Post-Oxidation Ductility of Machined-and-Polished E110
After Two-Sided Steam Oxidation at 1100˚C

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Introduction

A series of tests were conducted with machined (to ≈0.6-mm wall thickness) and polished E110 samples to determine weight gain kinetics prior to breakaway oxidation for comparison to the Russian alloy G110, which also exhibits good oxidation behavior, and to M5. Table 1 shows the test conditions and results [1]. All samples exhibited a lustrous black oxide layer up to the highest hold (1011 s) at 1100˚C. At this hold time, small white spots were observed on the outer surface oxide layer. The temperature history for the 1100°C oxidation tests with 0.61-mm wall-thickness is shown in Fig. 1. The weight gain results are plotted in Fig. 2 vs. the Cathcart-Pawel (CP) predicted weight gain. Unlike results at 1000°C [1], much better agreement is achieved between the E110 weight gain kinetics and the CP-predicted values. Similar behavior is observed for the M5 tested at ANL [2]. The ANL results are also in good agreement with the G110 alloy tested by RRC KI [3].

Table 1 Summary of Two-Sided Oxidation Test Results at 1100°C for Machined-and-Polished Samples; polished cladding outer diameter is 9.14 mm; machined-and-polished inner diameter is 7.76-7.98 mm and wall thickness is 0.58-0.69 mm; Δw_p = predicted normalized weight gain based on Cathcart-Pawel correlation; Δw_m = measured weight gain 9normalized to surface area); ECR = 1.437 (0.61 mm/h) Δw, where h is the actual wall thickness of the sample.

Wall Thickness mm	Hold Time s	CP ECR %	CP Δw_p mg/cm^3	Meas. Δw_m mg/cm^3	Comment
0.61	111	7.5	5.2	4.9	Lustrous black oxide
0.69	361	10.5	8.3	7.2	Lustrous black oxide
0.58	511	14.4	9.5	8.5	Lustrous black oxide
0.58	576	15.3	10.1	---	Lustrous black oxide
0.58	1011	19.4	12.9	10.7	Lustrous black oxide with small white spots

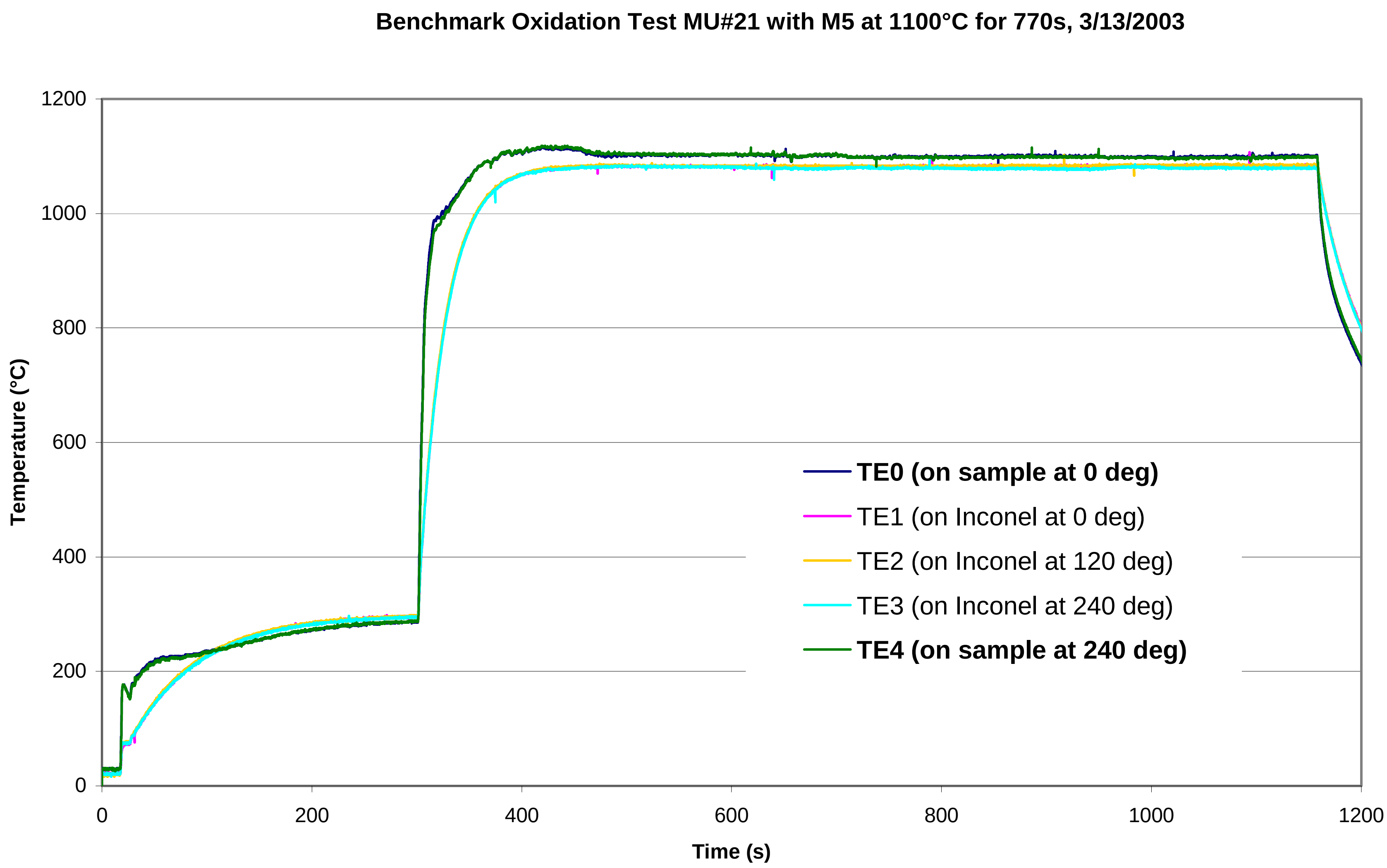

Fig. 1. Thermal benchmark results for 0.61-mm-wall M5 sample (MU#21) for a hold temperature of 1100°C.

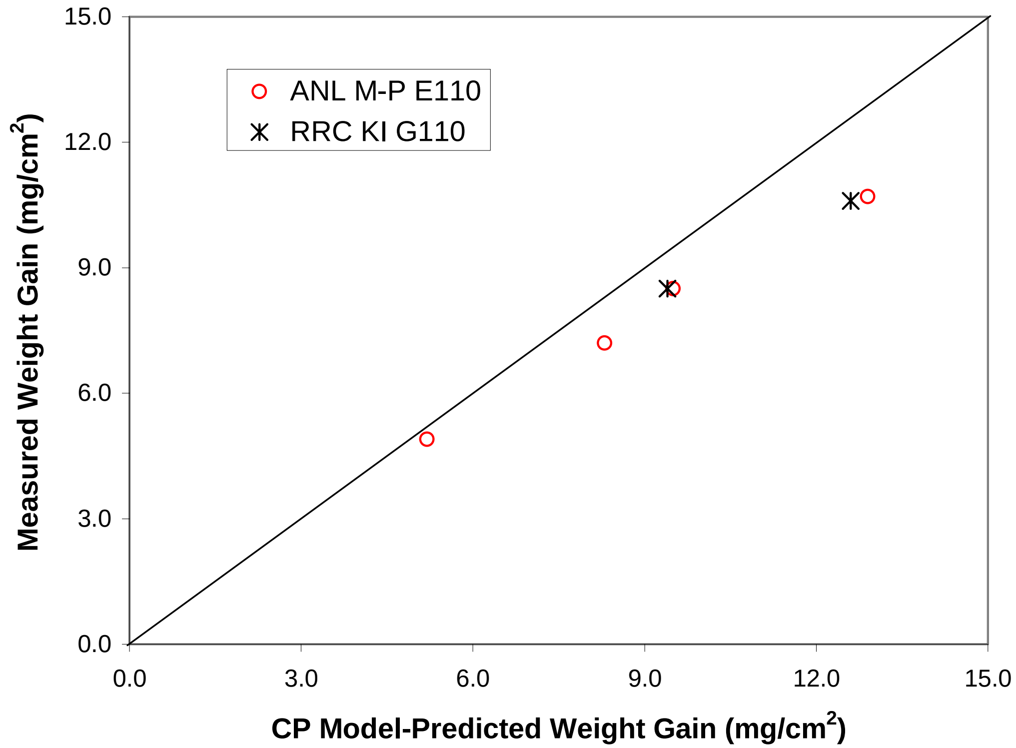

Fig. 2. Comparison of ANL weight gain results for machined-and-polished E110 samples with RRC KI results for Alloy G110 and the Cathcart-Pawel (CP) model predictions for steam oxidation at 1100°C.

Room-Temperature Ring-Compression Data

Although the original intent of the work with machined-and-polished E110 was to study time-temperature regimes for stable oxide growth on E110, as well as instability regimes of breakaway oxidation and hydrogen pickup, recently it was decided to perform room-temperature ring compression tests on four of the samples in Table 1. This was motivated by the Bochvar-Institute/TVEL presentation [4] on the RT ring-compression ductility of 7 lots of E110, with varying concentrations of Hf, Fe, and O (all within material specification ranges), as well as varying combined impurity levels (Ni, Al, Si, Ca, K, F, Cl, Na, Mg). The E110 tubing used in the Bochvar/TVEL study had an outer-diameter of 9.13 mm after "pickling". Although not specified, it is assumed that the tubing wall thickness was 0.71 mm. It is not clear whether or not the tubing was anodized – standard procedure to convert tubing into cladding – following pickling. 30-mm-long tubing segments were exposed to two-sided steam oxidation for 10 minutes (600 s). Both heating (300˚C to 1100˚C in ≈2 s) and cooling (1100˚C to 300˚C in 15-20 s) rates were relatively fast although the samples were not quenched. The authors claim a weight gain of ≈8 mg/cm^2 and an ECR of ≈10% for the samples exposed to steam at 1100˚C for 10 minutes. Ignoring oxidation during the heating and cooling ramps, the Cathcart-Pawel weight-gain model would predict $\Delta w = 9.76$ mg/ cm^2 and 12% ECR.

Four of the Bochvar/TVEL lots (1, 5, 6, 7) with a combined impurity level of 110-135 wppm exhibited outer-surface breakaway oxidation, high hydrogen pickup (200-600 wppm), and low RT ring-compression ductility (<5%). Three of the lots (2, 3, 4) with combined impurity levels of 25-45 wppm exhibited stable oxide growth (observed at low-magnification), lower hydrogen pickup (60-200 wppm) and higher RT ring-compression offset strain (5-10%). Based on ANL experience with the effects of surface finish (polishing vs. pickling) on oxide-growth stability, it was hypothesized that the Bochvar/TVEL samples would have exhibited more stable oxide growth, lower hydrogen pickup and higher ductility if the surfaces had been polished, rather than pickled. In order to test this hypothesis, four of the ANL machined-and-polished (M-P) samples in Table 1 were subjected to RT ring compression tests. The results of these tests are compared to the best of the Bochvar-TVEL results (Lots 2-4) in Table 2 and Fig. 3. The offset displacements used to determine offset strain and ductility were derived from the load-displacement curves given in Appendix A.

Discussion

The E110 tubing used in the ANL experiments was obtained from Fortum in Finland. In terms of Fe, Hf and O content, it falls within the range of the chemical composition of the Bochvar/TVEL lots 2-4. Although a limited chemical analysis was performed on two small tubing lengths, differences between the Fortum E110 and the Bochvar/TVEL Lots 2-4 appear to be higher N content (55 vs. 25-35 wppm) and higher C content (110-600 vs. 45-65 wppm). The impurities highlighted by Bochvar/TVEL (Ni, Al, Si, Ca, K, F, Cl, Na, Mg) were not measured for the Fortum cladding used by ANL. However, significantly higher post-oxidation ductility was achieved by ANL by machining the inner-surface to give a wall thickness comparable to M5 and by polishing both inner and outer surfaces. The Bochvar/TVEL samples, which were pickled rather than polished, may have exhibited higher post-oxidation ductility if the surfaces had been polished following pickling or if the surfaces had not been pickled at all.

Table 2 Comparison of E110 Post-Oxidation RT Ductility between ANL Machined-and-Polished (M-P) Cladding Samples and Bochvar/TVEL (BT) Lots 2-4 Pickled Cladding Samples [4] Following Two-Sided Steam Oxidation at 1100˚C

E110 Material	Wall Thickness mm	CP ECR %	Meas. ECR %	Offset Displacement mm	Offset Strain %
ANL M-P	0.61	7.5	7.0	5.47	60
ANL M-P	0.69	10.5	9.2	2.86	31.2
BT Lot 2	0.71*	12	≈10	---	10.0
BT Lot 3	0.71*	12	≈10	---	6.6
BT Lot 4	0.71*	12	≈10	---	4.4
ANL M-P	0.58	14.4	12.7	1.94	21.2
ANL M-P	0.58	19.4	16.0	0.58	6.3

*Assumed

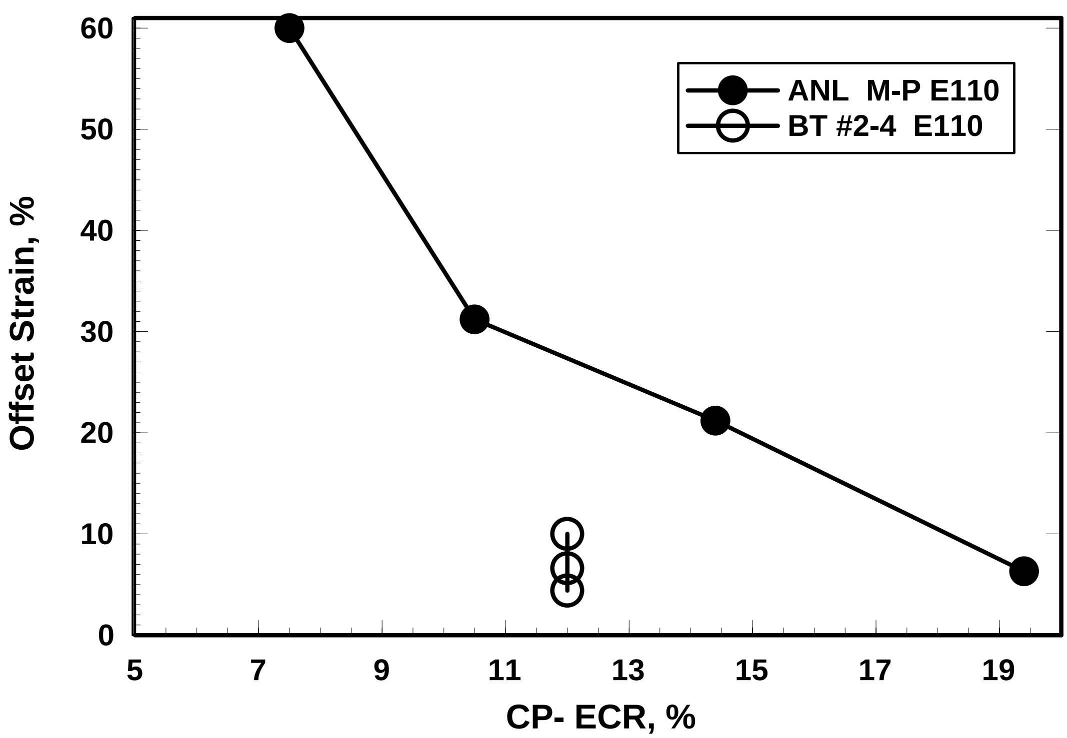

Fig. 3 Post-oxidation RT ductility vs. Cathcart-Pawel (CP) predicted ECR for ANL machined-and-polished (M-P) E110 cladding samples and Bochvar/TVEL (BT) Lots 2-4 pickled E110 cladding samples [4] following two-sided steam oxidation at 1100˚C.

References

1. Y. Yan, T. Burtseva and M. C. Billone, "Progress Report on E110 Post-Quench Ductility Test Program," letter report to NRC and RRC KI, May 31, 2003.

2. Y. Yan, T. Burtseva and M. C. Billone, "Post-Quench Ductility Results for Zry-4 and M5 Oxidized at 1000˚C and 1100˚C," letter report to NRC and Framatome, January 31, 2004.

3. L. Yegorova, K. Lioutov, V. Smirnov, A. Goryachev and V. Chesanov, "LOCA Behavior of E110 Alloy," Proceedings of the 2003 Nuclear Safety Research Conference," Oct. 20-22, 2003, NUREG/CP-0185, June 2004, pp. 123-139.

4. A.V. Nikulina, L.N. Andreeva-Andrievskaya, V.N. Shisov, and Yu. V. Pimenoc, "Influence of Chemical Composition of Nb Containing Zr Alloy Cladding Tubes on Embrittlement under Conditions Simulating Design Basis LOCA," presented at the ASTM 14th International Symposium on Zirconium in the Nuclear Industry, Stockholm, Sweden, June 13-17, 2004, to be published in Proceedings.

Appendix A

Load-Displacement Curves for Machined-and-Polished E110 Samples Oxidized at 1100˚C

and

Ring-Compression Tested at Room Temperature and

2 mm/minute Cross-Head Displacement Rate

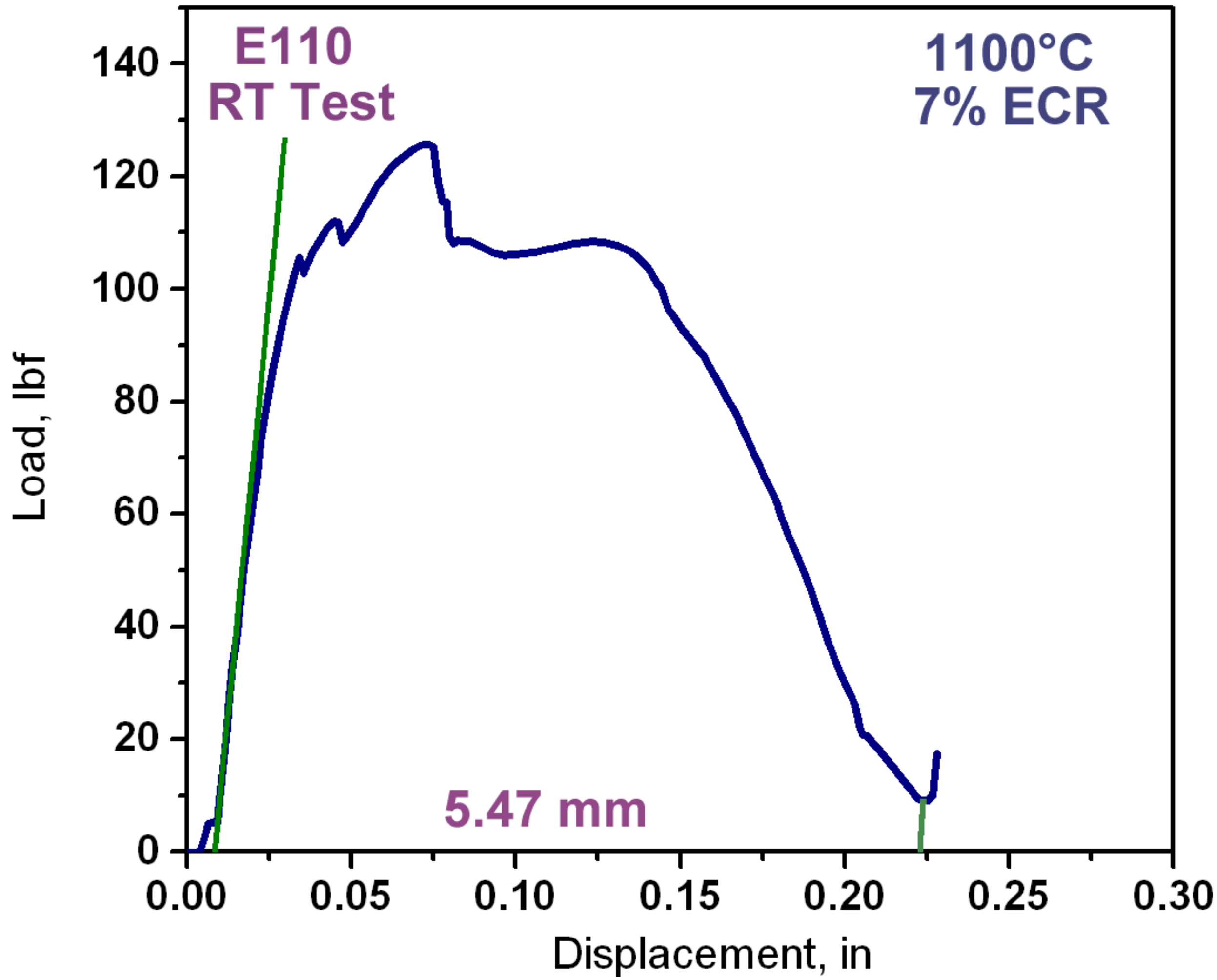

Fig. A.1 Ring-compression load-displacement curve for ANL machined-and-polished E110 sample (7.92-mm-long) oxidized at 1100˚C to a CP-predicted ECR = 7.5% and a measured ECR = 7.0%. Test was conducted at room temperature and a cross-head displacement rate of 2 mm/minute. Through-wall cracks were observed at bottom and side of sample, but they did not extend along the whole length of the sample. Offset displacement is 5.47 mm.

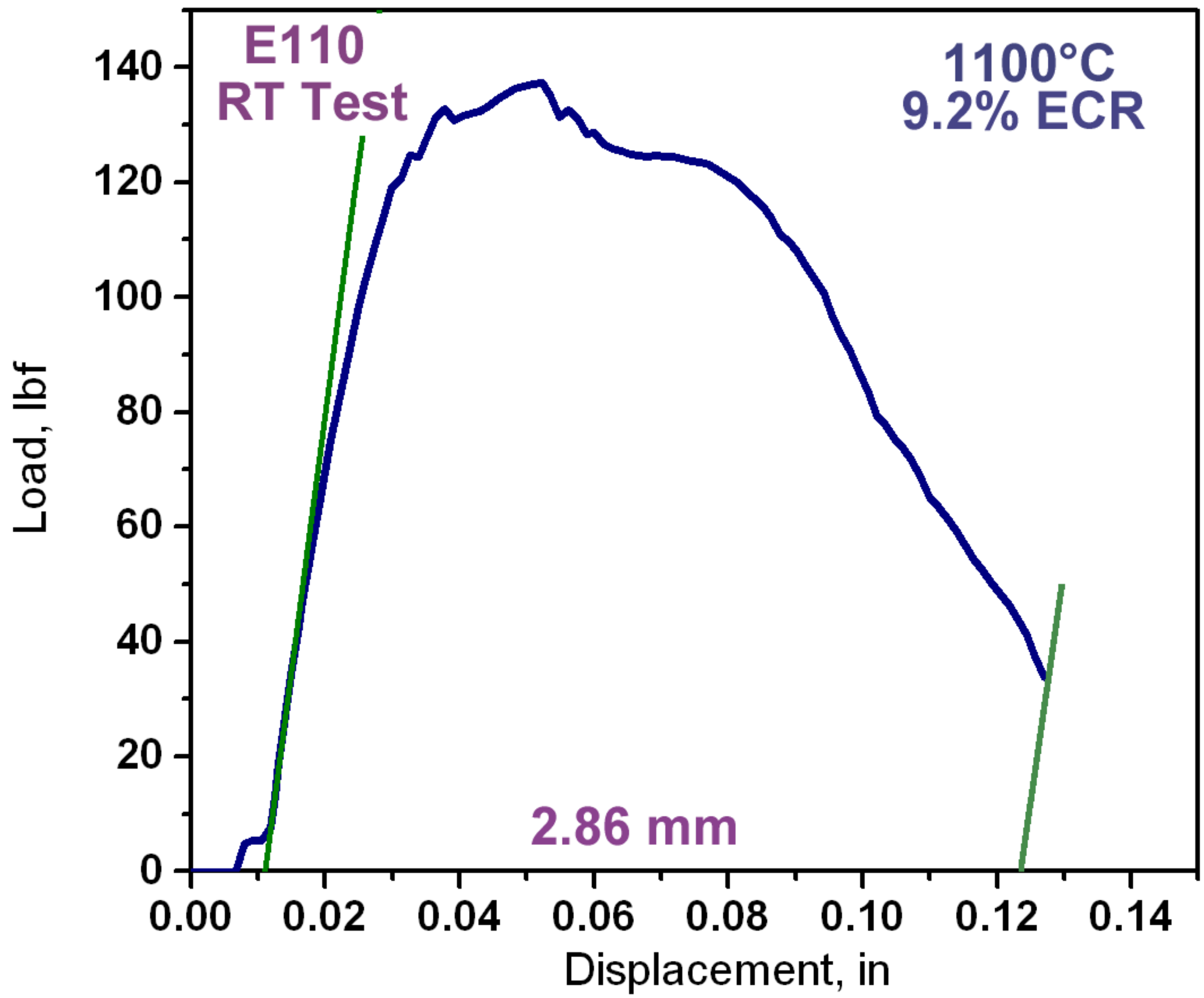

Fig. A.2 Ring-compression load-displacement curve for ANL machined-and-polished E110 sample (7.65-mm-long) oxidized at 1100˚C to a CP-predicted ECR = 10.5% and a measured ECR = 9.2%. Test was conducted at room temperature and a cross-head displacement rate of 2 mm/minute. Cracks were observed at top, side and bottom of sample, but it is not clear that they are through-wall cracks along whole length of sample. Offset displacement is 2.86 mm.

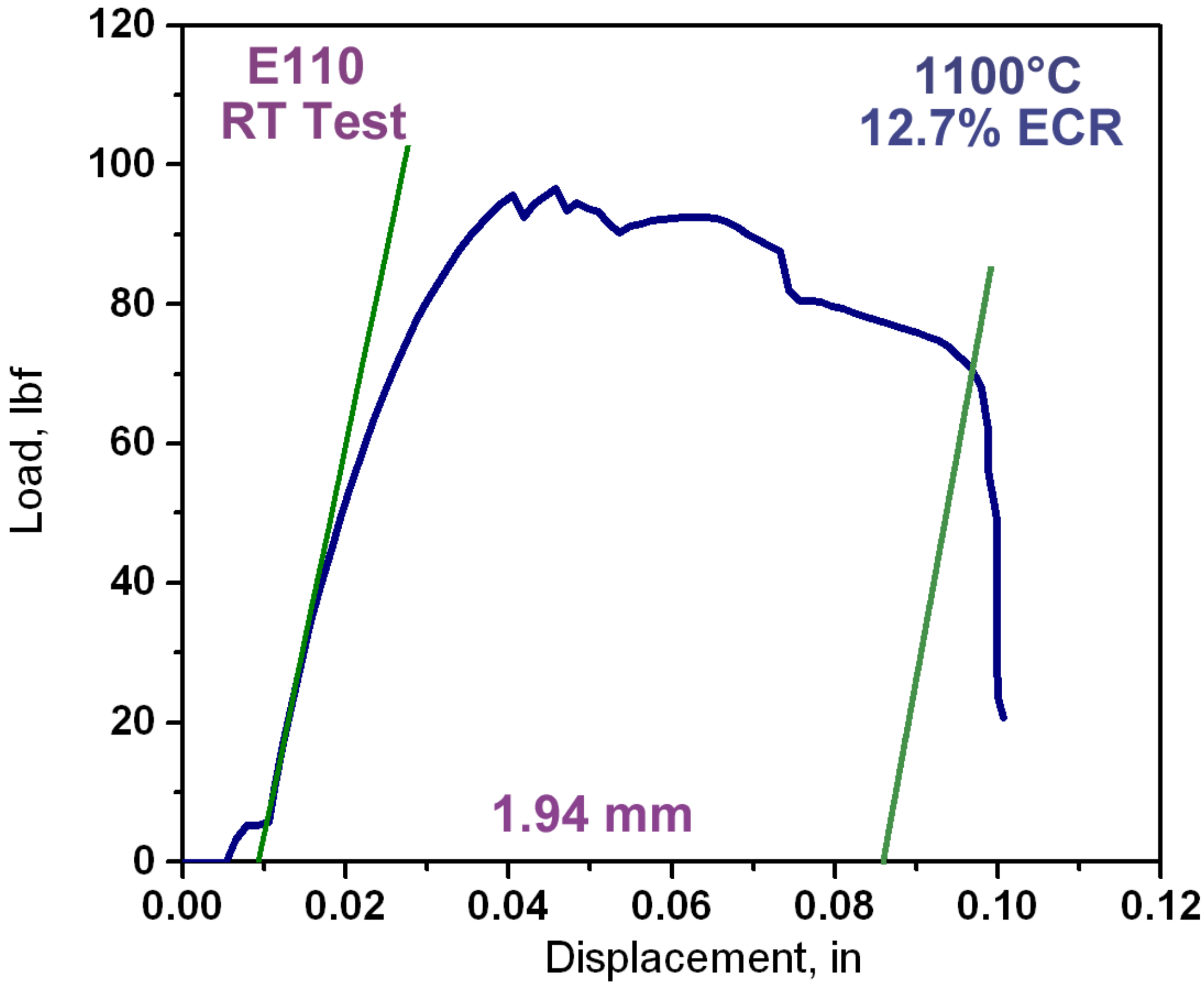

Fig. A.3 Ring-compression load-displacement curve for ANL machined-and-polished E110 sample (8.00-mm-long) oxidized at 1100˚C to a CP-predicted ECR = 14.4% and a measured ECR = 12.7%. Test was conducted at room temperature and a cross-head displacement rate of 2 mm/minute. Through-wall cracks were observed at top, side and bottom of sample. Offset displacement is 1.94 mm.

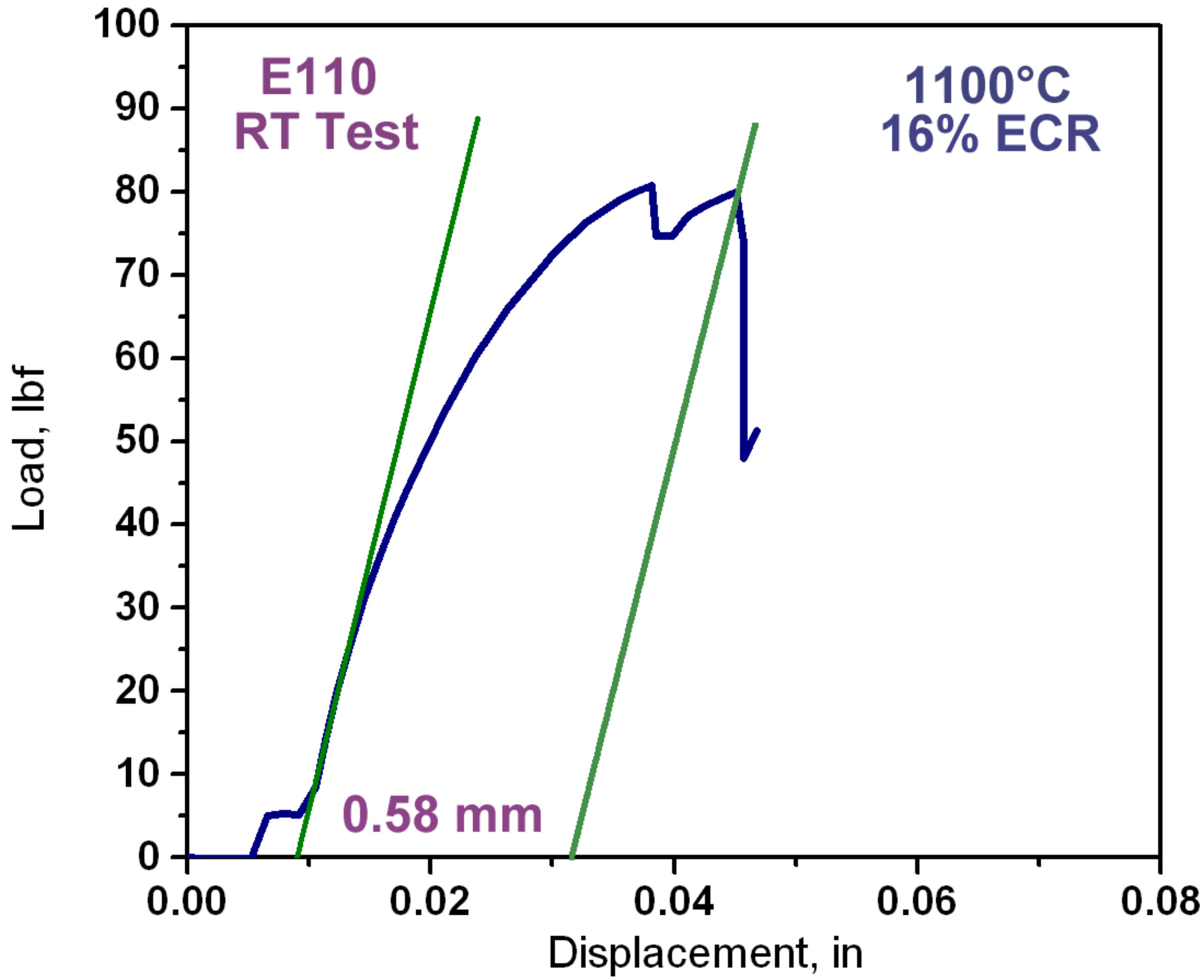

Fig. A.4 Ring-compression load-displacement curve for ANL machined-and-polished E110 sample (8.00-mm-long) oxidized at 1100˚C to a CP-predicted ECR = 19.4% and a measured ECR = 16.0%. Test was conducted at room temperature and a cross-head displacement rate of 2 mm/minute. A tight, through-wall crack was observed at the bottom of sample. Offset displacement is 0.43 mm.