ±2% in the best case and considerably better than 30% in the worst case.

This degree of correction is only possible if compensating cable is not used. The presence of compensating alloys in the irradiation zone could introduce an additional error which is not easily corrected for. It is strongly recommended that if compensating wires must be used, they should be restricted to areas outside the irradiation zone.

It is suggested that the method of in-situ calibration (paragraph 4.3) rather than resistivity measurement (paragraph 4.2) be used to determine the degree of transmutation of the thermocouples, because the in-situ method is simple and provides a correction dependent on the actual irradiation of the experimental thermocouples.

In experiments similar to the X-2 blowdown experiment, calibration thermocouples may be installed at a point in the endcap where the temperature does not exceed 1100°C (Figure 1). Since the neutron flux falls to zero somewhere along the hanger bar which is in the coolant flow, it should not be necessary to use high temperature lead wire thermocouples since the transition point temperature is defined by the coolant temperature. However, if accurate temperature measurements are required with the X-2 loop blown down, then the transition zone temperature must be measured, or high temperature lead wire thermocouples used.

In view of the published data on the transmutation of high temperature thermocouple alloys, some workers are searching for high temperature alloys of low neutron capture cross section. A recent paper recommends the alloys niobium/10% molybdenum and molybdenum/5% niobium(17). At the time of writing these alloys are not available in wire form and an attempt is being made to obtain wires of these compositions. The successful use of these alloys would remove the need to correct for transmutation effects and would improve the accuracy of nuclear fuel experiments.

6. CONCLUSIONS

A literature search has shown that the accuracy of high temperature thermocouple measurements is seriously impaired in a nuclear fuel environment.

The most serious errors are produced by the effects of induced change and transmutation of the thermocouple alloys. Errors due to induced change effects being radiation-induced, can be corrected by making temperature measurements immediately following reactor shutdown. Transmutation errors can be corrected by calibrating high temperature thermocouples against a chromel-alumel thermocouple in a cooler part of the nuclear experiment.

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