TOTAL EMISSIVITY OF ZIRCALOY-4 AT HIGH TEMPERATURES

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Abstract

*For the assessment of fuel behaviour during reactor accidents involving high temperatures, a knowledge of the emissivity of the fuel sheath is required. Current data on the emissivity of Zircaloy-4 fuel sheaths are limited to 1423 K only. This paper describes the development of a technique and the experiments conducted to determine the total emissivity of Zircaloy-4 *CANDU® (CANada Deuterium Uranium) fuel sheaths in argon and in steam for up to 120 s at temperatures ranging from 1423 K to 1973 K. The results show that in this temperature range, the total emissivity of an unoxidized, metallic bright fuel sheath in argon remains constant at a value of 0.20 compared with a constant value of 0.82 for a steam-oxidized, black sheath for oxidation time up to 120 s. The current data are in good agreement with reported measurements available up to 1423 K.*

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1. INTRODUCTION

The assessment of transient fuel behaviour following a loss-of-coolant accident coincident with the loss of emergency coolant injection is required for the safety analysis of CANDU reactors. The temperature of the fuel is expected to increase rapidly during such accidents. Because the radiative heat-transfer mechanism is important at these high temperatures, the total emissivity of the fuel sheath, Zircaloy-4, is required to calculate the heat transfer from the fuel to the pressure tube. A low fuel sheath emissivity would result in a high fuel temperature.

The total emissivity of zirconium alloys (Zircaloy-2 and Zr-2.5 Nb), measured in air in the temperature range 373 K to 673 K, showed that the emissivity of the unoxidized alloys increased from ~0.16 to ~0.6 when oxidized to produce a 2 µm thick oxide layer [1]. The effect of temperature on emissivity over that temperature range was negligible. The spectral and total emissivity of unoxidized and oxidized Zircaloy-4, measured at higher temperatures using the "hole-in-tube" method, had also been reported [2]. The spectral emissivity at a wavelength of λ=0.65 µm was ~0.65 and ~0.45 for unoxidized Zircaloy-4 and ~0.88 and ~0.73 for oxidized Zircaloy-4 at 1123 K and 1623 K respectively. However, the total emissivity of unoxidized Zircaloy-4 was reported to be constant at ~0.25 over the temperature range from 1173 K to 1573 K whereas, that of oxidized Zircaloy-4 was shown to increase from ~0.7 to ~0.78 over the temperature range from 1123 K to 1423 K.

A study was conducted in our laboratories to determine the spectral emissivity of CANDU fuel sheath at wavelengths of 1.0 and 2.3 µm [3]. The spectral emissivity of unoxidized fuel sheath decreased from a value of ~0.5 at 1673 K to ~0.2 at 1923 K at a wavelength of λ=1.0 µm. At a wavelength of λ=2.3 µm the emissivity of unoxidized fuel sheath decreased from ~0.32 at 1373 K to ~0.25 at 1923 K. Our study also confirmed that the spectral emissivity of oxidized fuel sheath was higher than that of unoxidized sheath. At λ=1.0 µm the emissivity was ~0.9 at 1573 K and decreased to ~0.8 at 1923 K. At λ=2.3 µm the emissivity of oxidized sheath remained constant at ~0.85 over the temperature range 1373 K to 1923 K. Because the spectral emissivity of oxidized fuel sheath at those high temperatures at both wavelengths was about the same (in the range from 0.8 to 0.9) the oxidized fuel sheath was expected to behave like a greybody.

The objective of this paper is to present the development work and the results of a study conducted to determine the total emissivity of a CANDU fuel sheath at high temperatures in non-oxidizing argon and oxidizing steam atmospheres for up to 120 s. The selected temperature was in the range from 1423 K to 1973 K. It should be remarked that no data are available in the literature for temperatures above 1423 K.

2. THEORETICAL CONSIDERATIONS

2.1 General Principles

The intensity of thermal radiation from a real surface or object, I, is always less than the thermal radiation from a blackbody. Total emissivity is defined as the ratio of the actual intensity from a real surface to the blackbody intensity, I^{BB}, at that temperature:

$$\varepsilon_t = \frac{I}{I^{BB}} \tag{1}$$

where ε_t = total emissivity of object at temperature, T.

For practical reasons, it is not possible to measure radiation at all wavelengths. Instead, a suitable range of wave lengths is generally chosen. The apparatus built for our studies was capable of measuring radiation at wavelengths ranging from 0.2 to 9.45 µm because a window and a lens made of calcium fluoride were placed between the object and the detector. Calcium fluoride has high transmittance in this range of wavelengths. A numerical integration of Planck's law from 0.2 to 9.45 µm for the temperature range 1400 to 2300 K yields the following relationship for a blackbody:

$$I^{BB} = \sigma^* T^{4.06} \quad (2)$$

where $\sigma^* = 3.463 \times 10^{-8}\ W \cdot m^{-2} \cdot K^{-4.06}$.

In this study, a thermopile detector was used for the measurement of radiation. The principles for its use are described and the relevant equations are given in the next section.

2.2 Principles for Using a Thermopile Detector

The radiant energy received by a thermopile detector not only is a function of the measured specimen temperature and the optical properties of its surface, but is also dependent on the energy reflected from the background.

The voltage generated by the thermopile detector is proportional to the net energy absorbed by the detector [4]:

$$V_d = \varepsilon_t R(\phi_s - \phi_b) \quad (3)$$

where R is the coefficient of response of the detector.

For a blackbody $\varepsilon_t = 1$; therefore, the voltage generated from a blackbody is:

$$V_d^{BB} = R(\phi_s - \phi_b). \quad (4)$$

From Equations (3) and (4) the total emissivity of the object is:

$$\varepsilon_t = \frac{V_d}{V_d^{BB}}. \quad (5)$$

If the detector is at the ambient temperature, T_a , and the true temperature of the object is T, using Equation (2), Equation (4) can be written as:

$$V_d^{BB} = R(\phi_s - \phi_b) = D(T^{4.06} - T_a^{4.06}), \quad (6)$$

where D is the detector system constant [4].

$$D = \frac{R\sigma^* A_t A_d}{\pi d^2}, \quad (7)$$

A_t = area of target in field of view
A_d = active area of the detector
d = distance between the object and the detector.

The detector system constant can be obtained from the experiment as will be discussed under Section 5.0.

Thus, with Equation (5) total emissivity of the object can be expressed as:

$$\varepsilon_t = \frac{V_d}{D(T^{4.06} - T_d^{4.06})} \qquad (8)$$

Equation (8) was used in this study to determine the total emissivity of Zircaloy-4. Equation (6), the theoretical relationship between the detector voltage and the blackbody temperature, was verified by the blackbody tests described in this paper.

3. EXPERIMENTAL APPARATUS

An experimental apparatus was designed and built at AECL Whiteshell Laboratories to measure the total emissivity of a CANDU fuel sheath. The underlying principle was to measure the radiant energy over a selected range of wavelengths from a fuel sheath and a blackbody, both held at the same temperature, using a thermopile detector. The ratio of the two values is equal to the total emissivity of the sheath as given by Equation (1). The fuel sheath tests were conducted in argon and in steam atmospheres, whereas the blackbody tests were conducted only in argon.

3.1 General Description of the Apparatus

A schematic diagram of the general layout of the test facility is shown in Figure 1. The sample, either a Zircaloy sheath or a blackbody (1), is contained inside a sealed quartz tube (3). The Zircaloy sheath sample ~15.3 mm O.D., ~14.5 mm I.D. and ~80 mm long) is heated by radiation from a graphite heater placed inside. The heater is powered by the power supply (11) and is maintained at the desired temperature by the controller (9) (see Figure 1).

For the blackbody measurements, the Zircaloy sheath sample is replaced by a blackbody made of spectral graphite having a cavity in the mid-section, 4.8 mm in diameter and 15.9 mm deep. The cross section of the graphite on both sides of the cavity is reduced to minimize the heat loss through the ends and thus maintain constant temperature inside the blackbody cavity.

During the tests, the true surface temperature of the sheath specimen (or the graphite cavity) was measured by a two-wavelength pyrometer (8) (see Figure 1). A steam generator (13) and superheater (12) produced steam at a temperature of about 1023 K. The flow rate of steam (0.08 g/s) was controlled by regulating the flow of water into the steam generator by a peristaltic pump (15). If an argon atmosphere was required inside the test chamber, the steam flow was shut off and superheated argon could be admitted to the test chamber. The test chamber can be sealed off from the outside atmosphere and evacuated by a rotary pump. This feature enables excellent control of the test chamber atmosphere.

The radiant energy, emitted by the specimen (or the blackbody), is channelled through a tube (4) and an optical system to the thermopile detector (5). The detector signal is amplified by the unit (7). The detector and pyrometer outputs are digitized by the voltmeter (18) so that they can be displayed and stored by the computer (19).

3.2 Selection of Optical and System Components

A Thermopile detector (Model 2M, Dexter Research Center Inc., Dexter, Michigan) was selected for this study. As mentioned under Section 2.2, the thermopile detectors produce a voltage proportional to the net radiant energy incident on the detector. The Dexter detector is fabricated from the dissimilar metals bismuth and antimony, forming a thermocouple. These are connected in series, so that their outputs add. Half the junctions are called "hot" and make up the active elements, and alternate junctions, the "cold" ones, are thermally bonded to a substrate. The detector casing was thermally bonded to an aluminum block to ensure that the detector remained at the ambient temperature. The thermopile detector is a voltage generating device and therefore it requires no bias voltage or current for operation. The spectral sensitivity of the detector depends on the choice of the window material. In this study the thermopile detector had a KBr window, which will transmit about 92% of the radiant energy up to a 20-μm wavelength. The physical characteristics and operating conditions of the detector are given in Table 1.

The geometry of the optics must allow the radiant energy from a defined surface area of the source to fall on the detector. The surface area must be equal for both the blackbody and the sheath specimens because the ratio of the intensities give the total emissivity, as defined in Equation (1). The geometry of the optics must therefore ensure that the entire image of the surface area of interest is focused on the detector surface. Since calcium fluoride was used as the window material of the test chamber and the lens, the transmission properties of this material are important in the final selection of the geometry of the optics. As mentioned under Section 2.1, calcium fluoride has high transmission between 0.2 to 9.45 µm. An optical ray tracing program (BEAM FOUR, Stellar Software, Version 4.42, Berkeley, California, U.S.A.) was used to determine the optimum dimensions of the geometry of the optical system, such as the size of the aperture in front of the sighting tube and the lens, as well as the distances between the specimen, the detector and the lens. The ray tracing program has a Monte Carlo random ray generator, that calculates the path a ray follows through a specified layout of lenses, apertures and detectors, and determines the distribution of the incidence of rays on the detector surface.

The ray distribution was calculated for various geometries. For the construction of the apparatus the following dimensions were optimized from a series of calculations:

Diameter of the aperture in front of the lens		=	3 mm
Distance between the object and detector		=	560 mm
Distance between the lens and the detector		=	60 mm
Detector area		=	2 mm x 2 mm
Radius of lens	R_1	=	20 mm
	and R_2	=	0 mm.

The observed, similar Gaussian distribution of the rays indicated that the detector surface would capture all the rays between 0.2 and 9.45 µm

4. EXPERIMENTAL PROCEDURE

To conduct a test the appropriate sample (either Zircaloy fuel sheath or blackbody) was assembled with the heater (see Figure 1). The quartz containment vessel (see (3) in Figure 1), was placed around the sample and adjusted so that the CaF_2 window exposed the sample to the detector and pyrometer. The cooling water for the copper conductors was turned on, and the radiometer amplifier was zeroed. The test chamber was evacuated, filled with argon, and a small flow rate (170 cm^3/s) of argon was maintained. The heater power supply was turned on and the sample was heated to the desired temperature.

4.1 Blackbody Tests

During blackbody tests, the argon was kept flowing through the containment vessel and the blackbody temperature was raised to 1373 K and maintained. The temperature was measured using the two-wavelength pyrometer focused on the inner surface of the blackbody cavity, and the detector voltage was measured. The measurements were repeated at 50 K intervals from 1423 K to 1973 K.

4.2 Sheath Oxidation Tests

For sheath oxidation tests, the sample temperature was raised to 1373 K and maintained in argon while the detector voltage and the sheath temperature were measured. After maintaining the temperature constant, (for about 50 s), the argon valve was shut off and steam (0.08 g/s) was allowed to enter the containment vessel. The power input to the graphite heater was controlled to maintain the initial temperature. After about 120 s of steam flow, the power and the steam flow were shut off, and the specimen was allowed to cool to room temperature. The measurements were repeated at 50 K intervals from 1423 K to 1973 K.

For selected samples, the part of the sheath exposed to the detector was sectioned and examined with an optical microscope.

5. RESULTS AND DISCUSSION

The detector signal in mV from the blackbody tests was plotted against $[T^{4.06} - T_a^{4.06}]$, where T and T_a are the absolute temperature of the blackbody and the ambient temperature respectively. The results are shown in Figure 2. The measurements fall on a straight line as expected (see Section 2.2, Equation 6). The following equation was obtained by linear regression:

$$V_d^{BB} = 2.82 \times 10^{-14} (T^{4.06} - T_a^{4.06}) \tag{9}$$

The detector system constant defined by Equation (7) is therefore given by:

$D = 2.82 \times 10^{-14}\ \mathrm{mV/K^{4.06}}$.

The operating conditions for the thermopile detector specify that the maximum incidence on the detector should not exceed 10^3 W/m^2 (see Table 1). The maximum incidence on the detector, I^{BB}, is derived from Equations (6) and (7):

$$I^{BB} = \frac{\Phi_s - \Phi_b}{A_d} = \frac{\sigma * A_t}{\pi d^2} \left[T^{4.06} - T_a^{4.06}\right] \tag{10}$$

With $A_t = 7.068 \times 10^{-6}$ m^2, d = 0.56 m for the experimental set up and for the maximum test temperature, T = 1973 K and for the ambient temperature T_a = 293 K, we get I^{BB} = 5.93 W/m^2. Therefore the operating conditions do not exceed the specified maximum incidence for the detector.

Equation (9) was programmed into the computer so that for a sheath specimen temperature, T, the expected blackbody voltage could be calculated. With the measured detector voltage, V_d for the sheath specimen the oxidized sheath emissivity could then be computed directly using Equation (8) and displayed as a function of time.

Results of a typical steam oxidation test conducted at 1973 K are shown in Figure 3. In this test, the fuel sheath was held in argon for about 50 s and then steam was turned on. The total emissivity increased rapidly from a value of about 0.2 to about 0.82 during steam oxidation. The sheath temperature of the specimen was maintained at about 1973 K for about 170 s; the test was then terminated. Sheath specimens removed from argon atmospheres were bright metallic compared with black ones after steam oxidation tests.

From the results of various tests, the steady-state emissivity is plotted against the temperatures for oxidized (closed circles) and unoxidized fuel sheath (open circles) in Figure 4, and compared with available data from the literature [2]. This study shows that, in the temperature range from 1423 K (1150°C) to 1973 K (1700°C), the total emissivity of unoxidized Zircaloy-4 remains constant at a value of 0.20 compared with 0.82 for steam-oxidized Zircaloy-4. The steady-state emissivity for the steam oxidation test is the value measured after 120 s of steam exposure. The thicknesses measured from metallographic examination indicated growth of several tens of microns of oxide. The measured steady-state emissivity is found to be in good agreement with the data on the spectral emissivity of Zircaloy-4 measured at wavelengths of 1.0 and 2.3 μm [3].

6. CONCLUSIONS

The experimental method developed here was used to determine the total emissivity of a Zircaloy fuel sheath at high temperatures. In the temperature range from 1423 K to 1973 K the total emissivity of unoxidized fuel sheath is constant at a value of about 0.2 and the specimen surface has a bright metallic shine. Under the experimental conditions, the surface oxide formed in steam is black and the emissivity of the oxidized sheath increased to a constant value of ~0.82.

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TABLE 1

PHYSICAL CHARACTERISTICS AND OPERATING CONDITIONS OF THERMOPILE DETECTOR, MODEL 2M, DEXTER RESEARCH CENTER

Physical Characteristics	Operating Conditions
Number of Junctions: 48	Temperature Range: 208 to 358 K
Sensitive Area: 2 mm x 2 mm	Maximum Incidence: 10^3 W/m^2
Window Material: KBr	Spectral Response: flat from UV to far IR
Encapsulating Gas: Ar	Signal Output: linear from 10^{-2} to 10^3 W/m^2

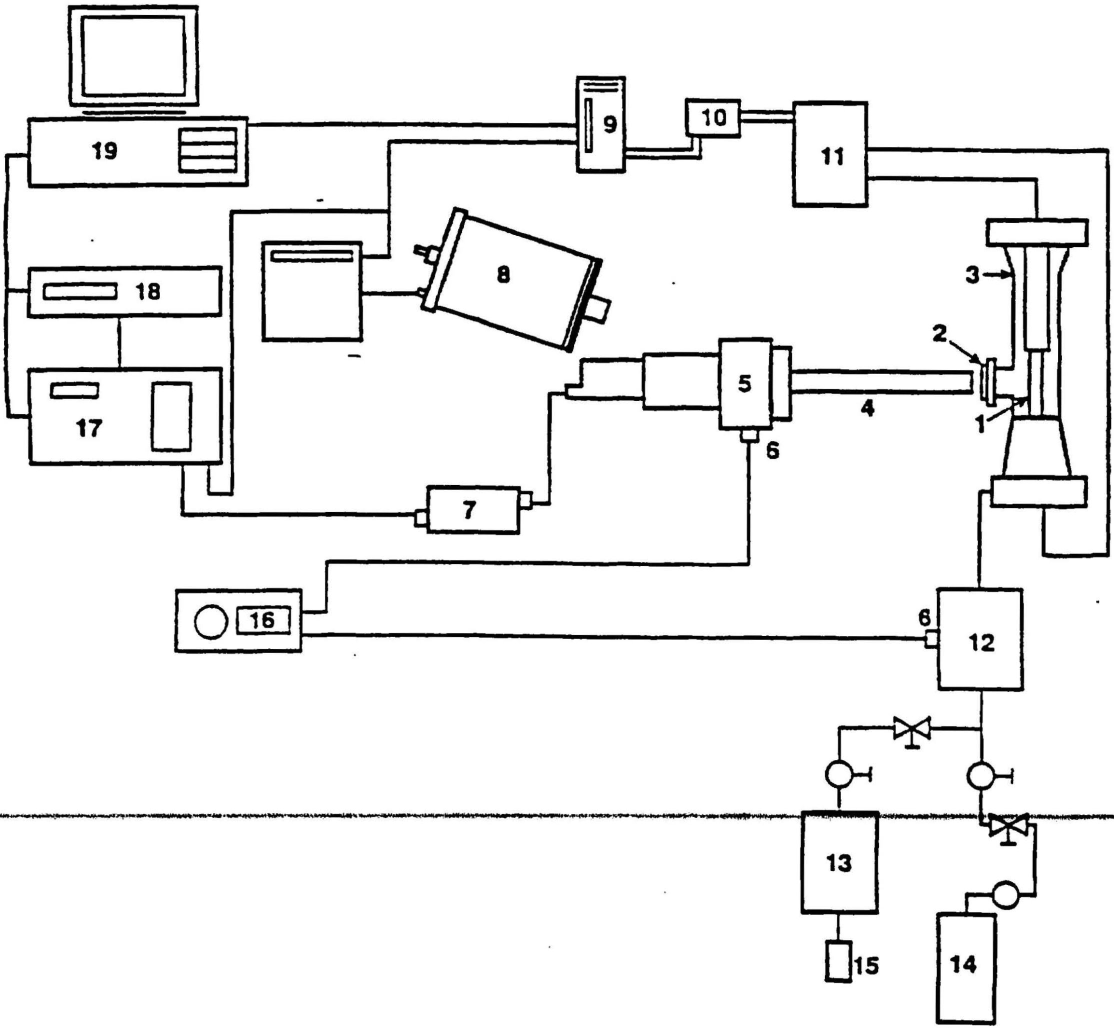
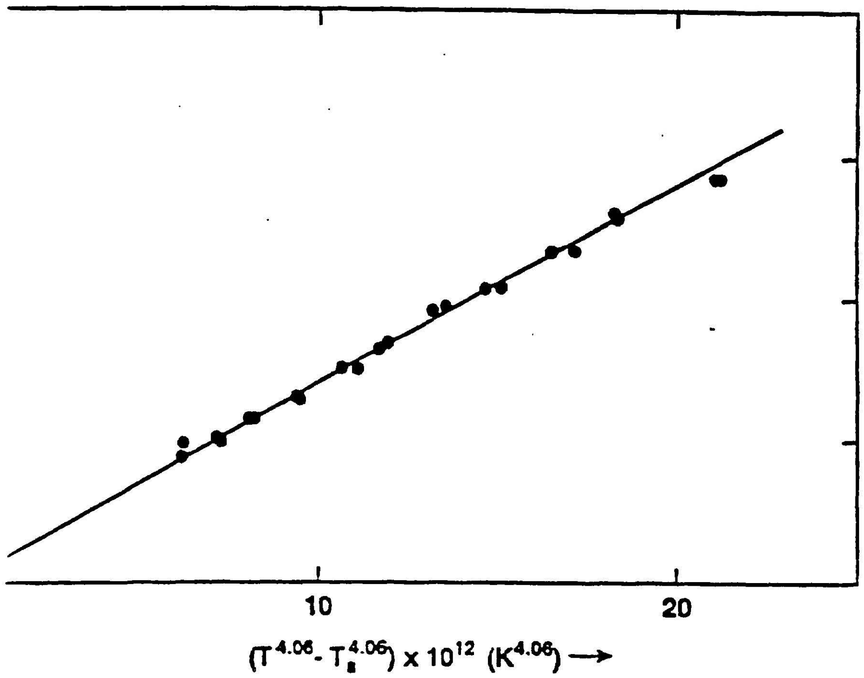
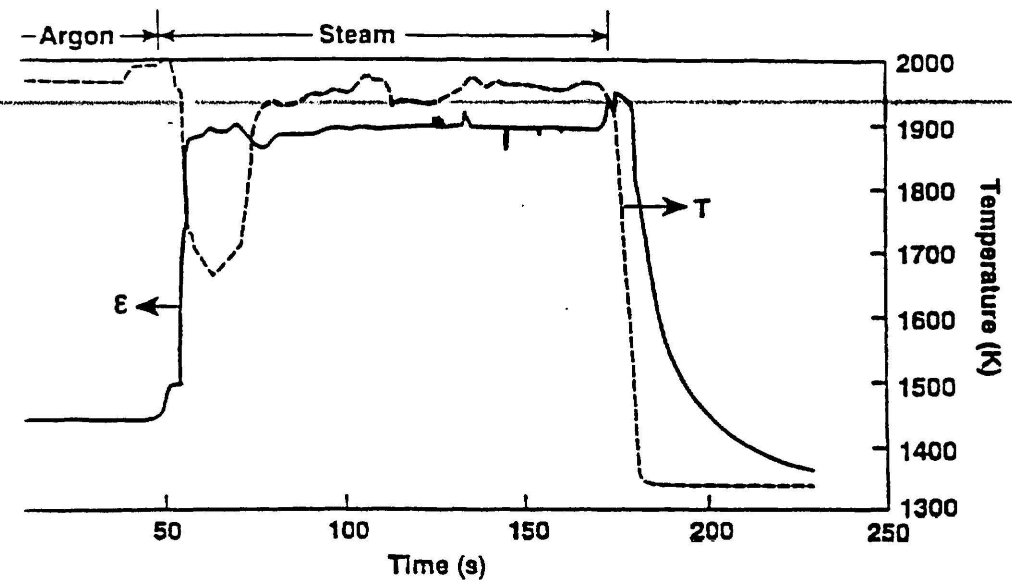


FIGURE 1: GENERAL LAYOUT OF THE TEST FACILITY

(1 = sample, 2 = CaF_2 window, 3 = Quartz containment vessel, 4 = sighting tube, 5 = thermopile detector, 6 = thermocouples, 7 = amplifier, 8 = two-wavelength pyrometer, 9 = P.I.D. process controller, 10 = SCR power controller, 11 = 100 KVA power supply, 12 = steam superheater, 13 = steam generator, 14 = argon buffer, 15 = peristaltic pump, 16 = thermocouple display, 17 = scanner, 18 = digital voltmeter, 19 = computer)



.ATIONSHIP BETWEEN THE DETECTOR OUTPUT AND THE ABSOLUTE
.PERATURE OF THE BLACKBODY



:IATION OF TOTAL EMISSIVITY AND TEMPERATURE DURING A Zr-4 STEAM-
DATION TEST AT 1973 K

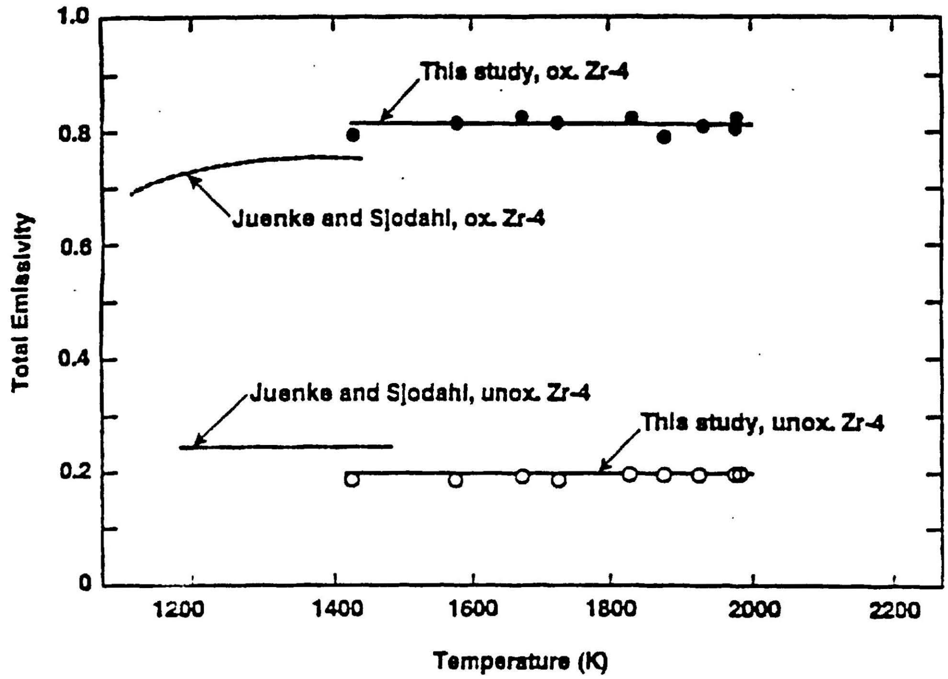


FIGURE 4. TOTAL EMISSIVITY OF UNOXIDIZED AND STEAM-OXIDIZED Zr-4 AS A FUNCTION OF TEMPERATURE