PROGRESS IN DEVELOPMENT OF A TECHNIQUE TO MEASURE THE AXIAL THERMAL DIFFUSIVITY OF IRRADIATED REACTOR FUEL PELLETS

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ABSTRACT

A new technique, based on pulsed high-energy (≈12 MeV) electron-beam heating, is being developed for measuring the thermal diffusivity of irradiated reactor fuel. This paper reports on the continuing development work required to establish a practical technique for irradiated materials at high temperatures (1000 to 1500°C). This includes studies of the influence of thermocouple surface contact resistance, of the sheath and the pellet mounting system, of internal cracks in the pellet, and of the chamber atmosphere. Calibrations with a NIST standard and measurements on fresh UO_2 were done. Progress during the past year in these various areas is reviewed, and initial experiments with a specimen of high-burnup CANDU fuel are discussed.

1. INTRODUCTION TO THE EXPERIMENTAL TECHNIQUE

AECL has been systematically developing the following technique for measuring thermal diffusivity of irradiated reactor fuel, based on 12-MeV electron-beam pulsed heating. A cylindrical sample is mounted in a small conventional furnace and brought to a specific temperature. Then, one end of the sample is rapidly pulse-heated with an electron beam, and the transient temperature response of the other end is measured with a thermocouple. The transit time for the heat pulse yields the thermal diffusivity.

Early work concentrated on developing the PHELA electron accelerator beam delivery systems and the high-speed thermocouple temperature data collection systems. Proof-of-principle measurements were done on unirradiated unsheathed UO_2 up to 400°C [1]. An irradiated fuel transfer flask/target chamber system was completed in 1996, and hot-cell loading and transfer systems then developed. The sample mounting and heating system (Figure 1) holds the fuel slice firmly so that pressure is always applied to a spring-loaded thermocouple on the back face of the pellet, thus ensuring reproducible surface contact. To reduce conductive heat loss, a sample mounting technique, compatible with hot-cell loading, was chosen consisting of 3 axial wires for lateral confinement, 3 small axial wires for locating the back face (i.e., thermocouple tip location), and 3 axial wires against the upstream side (front face) to hold the pellet against the slight pressure of the thermocouple spring-loading. To minimize radiant cooling of the pellet during each transient temperature increase, the upstream end of the pellet holder is covered with a 25-μm-thick sheet of Ti6Al4V foil, which is heated by the heater. A thermal baffle is also mounted

on the frame behind the pellet back face. The thermal diffusivity experiment must be performed in a controlled atmosphere, usually in good vacuum (<10^{-5} Torr) to maintain the fuel stoichiometry. At the beginning of 1997, much development work remained in the path to an established practical technique for irradiated materials at high temperatures (1000 to 1500°C). The integrity of the sliced irradiated fuel pellet, and the influence of internal cracks in the pellet and of the chamber atmosphere were as yet unconsidered. The influence of thermocouple surface contact resistance, of the sheath (it seemed clear that the sheath could not be removed without destroying the integrity of the pellet), of the pellet mounting system—all needed to be studied. Finally, measurements were required on known standards at each stage of hardware and software development, to verify each development step. Progress during the past year in these various areas will be outlined in this paper.

2. CHOICE AND MODELLING OF THE THERMOCOUPLE SYSTEMS

The present sample heating furnace is limited to a maximum of 1000°C, so a type K thermocouple was used with a response time similar to the length of the electron beam macropulse (≈100 ms). The electron accelerator vault is an electrically noisy environment, so a shielded thermocouple is necessary. To minimize the heat load on the back face of the pellet, and to maximize the thermocouple-pellet response time, the smallest practical thermocouple diameter was chosen, a 0.25-mm-diameter, stainless steel, sheathed, grounded commercial unit. The response-time of the thermocouple junction was measured by inserting the thermocouple into a 2-mm-deep, 0.28-mm- diameter hole in the back face of an 8-mm-long, 13-mm-diameter OFHC copper pellet and doing a standard pulse heating experiment. The response of the copper is very fast (thermal diffusivity = $113.5*10^{-6}$ m^2/s), and a fit to the total system response suggested a junction response time, $\tau \approx 70$ ms.

When, however, the response was measured in the usual measurement configuration in which a spring-loaded thermocouple was simply touching the back face of the copper sample, a different response was obtained (Figure 2), which is not a simple exponential. To improve the modelling of the spring-loaded, surface-contacting response, a thermal model of the thermocouple was conceived, based on the assumed physical geometry and surface contact characteristics (Figure 3). The sheath is assumed to be the main heat conduction path away from the thermocouple tip, while the heat capacity is primarily that of the magnesia insulation. Clearly a distributed model for the thermocouple would be best, in principle, but it was hoped that a lumped response would be adequate. This model suggests that only the value of the contact resistance should be different for each sample, but that the values of τ_2 (the junction response time) and τ_4 (the thermocouple sheath/insulation response time) should depend only on the thermocouple used. Theoretical fits that were indistinguishable from the copper data were obtained with values of $\tau_2 \approx 0.1$ s, $\tau_3 \approx 0.3$ s, and $\tau_4 \approx 1.7$ s.

The theory of thermal contact resistance suggests that, for a given force, F_{el}, and fixed geometry, the contact area, A_{el}, between two materials (which is inversely proportional to the contact resistance) is proportional to $(F_{el}/E')^{2/3}$, assuming the applied loads are low enough to not cause plastic deformation. Here, E′ is the combined elastic modulus of the two materials. The

value of $(E')^{-2/3}$ should be proportional to the contact resistance and thus the time constant, τ_3. The values for the materials used in this work are listed in Table 1 .

The sensitivity of the experimentally determined diffusivity, α, to the value of these time constants was determined by fitting measurements on sheathed and unsheathed fresh fuel pellets with the complete time-response function, using various fixed values of the thermocouple time constants, but varying α to get a best-fit value. A variation of τ_4 between 1.35 and 2.35 seconds makes ≤2% change in the best-fit value of the thermal diffusivity, even for the worst case of relatively large values of the $\alpha \approx 2.6 * 10^{-6} m^2/s$. The sensitivity of the "best-fit" diffusivity value to τ_3 is stronger. A factor of 2 increase in τ_3 produced a 7.6% increase in the "best-fit" diffusivity value (for $\alpha = 2.5 * 10^{-6}$ m^2/s at 50°C, with an 8-mm-long pellet). For the lower diffusivity values encountered at high temperatures, this influence is proportionally lower.

3. INFLUENCE OF THE SHEATH ON THE TRANSIENT THERMAL RESPONSE

The fragile nature of irradiated fuel makes it impossible to remove the sheath from a pellet and still maintain the physical integrity of the pellet. Measurements were done on a 7.56-mm-long fresh fuel sample to see how the presence of the sheath modified the measured pellet back face response. The sheathed diffusivity values were ≈7.5% lower at room temperature and ≈6% lower at 300°C. At 500°C, the sheathed and unsheathed values differed by less than 4%, and were within systematic error.

The finite-element code ABAQUS was used to perform a simulation of the thermal transient response of the pellet and sheath, using varying values of gap conductance. At low pressure and 300°C, adequate agreement has been obtained with models using a fuel-to-sheath heat transfer coefficient, h, of ≈0.0005 W/(mm^2•K) over the surface area in nominal contact with the sheath (≈30% of circumference). In vacuum the value of h over the open-gap region (≈70% of the circumference) decreases to h ≤ 0.0002 W/(mm^2•K). The ABAQUS calculations showed that, at 100°C, the influence of the sheath should be negligible, regardless of the value of "h" (the fuel-to-sheath heat transfer coefficient), and that only at high gap conductance and elevated temperatures was an apparent increase in diffusivity of 12%, induced by the presence of the sheath.

This suggests that the lower values measured for the sheathed fuel is a result of some other systematic error in our mounting, measuring or surface contact analysis technique, and not the result of axial heat conduction by the sheath. The agreement at higher temperatures suggests that the fuel-to-sheath heat transfer coefficient has very little influence if a gap exists between the fuel and the sheath. For CANDU engineering purposes, only values above 300°C are of interest, and the sheath-induced error at these higher temperatures appears to be less than 4%.

4. VERIFICATION OF ELECTRON PULSE TECHNIQUE WITH NIST STANDARD

Verification of the present technique using known standards was difficult, primarily because the density of UO_2 is high, and for many reasons, including rate of energy loss and beam multiple-scattering effects on the energy deposition, only a high density material with a length ≥8 mm would provide a good calibration of the measurement.

The NIST standards group have not yet qualified any thermal diffusivity standards, and could only supply thermal conductivity standards. To derive thermal diffusivity values for these materials, one needs accurate specific heat measurements. The only suitable low thermal conductivity standard was NIST SRM#1461, a high nickel steel of density 8.007 ± 0.002 gm/cm^3, with thermal conductivity varying from 14 to 25 $W/(m \cdot K)$ between 25°C and 900°C. The specific heat of NIST SRM #1461 was measured at the Chalk River Laboratories (CRL) using the new modulated differential scanning calorimetery (DSC) system. The standard formula relating thermal conductivity, specific heat and density was used to determine the thermal diffusivity values of the NIST SRM#1461, with an uncertainty estimated at ±4% at ambient and up to ±7% at 300-400°C.

Two pellets of SRM#1461 were used for thermal diffusivity measurements, one 11.0 mm long and one 22.0 mm long. The beam energy was taken as 11.9 MeV, and energy loss curves were derived from the ITS Tiger code runs. The uncertainties in the thermocouple-to-surface contact thermal resistance precluded using the spring-loaded thermocouple surface-contacting technique for verifying the fundamental accuracy of the diffusivity measurement system. Instead, each sample had a 2-mm-deep, 0.325-mm-diameter hole drilled on-axis in the pellet back face, and the 0.25-mm-diameter thermocouple was inserted into this hole (a relatively snug fit, given tolerances). The data analysis presumed that the thermocouple sheath conduction term was short-circuited by inserting the thermocouple in the snug hole. The temperature versus time calculation was done in the usual manner using the exact pellet length, but the temperature was calculated at a position 1.5 mm upstream from the pellet back face, where the thermocouple junction was presumed to be located (i.e., 0.5 mm from the thermocouple tip). The optimum fit to the data for the 2 different pellet lengths yielded a mean value of thermal diffusivity, α, of $3.65 \pm 0.13 * 10^{-6}$ $m^2 \cdot s^{-1}$, with a 2 standard deviation confidence level. The expected value for the NIST 1461 thermal diffusivity at 25°C is $3.58 \pm 0.14 * 10^{-6}$ $m^2 \cdot s^{-1}$. This demonstrates very good agreement between our measurement technique and the accepted standard.

A set of normal surface-contacting spring-loaded thermocouple diffusivity measurements was done to determine the surface contact time constants for SRM#1461 (Table 2) and to give an absolute normalization point for the contact thermal conductance considerations in Section 2 (and thus tentative values to use for the UO_2 measurements). The surface contact thermal resistance clearly decreased with increasing temperature. The dominant time constant, τ_3, which determines the back face temperature rise time, decreased by a factor of two between room temperature and 315°C. As well, the ratio (τ_3/τ_4) showed a consistent decrease with increasing temperature, from ≈0.4 at room temperature to ≈0.25 at 315°C.

The surface contact resistance should have the temperature dependence implied in Table 1 if the thermocouple-pellet interface stresses remain in the elastic regime, and if surface contamination and oxidation are eliminated. However, the experimentally determined values in Table 2 exhibit a stronger temperature dependence than predicted, and thus leaves some uncertainty in selection of the values for fresh and irradiated fuel. Fortunately, this is likely to be a small effect (Section 2), especially for low conductivity materials.

5. VERIFICATION OF THE TECHNIQUE FOR UNIRRADIATED UO_2

Two short cylinders of commercially manufactured CANDU fuel (7.56 and 8.00 mm long, 12.19 mm diameter) were each inserted into pieces of CANLUB-coated sheathing (cut to matching lengths). This sheath material was specifically designated for use with this fuel, and thus had the usual small but definite (≈25 μm) gap between the fuel and the sheath. Thus the fuel inserted easily into the sheath. Three very small crimps in the outer end of each sheath held the fuel in place. The sheath was removed from the shorter sample for one measurement to determine the influence of the sheath. The fuel had been used to fabricate bundle SN#L50957C, and had a density of 10.726 ± 0.020 g/cm^3.

Sets of measurements were done on the 8.00-mm-long sheathed pellet between 96/11/26 and 97/07/03 (Figure 4). The results were all obtained under good vacuum ($\leq 10^{-5}$Torr), with gently polished thermocouple contact surfaces (to remove possible oxides). The integrity of the thermocouple spring-mount was maintained during these runs. The measured values of the axial diffusivity for the 2 different sheathed pellets agree to within 5% over the whole temperature range. However, these axial diffusivity results for the fresh commercial CANDU fuel are about 15% lower than the results of Lucuta et al. [2] and of the MATPRO [3] predictions for stoichiometric UO_2 of density 10.776 gm/cc. The slightly lower density of the pellet used in the present work is not responsible for this large difference. The measured differences between sheathed and unsheathed samples also do not account for it. This disagreement with the published data is still under study.

6. INITIAL EXPERIMENTS ON HIGH-BURNUP CANDU FUEL (BRUCE, BUNDLE J03311W, 540 MW•h/kg U)

Since the present technique is being developed with the aim of measuring irradiated fuel properties, it was considered important to try a few initial runs on a high-burnup fuel, just to identify the eventual problems to be encountered in a "worst-case" material. The Bruce J03311W fuel (540 MW•h/kg U) is much studied and fairly well documented [4,5], and it appeared that, up to 900°C, only a small amount of radioactive krypton would be released from the fuel, making it suitable for an initial test of the irradiated fuel measurement system in the PHELA facility.

The values of the thermal diffusivity, α measured under high vacuum on 3 different low-to-high-temperature runs on the same sample, are shown in Fig. 5. These values are much lower than expected, and increase with temperature (opposite to the expected dependence - Ref.[2,3]).

This dependence was assumed to be due to cracks whose effect diminishes with temperature because of radiant transfer of heat across a gap. At room temperature the measured diffusivity value was $\approx 0.07 * 10^{-6}$ m^2/s, a factor of $\approx$30 lower than expected. However at 960°C (1235K) the measured value was $\alpha \approx 0.6 * 10^{-6}$ m^2/s, only slightly lower than the values measured by the JAERI group [6] on HBWR high-burnup (~1500 MW•h/kg U) fuel .

The influence of backfilling the chamber to various partial pressures of helium is also shown in Figure 5. At room temperature, filling the target chamber with helium at 760 Torr increased the "effective" value of the determined diffusivity to $\approx 0.65 * 10^{-6}$ m^2/s, and lowering the helium pressure to 1 Torr decreased this value by a factor of 3. At 600°C, the influence of 10 Torr helium was reduced to a 30% effect on the measured "effective" diffusivity. This suggested the sample must have transverse cracks of significant size and number; this was borne out by the metallographic examination, which showed a large transverse void and high porosity in the central high-temperature region of the fuel.

7. DISCUSSION OF RESULTS AND SUGGESTIONS FOR FUTURE DEVELOPMENT

The primary goal of the present development work is to do axial diffusivity measurements on sheathed irradiated CANDU fuel. However, the developement process required measurements on a known standard and on fresh fuel. The high-energy electron-accelerator-based system has demonstrated quite accurate results on a NIST metal thermal conductivity standard (SRM 1461). However, the results for a set of commercially manufactured, fresh, unsheathed CANDU fuel pellets are 20% lower than the accepted values for stoichiometric UO_2 of the same density. Experiments are in progress to carefully characterize this specific fuel (density, stoichiometry), and to determine the reason for the discrepancy.

The presence of the sheath on a fresh fuel pellet was shown to decrease the measured apparent axial thermal diffusivity by an amount that decreased with temperature ($\leq$7.5% at RT, $\leq$4% at 500°C). This trend is opposite to that suggested by the ABAQUS simulations, and thus should be investigated further. However, the influence of the sheath, at least for fresh fuel-sheath-to-fuel-gap conditions, is minimal ($\leq$4%) over the range of CANDU interest (i.e., above 350°C).

Axial diffusivity measurements on sheathed pellets of high-burnup (540 MW•h/kg U) Bruce fuel (J03311W) demonstrated the strong influence of cracks and macroscopic porosity in reducing the determined effective diffusivity, especially at low temperatures. The influence of the cracks decreases as the temperature increases, and at high temperatures agreement with the laser-flash values is reasonable (The laser-flash technique generally uses small (1 to 2 mm thick) samples with no macroscopic cracks). Backfilling the target chamber with helium to allow thermal conduction across the cracks has been shown to dramatically increase the "effective" diffusivity. However, the question of whether useful quantitative numbers can be extracted from the measured effective axial diffusivities remains open. Another set of measurements on a standard burnup (220 MW•h/kg U) Darlington fuel (Q27022C), will be done to see if the influence of cracks is still dominant. A set of experiments on fresh fuel with a known axial crack, under different gases and pressures, may be useful.

The measurement system produced good results for the NIST metal standard and for fresh metal fuels, and thus should be applicable to irradiated metal fuels and in general to any uncracked fuel, fresh or irradiated. The rapid delivery of a heat pulse deep into relatively large bulk samples, makes this a unique analytical technique that may find other applications in the study of reactor materials.

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TABLE 1

THE COMBINED ELASTIC MODULUS E', AND THE CONTACT RESISTANCE RATIOS FOR THE THREE COMBINATIONS OF MATERIALS

MATERIALS	E' (25°C)	E' (327°C)	E' (500°C)	CONTACT RESISTANCE RATIO	
				25°C	327°C
Copper - Stainless Steel	81 GPa			1.2	
NIST 1461 - Stainless Steel	107 GPa	93 GPa	78 GPa	1	0.91
Polycrystalline UO_2 - Stainless Steel	107 GPa	100 GPa	89 GPa	1	0.96

TABLE 2

RESULTS OF FITTING SURFACE CONTACT TIME CONSTANTS OF 0.25 MM SHEATHED OMEGA TYPE-K THERMOCOUPLES IN CONTACT WITH NIST SRM#1461 (STAINLESS STEEL)

T (°C)	τ_3(s)	τ_3/τ_4	τ_4
30	0.75 ± 0.25	0.4 ± 0.1	1.88
315	0.40 ± 0.10	0.25 ± 0.1	1.6
505	0.30 ± 0.10	0.3 ± 0.15	1.0
709	0.05 ± 0.05	0.3 ± 0.15	0.2

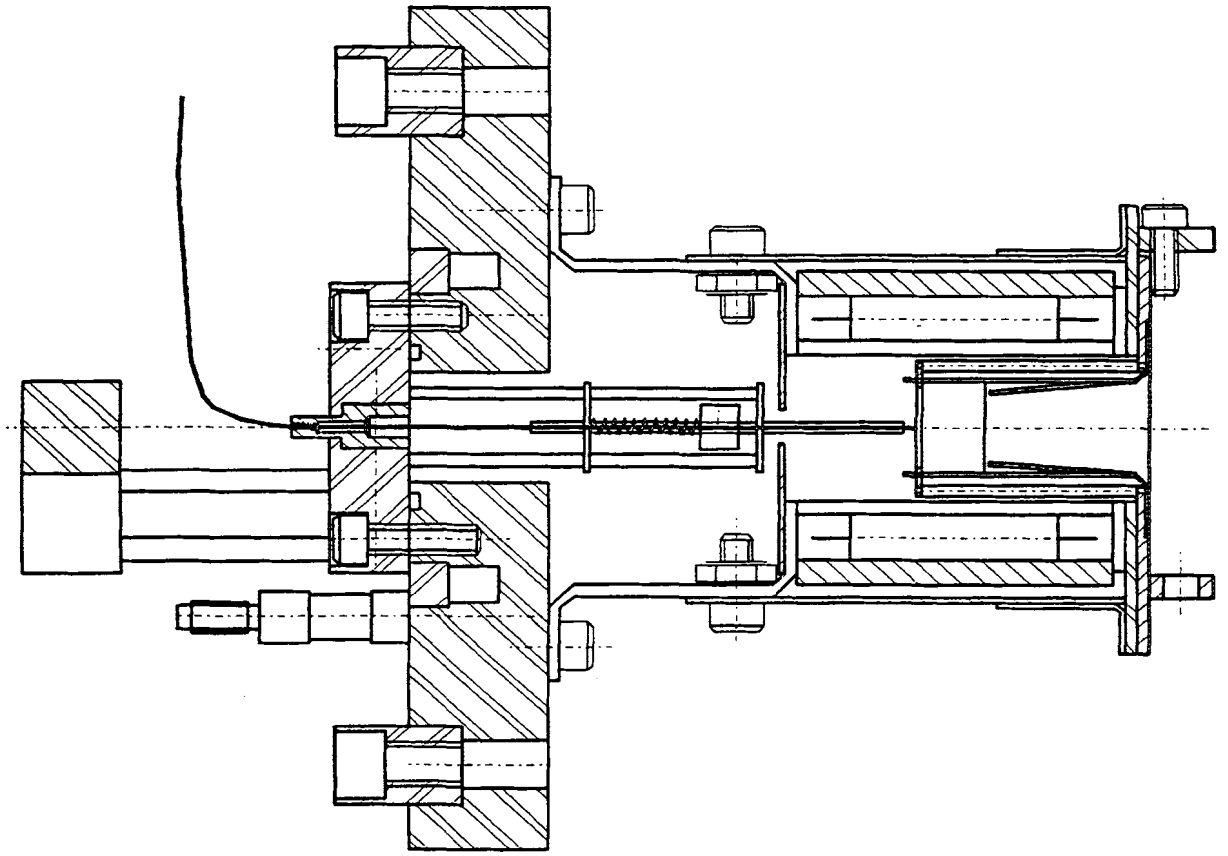


Figure 1 Schematic cross section of the pellet suspension basket, the thermocouple mounting system, and the conventional heater around the pellet. Two 0.25-mm-diameter, sheathed type K thermocouples are used, one touching the back face of the pellet, and one with a ~ 1-mm gap to to the pellet back face. The electron beam is incident from the right.

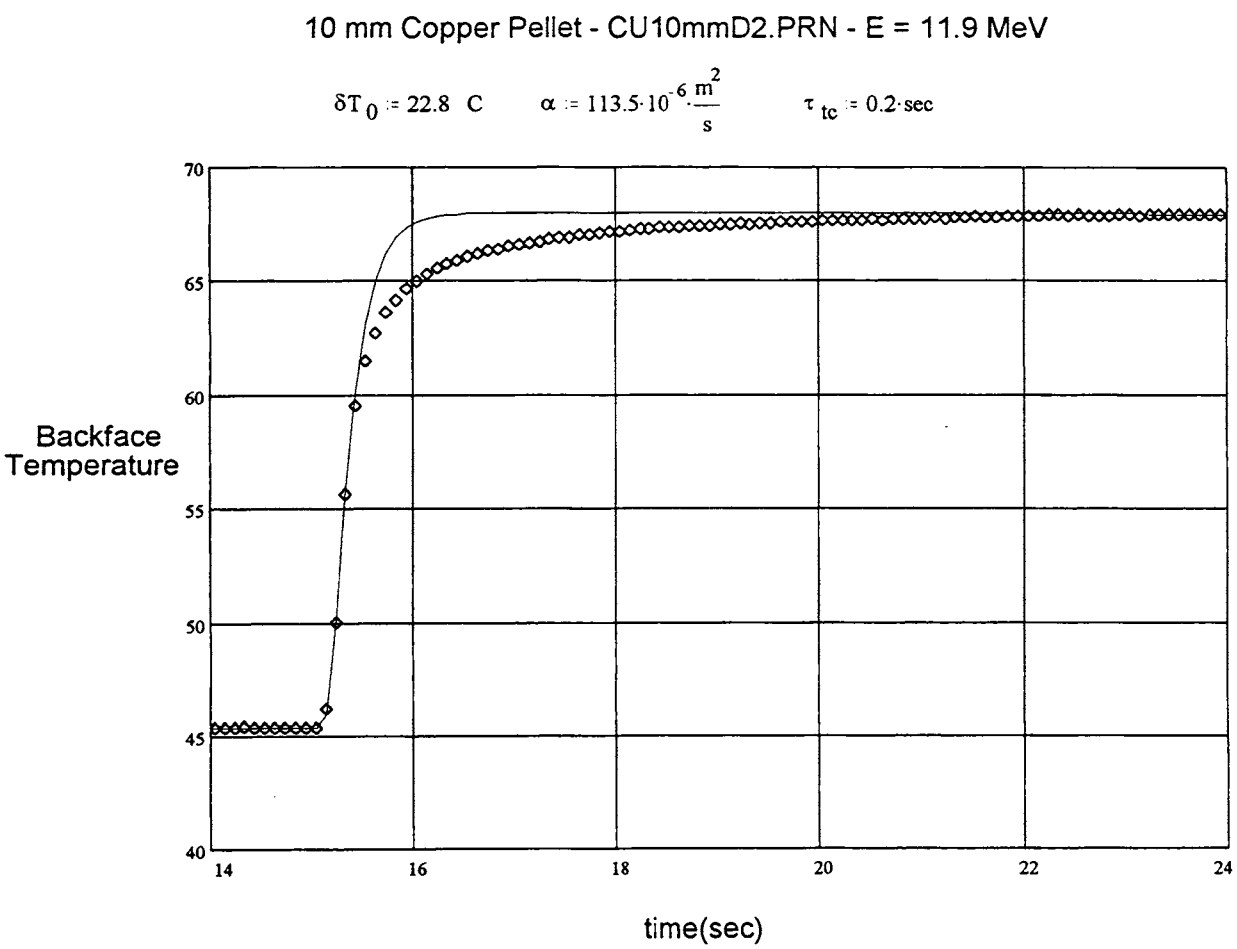
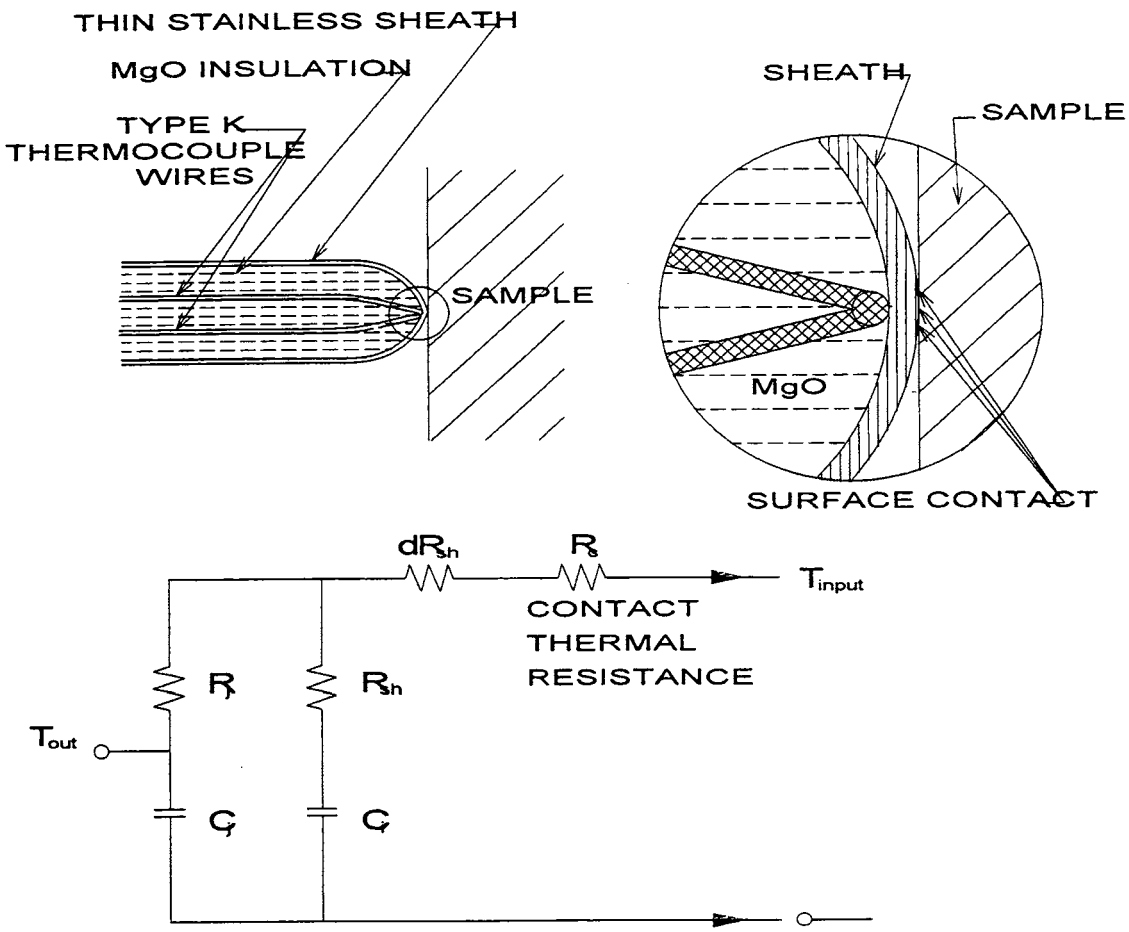


Figure 2 Transient temperature on the backface of a 10-mm-long copper sample, measured using a spring-loaded ,sheathed, 0.25-mm-diameter, type K thermocouple. The solid line is the expected response of the pellet backface,folded with a single time-constant thermocouple response .



R c = Contact thermal resistance
R sh = Equivalent sheath thermal resistance
dR sh = Resistance of length of sheath between junction and contact point of sheath with sample
C j = Junction heat capacity
R j = Junction to sheath contact resistance
C i = Equivalent heat capacity of magnesia insulation and sheath

$t_2 = R_j C_j$ $t_3 = (R_c + dR_{sh}) C_i$ $t_4 = R_{sh} C_i$

Figure 3 Schematic view of the thermocouple surface contact and time-response model, for a sheathed, MgO-loaded thermocouple. A simplified "electrical equivalent circuit" schematic is shown, suggesting the origins of the response terms.

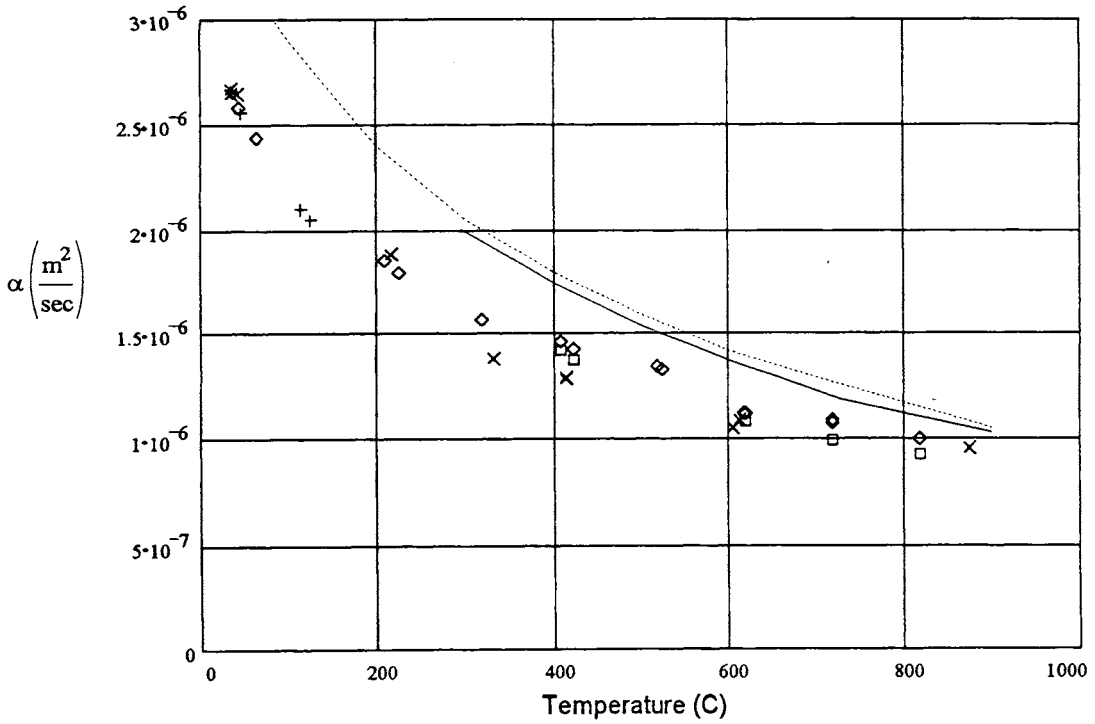


Figure 4 A comparison of axial thermal diffusivity values measured over a period of six months using the pulsed electron beam technique on the same 8.00 mm long pellet of fresh sheathed Bruce fuel (Zircaloy 4 sheath with CANLUB). The dotted line represents the measurements of Lucuta et al. on 98.3 % density stoichiometric fresh UO2 , the solid line MATPRO values for 98.0 % UO2.

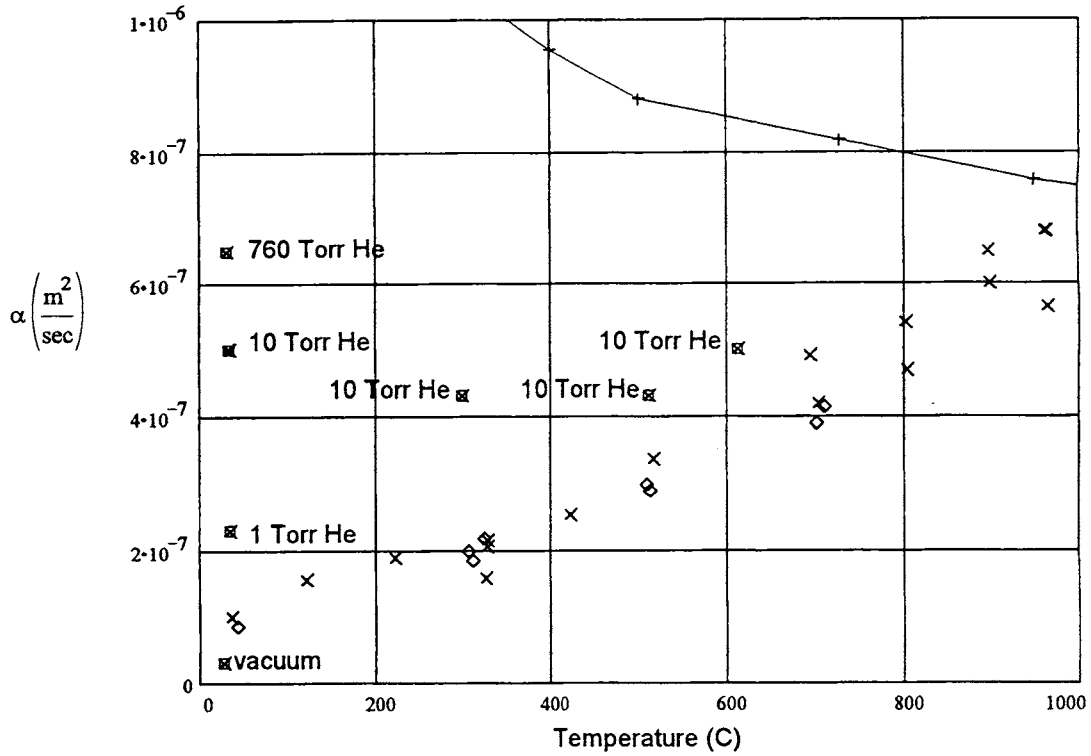


Figure 5 Measured values of the "effective" axial thermal diffusivity of the J03311W, Element #3, Sect.#3 sample. Runs were done on three different days . The marked squares are measurments showing the influence of backfilling the chamber to various pressures of helium. The line with crosses represents the JAERI measurements on 1500 MWh/KgU HBWR fuel .