RELOCATION OF MOLTEN ZIRCALOY IN A CANDU SUBCHANNEL GEOMETRY

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ABSTRACT

The results of the experiments described in this paper helped identify two mechanisms that will restrict bundle temperature escalation in those postulated CANDU® nuclear power reactor accidents in which considerable heat is generated by the Zr/steam reaction. One mechanism involves the formation of a zirconium oxide crucible around the fuel pins and the other, the relocation of molten sheath within the crucible. Both mechanisms result in a reduction in the area of Zircaloy-4 sheath exposed to the steam. Because the rate of heat generation during a reaction is proportional to area, reduction in the area of exposed sheath will limit the temperature escalation during an accident.

INTRODUCTION

One of the key phenomena in a number of postulated severe nuclear reactor accidents is the relocation of molten Zircaloy-4 sheath. In the CANDU nuclear reactor geometry, relocation of the molten sheath is believed to be an important factor in the mitigation of the effects of the highly exothermic Zr/steam reaction, in particular, resulting in a reduction in the rate of heat generation from the reaction and, consequently, a lower-than-expected maximum fuel temperature. As observed in tests at Westinghouse Canada [1], melt relocates to gaps between the fuel pellets as well as into existing cracks in the pellets, reducing the area of Zr exposed to the steam. Akalin [2] used this observation to develop a model which predicted that the area reduction from relocation alone could dramatically reduce the maximum fuel temperature reached during a postulated reactor accident. On the basis of these developments, experiments were undertaken at AECL Whiteshell Laboratories to evaluate the influence of relocation on the heat generation rate and to identify other mechanisms that could have the same effect.

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EXPERIMENTAL

The facility used in the experiments is shown schematically in Figure 1. The heart of the facility is a vertically-oriented tube furnace heated by a graphite resistance element enclosed in an inert-gas-filled chamber. The furnace is capable of a maximum internal hot zone temperature of about 1850°C. Specimens were loaded into the bottom of the furnace tube, the system closed, evacuated and back-filled with Ultra-High Purity argon. The sample was then inserted part way into the furnace hot zone by a motorized ram, to be preheated to either ~1000°C or ~1400°C. High temperature steam was then allowed into the bottom of the furnace tube while the specimen was being inserted into the centre of the hot zone; these two actions were timed such that the specimen was exposed to the steam shortly before its arrival at the centre of the hot zone. Insertion rates were varied between 3.5 and 68 mm/s to obtain a range of specimen heat-up rates.

The specimen geometry used in most of the experiments is shown in Figure 2. The specimen consists of two and one-half sheathed, hollow UO_2 fuel pellets arranged as a trefoil, the "close-packed" element configuration in a CANDU fuel bundle. A deflector made from the bottom of a round-bottom thoria crucible directed the upward-flowing steam into the triangular region between the three pellets. Some horizontally- and vertically-oriented sheathed single-pellet specimens, and horizontally-oriented specimens consisting of two side-by-side sheathed pellets were also included in the program to provide conditions where little or no relocation took place.

A thermocouple welded to the sheath monitored the specimen temperature up to ~1700°C and a two-colour pyrometer focussed at the centre of the hot zone monitored the specimen temperature from its arrival at the centre of the hot zone to the end of the test. Temperature was recorded as a function of time; the temperatures were used as input data into the CATHENA (Canadian Algorithm for Thermalhydraulic Numerical Analysis) code to analyse the results and to model the test.

Throughout all of the experiments, the rate of hydrogen evolution from the Zr/steam reaction was used as a measure of the extent of the oxidation and thus of the heat generated by the reaction. The hydrogen was collected in a pair of water displacement units; the volume of hydrogen generated was measured as a function of time. The measured hydrogen production was then compared to that calculated by the CATHENA code using the above temperature/time data and either the original Zircaloy-4 surface area or the exposed surface area measured after the test.

Each trefoil sample was sectioned in three orthogonal directions to evaluate the extent of the relocation and the surface area of the relocated material exposed to steam. The composition of the various phases found in these samples was determined using a Tracor-Northern TN-2000 energy dispersive analysis of X-rays (EDAX) unit installed on an ISO DS-100 scanning electron microscope. The other two specimen configurations were generally sectioned in one plane, the single pellet specimens parallel to the pellet axis and the two-pellet specimens perpendicular to this axis.

RESULTS

A photograph of a trefoil specimen after testing is shown in Figure 3 before being sectioned, and in Figure 4 after being sectioned perpendicularly to the pellet axis. The microstructural characteristics in Figure 4 were typical of those of all the other trefoil specimen cross-sections. A white outer oxide (A, ZrO_2) has formed around all three pellets (Figure 4). The darker material beneath this outer layer (at B, for example) is oxidized previously-molten material containing U ($(Zr,U)O_{2-x}$). Some of the molten material relocated only a short distance or remained essentially in place (at B, for example); other molten material relocated either to the bottom of the upper two pellets or into the triangular region between the three pellets (C). In some specimens, the material in the triangular region was only partially oxidized; material that remained unoxidized contained Zr, U and the Zircaloy-4 alloying elements (D).

The oxide layer completely surrounding the three pellets of the trefoil was termed the "crucible". It is evident from Figure 4 that steam access to the material beneath the oxide would be restricted once the crucible was formed. This would particularly be the case for material that had relocated into the triangular region between the three pellets. In addition to the crucible, steam access to the triangular region was also restricted from the ends of the specimens. Evidence of this effect is presented in Figure 5; a web of material has formed across the triangular opening, restricting access of the steam to the inside of the triangular interstice. In addition, Figure 5 also shows that the ends of the sheath have "sealed" to the pellets preventing escape of the liquid from beneath the outer oxide or ingress of the steam. Effectively, all of the molten sheath must be oxidized perpendicularly to the sides of the pellets or from the ends of the triangular void.

It was found that the circumference of the crucible was less than the sum of the original circumferences of the sheaths, resulting in a reduction of the area of the sheath exposed to the steam. Additional area reduction is brought about by relocation; only the accumulated lengths of molten material still clinging to the outer oxide shell (B, Figure 4) remain to be oxidized by the steam in the direction perpendicular to the sides of the pellet. The remaining exposed area, the end closure, was estimated from the type of cross-section shown in Figure 5, made from each trefoil sample.

Typical results of the CATHENA analyses are presented in Figure 6B. Figure 6A is a plot of the measured variation in temperature with time over the duration of a test. There was always a gap between the last recorded temperature before the thermocouple failed and the time of arrival of the specimen within the focal length of the pyrometer, where it was reasonable to assume that the actual specimen surface temperature was being recorded. In general, the thermocouple trace was simply extended to the intersection of the pyrometer trace and the furnace temperature trace.

The modelling of a typical test in which significant relocation occurred is shown in Figure 6B. This graph is a plot of the measured volume of hydrogen produced by the Zr/steam reaction as a function of time, as well as the hydrogen production calculated by the CATHENA code utilizing the different measured areas of sheath exposed to steam. The beginning of

relocation was estimated by obtaining the minimum oxide thicknesses at the location where the melt had relocated from and using the CATHENA code to calculate a time to produce this thickness of oxide. This time coincided approximately with the point at which the measured hydrogen production curve began to deviate from the hydrogen production curve calculated by the CATHENA code using the temperatures from a plot such as Figure 6A and assuming no change in the original area of Zircaloy-4 throughout the test. Using the original Zircaloy-4 area overestimates the rate of hydrogen evolution beyond the beginning of relocation, provided the test conditions promote relocation.

Calculating the hydrogen production after the start of relocation using only the combined area reduction from relocation and that from the crucible formation underestimates the rate of hydrogen evolution. Closure of the ends of the triangular region between the three pellets increases the area of exposed Zircaloy-4; when the end effects are included, the calculated and the measured variation of hydrogen evolution with time agree quite well. Because the rate of hydrogen production is proportional to the rate of heat evolved from the Zr/steam reaction, it can be concluded that the reduction of the area of Zr-bearing material exposed to steam is a major factor in the mitigation of the temperature escalation and the maximum temperature reached in many of the tests designed to simulate severe accidents such as the Westinghouse tests [1].

A number of combinations of kinetic relationships can be chosen within the CATHENA code to calculate the consequences of the Zr/steam reaction. All of the calculations in this study were based on the kinetic relationships developed by Cathcart et al. [3] below the tetragonal-to-cubic ZrO_{2-x} transition (~1500°C) while the Prater and Courtright kinetics [4] were used above this transition. This combination provided the best agreement with the data from the present experiments.

Figure 7 shows that specimen heating rate also influences the extent of the area reduction developed in a temperature transient of the type studied. The heating rates were taken as the initial slopes of the measured temperature/time curves (e.g., Figure 6A). It appears from Figure 7 that little area reduction and relocation will take place below a specimen heating rate of 25°C/s. Furthermore, the exposed Zircaloy-4 area established at 25°C/s of ~25% of the original sheath area does not change appreciably up to a heating rate of ~60°C/s, suggesting that the mitigating effect of the two mechanisms of reducing the area of Zircaloy-4 exposed to steam also remains constant over this heating rate range.

It should be noted that the measured and the calculated hydrogen production rates agreed for those samples in which little or no relocation occurred when the calculated rate was based on the original Zircaloy-4 sheath surface area.

DISCUSSION

The microstructures in Figures 4 and 5, the agreement between the measured and the calculated start of relocation (Figure 6A) and the composition of the phases found in those microstructures, indicate that the original area of the sheath is initially oxidized, followed by the

formation of the crucible, and the melting and relocation of the remaining non-oxidized sheath. The presence of U in the melt is probably a result of a combination of solid-state interaction between the Zircaloy-4 and the UO_2 pellets during the preheat step and during heating to temperature, and the dissolution of some of the UO_2 after melting and during relocation. The closure of the ends of the triangular space between the pellets appears to be caused by a combination of the expansion of the oxide after relocation and the melt running down the inside of the outer oxide layer.

The formation of a crucible around trefoils of pellets was noted in the Westinghouse tests [1]. The formation of the crucible clearly depends on the heating rate (Figure 7) and probably on the proximity of neighbouring pellets. If the neighbouring pellets are close enough, the thermal expansion of the components of the trefoil specimen may bring the sheath on adjacent pellets into contact, allowing the oxide in the area of contact to dissolve and the two sheaths to bond. The pressure of the melt relocating into the area between the pellets will force the oxide to bulge out in this region; continuing oxidation seals the contact region between the sheaths from the upper two pellets. A similar mechanism bridges the gaps between the upper two pellets and the lower half-pellet of the trefoil.

The need to include the end-effect to bring the calculated and the measured hydrogen production rates into agreement is essentially unique to the length of trefoil specimen used in this study. The end effect would become increasingly inconsequential as the length of specimen increased. Indeed, it is anticipated that for longer trefoil specimens, the combined reduction in area from the crucible formation and the relocation of molten sheath would be the only factors necessary to model the oxidation behaviour of the sheath on a trefoil of fuel elements in the type of transient used in this investigation.

CONCLUSIONS

1. A sequence of events that reduces the rate of hydrogen production from the Zr/steam reaction when a trefoil of CANDU-sized fuel elements is subjected to a high temperature transient in steam has been identified.

2. Satisfactory agreement between the measured and the calculated hydrogen production rates can be obtained by considering only the area changes that occur in a trefoil sample undergoing a high temperature transient in steam. In effect, the major contributor to the mitigation of the temperature escalation during such transients is the decrease in the area of Zircaloy-4 exposed to the steam.

3. Two mechanisms of area reduction have been found to operate in trefoil samples, one involving the development of an oxide crucible surrounding all three pellets and one involving the relocation of molten sheath. Neither mechanism operates when the heat-up rate of the specimen is less than 25°C/s.

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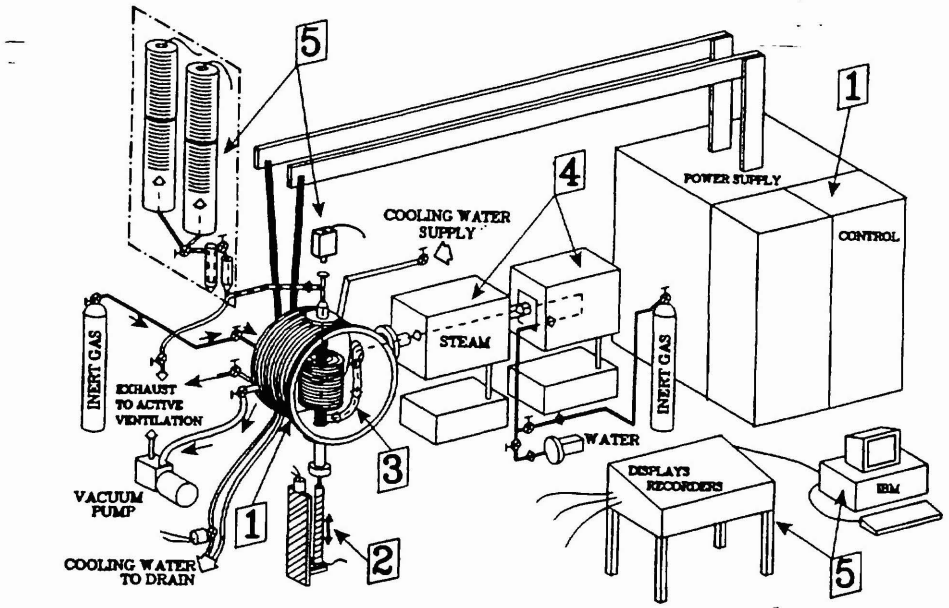

FIGURE 1: SCHEMATIC DIAGRAM OF THE TEST FACILITY. (1) FURNACE CHAMBER, POWER SUPPLY AND CONTROL, (2) SPECIMEN INSERTION MECHANISM, (3) STEAM LINE, (4) STEAM GENERATOR, (5) TEST MONITORING SYSTEM

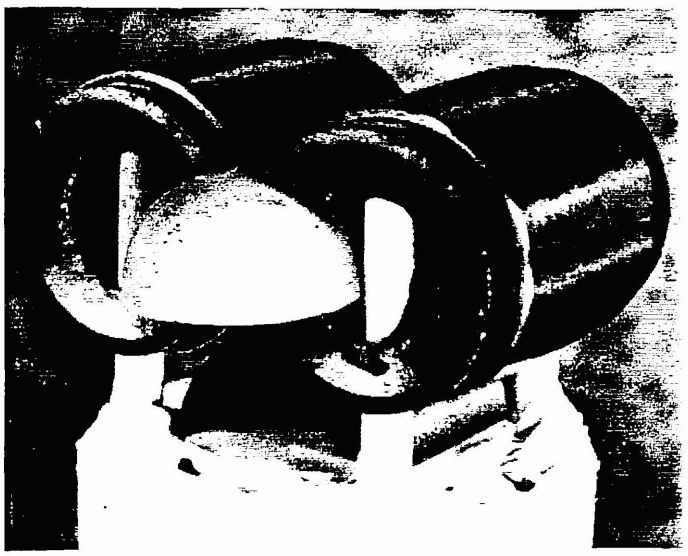

FIGURE 2: VIEW OF A TREFOIL SAMPLE BEFORE TESTING

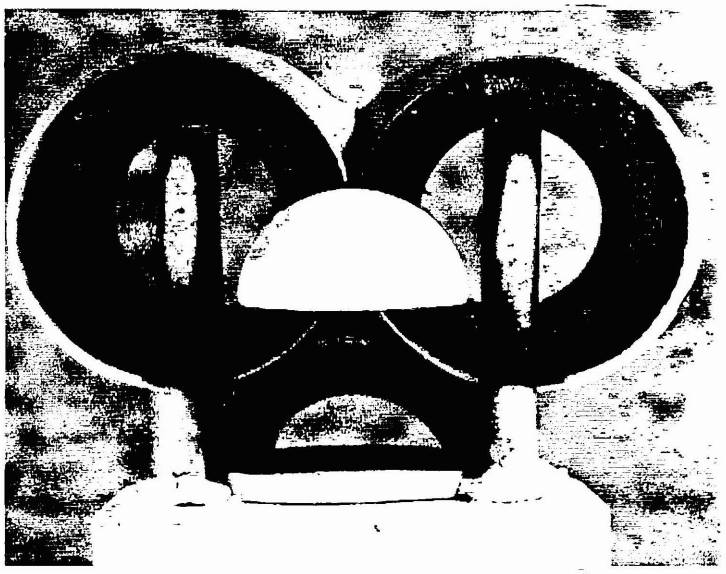

FIGURE 3: PHOTOGRAPH SHOWING THE APPEARANCE OF A TREFOIL SAMPLE AFTER TESTING

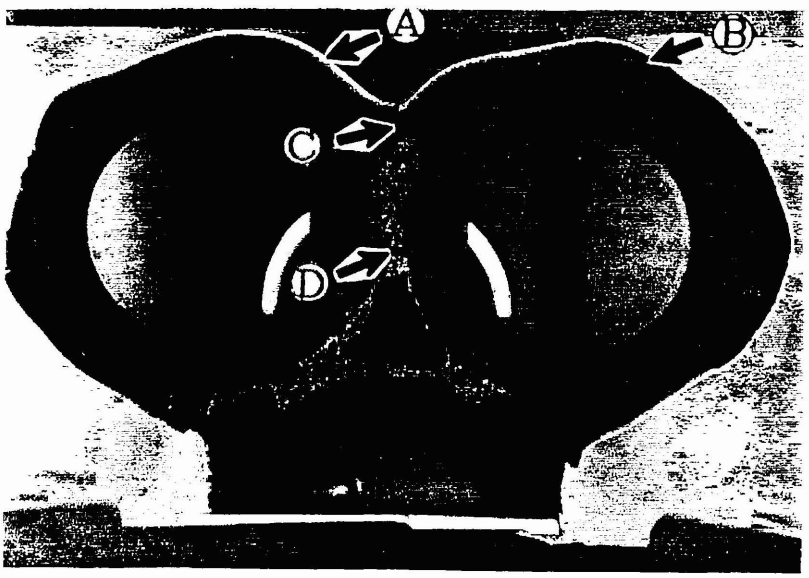

FIGURE 4: TYPICAL TREFOIL CROSS-SECTION. A: CRUCIBLE, B: $(Zr,U)O_{2-x}$, C: RELOCATED MOLTEN SHEATH, D: NON-OXIDIZED PREVIOUSLY-MOLTEN SHEATH

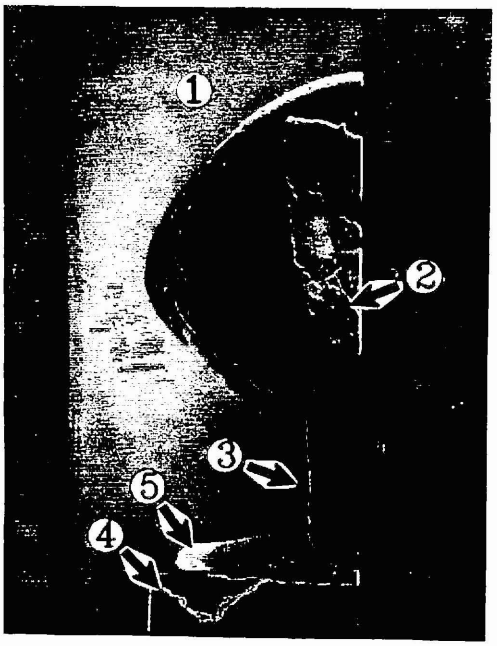

FIGURE 5: CROSS-SECTION BETWEEN UPPER TWO PELLETS OF A TREFOIL SHOWING WEB (3) FORMED OVER THE TRIANGULAR REGION BETWEEN THE PELLETS (1) OXIDIZED SHEATH ON UPPER PELLET, (2) NON-OXIDIZED, PREVIOUSLY MOLTEN MATERIAL (3) WEB, (4) LOWER HALF-PELLET, (5) OXIDIZED SHEATH ON LOWER HALF-PELLET

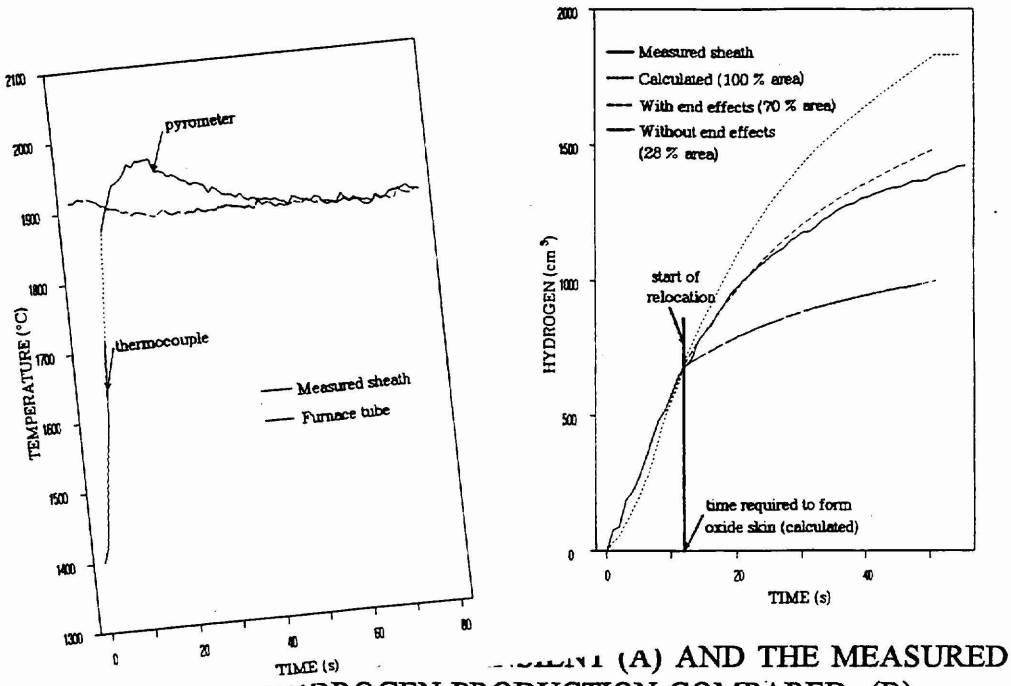

FIGURE 6: [illegible] (A) AND THE MEASURED AND [illegible] HYDROGEN PRODUCTION COMPARED (B)

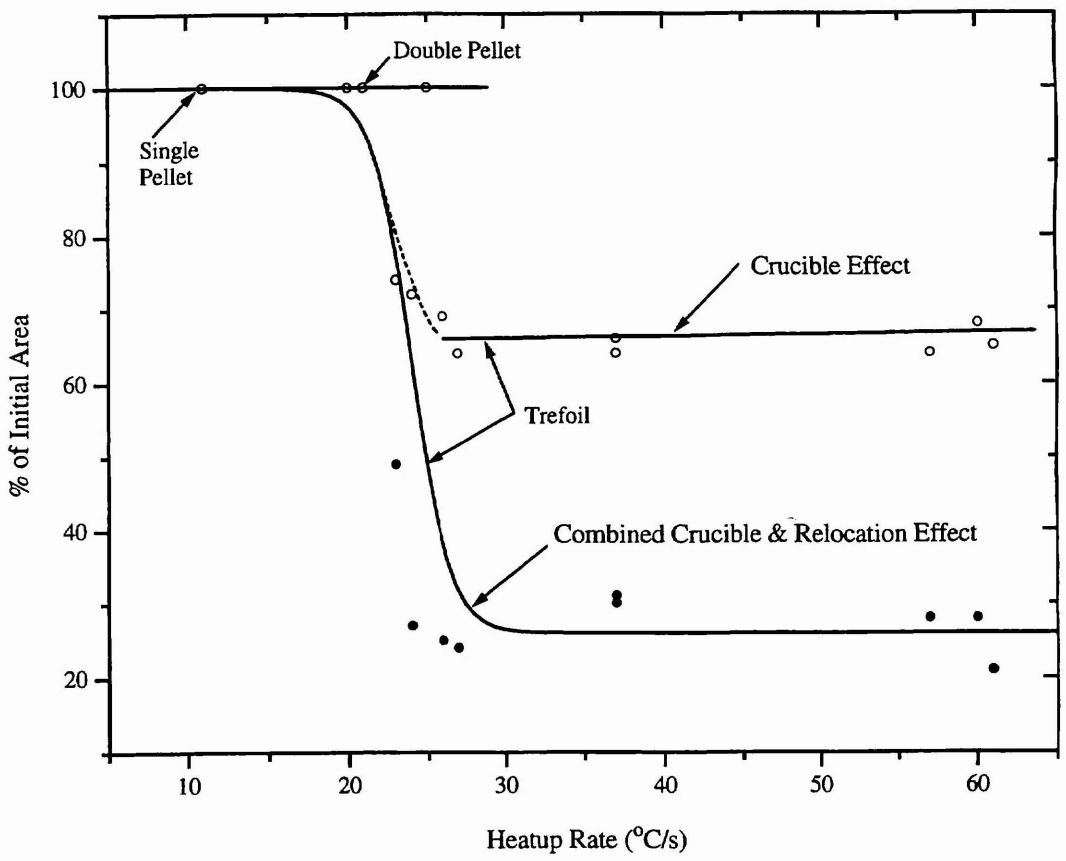

FIGURE 7: RELATIONSHIP BETWEEN THE SPECIMEN HEAT-UP RATE AND THE EXPOSED SHEATH AREA