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SUMMARY

TEMPERATURE ESTIMATES FROM THE ZIRCALOY OXIDATION KINETICS IN THE α PLUS β PHASE REGION

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Oxidation rates of zircaloy in steam were measured at temperatures between 961 and 1264 K and for duration times between 25 and 1900 seconds in order to calculate, in conjunction with measurements from postirradiation metallographic examination, the prior peak temperatures of zircaloy fuel rod cladding. These temperature estimates will be used in light water reactor research programs to assess (a) the accuracy of temperature measurements of fuel rod cladding peak temperatures from thermocouples attached to the surface during loss-of-coolant experiments (LOCEs), (b) the perturbation of the fuel rod cladding LOCE temperature history caused by the presence of thermocouples, and (c) the measurements of cladding azimuthal temperature gradients near the thermocouple locations.

Zircaloy-4 samples about 2.3 cm long were cut from unirradiated tubing (1.07-cm outside diameter by 0.0601 cm thick) fabricated according to commercial specifications used for the Loss-of-Fluid Test (LOFT) fuel rods. The samples were heated in a tube furnace with a steam flow of 20 g per minute. A Chromel-Alumel thermocouple placed inside the tube sample was used to record the sample temperature as a function of time. Effective time durations at temperature were calculated from temperature-time plots. Weight gain measurements were obtained from deflections of a quartz spring and from pre- and posttest weight measurements. Oxide and oxide plus

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oxygen-rich alpha thicknesses were obtained from metallographic measurements.

The weight gain from posttest measurements followed parabolic kinetics given by Equation 1:

$$W = 479.02 \left(\frac{+23.3\%}{-18.9\%} \right) \exp \left(-20,400 \left(\frac{+2.3\%}{-2.3\%} \right) / RT \right) t^{1/2} \quad (1)$$

where

W = weight gain (mg O/cm^2)

R = 1.987 cal/mole-K

T = temperature (K)

t = time (s).

The growth of the oxide plus oxygen-rich layer also followed parabolic kinetics and is given by:

$$X = 2.52611 \times 10^4 \left(\frac{+38.5\%}{-27.8\%} \right) \exp \left(-23,920 \left(\frac{+3.1\%}{-3.1\%} \right) / RT \right) t^{1/2} \quad (2)$$

where X is the thickness (micrometers).

The weight gain rate data lie below Leistikow's data¹ which substantially follow cubic kinetics at low temperatures. The rate data for reaction layer thicknesses lie slightly below Biederman's data² with reasonably good agreement.

Zircaloy oxidation kinetics were applied to a zircaloy cladded electrical heater rod subjected to four LOCEs. Based upon the oxidation kinetics for oxide layers (Equation 2), the temperatures near two

surface-attached thermocouples and one embedded thermocouple were 34, 19, and 16 K, respectively, higher than the values determined by thermocouples. These differences are within the 4% uncertainty (50 K) estimated by linear error propagation techniques. Measurements of the oxide thickness indicated the peak temperature of 1191 K occurred between but opposite the two surface attached thermocouples.

The results presented here extend the kinetics of zircaloy oxidation to reaction layer thicknesses in intermediate temperatures in the two-phase alpha plus beta temperature region, and with Biederman's results establish parabolic temperature kinetics in this temperature region. These oxidation kinetics can be used to estimate prior cladding peak temperatures achieved during LOCEs from postirradiation examinations.

REFERENCES

1. S. Leistikow et al., Kinetik und Morphologie der Isothermen Dampf-Oxidation von Zircaloy 4 bei 700-1300°C, KfK 2567 (May 1978).
2. R. R. Biederman et al., "A study of Zircaloy-Steam Oxidation Reaction Kinetics, First Interim Progress Report," October 1976.