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Grain refinement of T91 alloy by equal channel angular pressing

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ABSTRACT

We report on the grain refinement of modified 9Cr–1Mo ferritic-martensitic steel (T91) by equal channel angular pressing, a severe plastic deformation method. Microstructural refinement depends on processing temperature (300–700 °C) and extrusion strain (1.2–2.3). The average grain size has been refined by over an order of magnitude down to 300 nm, accompanied by a hardness increase of up to 70%. The refined microstructure undergoes little grain growth or softening up to 500 °C. At an annealing temperature of 700 °C or higher, significant softening occurs as a result of grain growth.

1. Introduction

Modified 9Cr–1Mo steel, T91, is a ferritic-martensitic steel currently used in power plant superheaters, and is a potential candidate for structural steel in nuclear reactors. This alloy must have creep resistance, corrosion resistance, and radiation tolerance in order to serve for decades as a structural material in generation IV reactors. Radiation damage tolerance can be especially critical in some other applications such as beam windows for accelerator driven transmutation systems where the damage rate can be tens of displacement per atom (dpa) per year [1]. Thus, the improvement of high temperature mechanical strength and radiation tolerance is critical for future applications of T91 and similar alloys.

One method to improve the mechanical strength of metal is to refine its microstructure. Dislocation activity is diminished within smaller grains, and strengthening due to grain refinement at the submicron or greater length scale is typically described by the Hall–Petch model based on dislocation pile-ups, i.e., the hardness of metals or alloys typically increases linearly with $d^{-0.5}$, where d is the average grain size. Equal channel angular pressing, referred to as ECAP, is a severe plastic deformation technique that has been used to produce a variety of ultra fine grained (UFG) materials [2–5]. ECAP is carried out by pressing a material through a channel with a sharp angle as shown in Fig. 1. In a 90° die, this results in a Von Mises strain of ~1.5, equivalent to a 69% area reduction in conventional rolling. Unlike conventional rolling, the ECAP work piece has essentially the same dimensions as the original material, so the process can be repeated numerous times to achieve a desired level of cumulative strain for microstructure refinement. Additionally, the texture of the material produced by multi-pass ECAP can be varied by changing the orientation of the billet between extrusions [6]. Details on microstructure evolution after ECAP have been summarized in several review articles [7,8]. In spite of the promise of grain refinement via ECAP, there is no study on the influence of ECAP processing parameters on the microstructure refinement of T91 and similar potential candidates for reactor steels. In this paper, we present the first exploration of ECAP processed ferritic T91 alloy. This work is an initial study on the evolution of microstructure and mechanical properties due to ECAP processing and heat treatment.

2. Experimental

The material used in this study was from Oak Ridge National Laboratory heat 10148. The chemical composition of is detailed in Table 1. ECAP processing was carried out in a sliding wall die using an MTS-controlled 225 ton hydraulic press. The ECAP extrusions were carried out on 7 mm diameter T91 rods encapsulated in commercially pure Ni. The Ni can was used to bring the work piece to the 2.4 × 2.4 cm cross-section required by the ECAE tooling utilized for this experiment. The extrusion rate was 1.27 cm/s in all cases. Extrusions were performed at elevated temperature, 300–700 °C. Actual processing temperatures were estimated to be within 20 °C of reported temperatures.

The as-received T91 alloy was fully tempered and predominantly ferritic. The starting materials and processing methods are detailed in Table 2 along with identifications used for the materials. Two-pass billets were rotated by 90° around their long axis prior to the second extrusion. This processing route is referred to as 2B. A specimen extruded using such a route at 300 °C is represented by 2B300. Post-processing heat treatment was carried out in a vacuum tube furnace at a base pressure of 2×10^{-6} torr. The hardness and elastic modulus were measured by a Fischerscope 2000XYp using a Vickers diamond tip and a depth of 4 μm. Transmission electron microscopy (TEM) was performed with a JEOL

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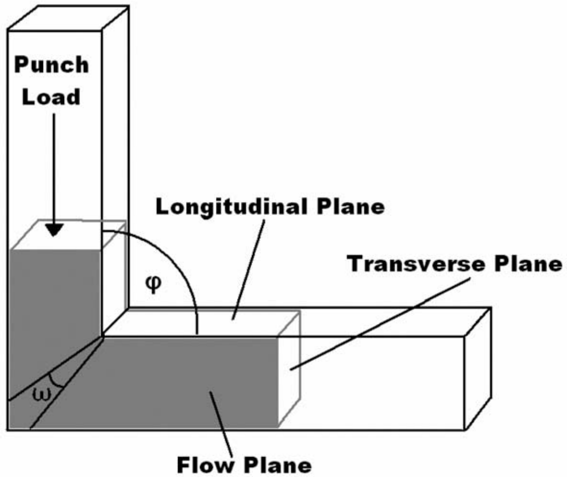

Fig. 1. Schematic of ECAP with a die angle φ. The shear zone has fan angle ω rather than the ideal plane.

Table 1
The chemical composition (wt%) of as-received T91 alloys from supplier.

Cr	Mo	Mn	Ni	C	Si	V	Nb	P	S
9.24	0.96	0.47	0.16	0.089	0.28	0.21	0.054	0.021	0.006

2010 microscope, and selected area diffraction patterns accompanying micrographs were acquired with an SAD aperture of 2.5 μm in diameter.

Table 2
ECAP processing conditions and corresponding billet identifiers.

Identifier	ECAP route	ECAP temperature (°C)
2B300	2B	300
1A700	1	700
2B700	2B	700, 600
AR	As-received	

3. Results

TEM studies were performed to reveal microstructural evolution in detail. The as-received materials had fairly large grain sizes, ~5 μm, as shown by a representative TEM micrograph in Fig. 2(a). The inserted selected area diffraction (SAD) pattern shows a dotted pattern, an indication of rather large grain size. After the single extrusion at 700 °C (1A700), the average grain size dropped moderately to ~1 μm as shown in Fig. 2(b). After two extrusions at this temperature, the 2B700 specimen had a much smaller grain size, approximately 500 nm, with a higher density of high-angle grain boundaries as indicated by the SAD pattern in Fig. 2(c). When the same strain was applied to extrude specimens at a lower temperature, 300 °C, the average grain size was approximately 350 nm with primarily high-angle grain boundaries, as shown in Fig. 2(d). The SAD pattern of 2B300 specimen was nearly ring-like, an indication of randomly oriented very fine grains, and the existence of high angle grain boundaries.

Heat treatment (annealing) was performed for 10 h on the ECAP processed specimens. Systematic studies showed that in general grain coarsening started at 600 °C, and took off rapidly thereafter. Fig. 3 shows the bright field TEM micrograph of 2B300 T91

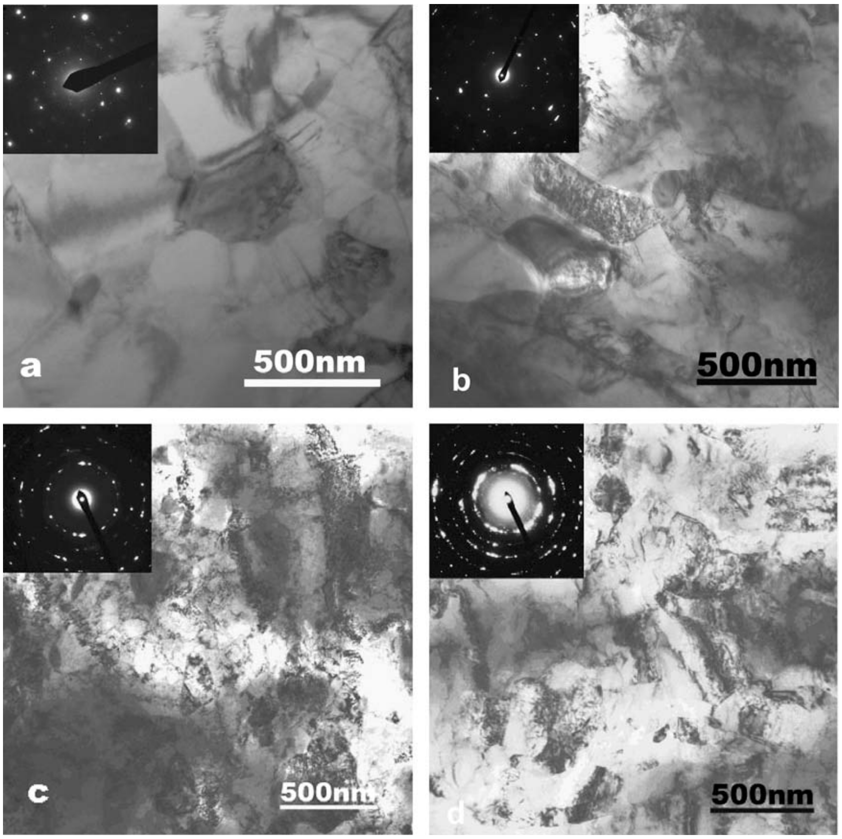

Fig. 2. Representative bright field TEM micrographs with selected area diffraction (SAD) patterns of as-received, and ECAP processed T91 alloys. (a) As-received T91 alloys have an average grain size of ~5 with course structure, (b) 1A700 showing additional boundaries but little change in diffraction pattern, (c) 2B700 showing a refined microstructure, (d) 2B300 showing a highly refined microstructure.

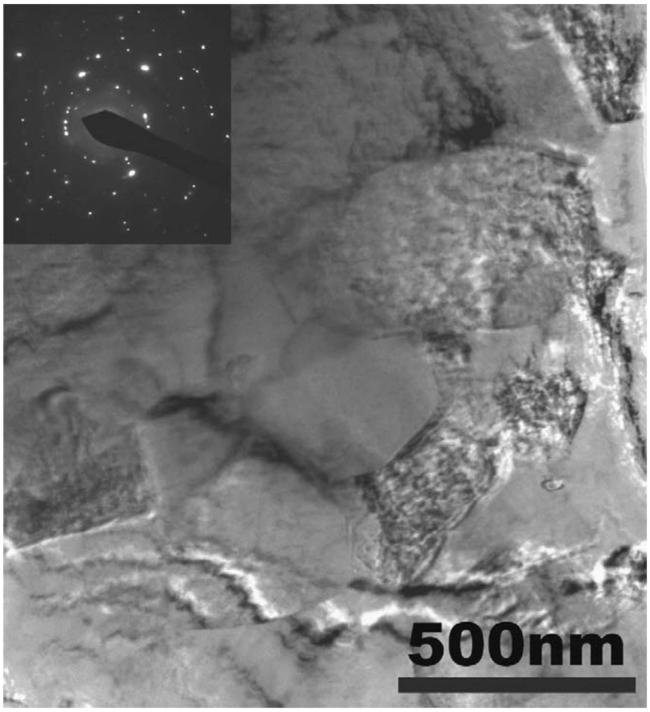

Fig. 3. TEM bright field micrograph with diffraction pattern of 2B300 material after annealing at 600 °C.

specimen annealed at 600 °C. The average grain size of this specimen (from statistical analysis of several micrographs) was determined to be ~500 nm. TEM studies (not shown here) also indicated significant grain growth, upto ~5 μm, for the same specimens annealed at 700 °C.

The evolution of indentation hardness with $d^{-1/2}$, where d is the average grain size, is shown (as solid squares) in Fig. 4. In general, a smaller average grain size lead to a higher hardness. It is unsurprising that the hardnesses of 2B700 and 2B300 were similar given that their average grain sizes were comparable. Fig. 5 shows the hardness evolution of as-received and ECAPed specimens after 10 h of isothermal annealing. As-received specimens showed a monotonic reduction of hardness to 2 GPa, whereas most ECAP processed specimens showed retentions of high strength up to ~500 °C. The onset of softening occurred at ~600 °C. The ECAP processed steel softened considerably after annealing at 700 °C.

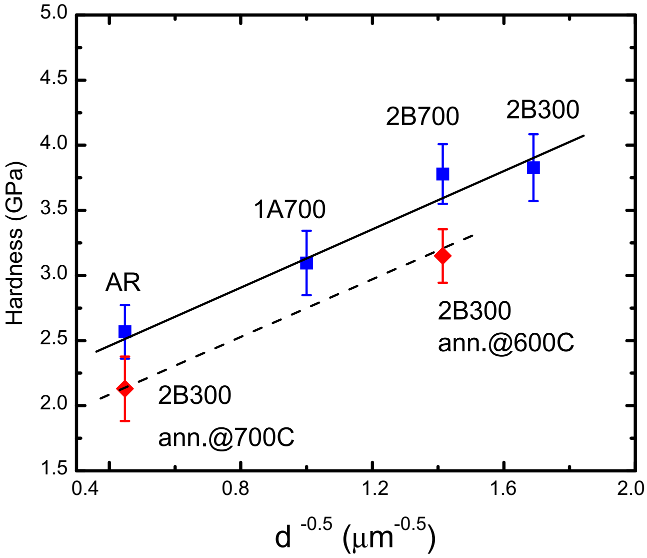

Fig. 4. Hardness vs. grain size. Hardness increases with decreasing grain size for the ferritic T91 materials, following the Hall–Petch model.

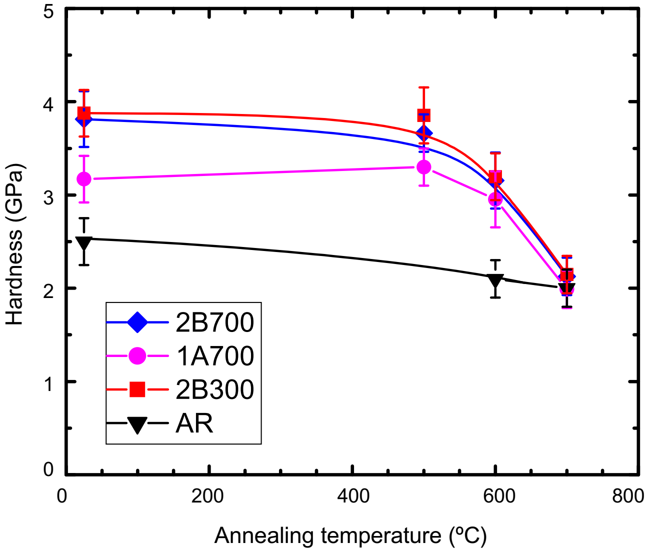

Fig. 5. Hardness vs. annealing temperature for 10 h heat treatment with furnace cooling. Hardness tests were performed at room temperature.

4. Discussion

During ECAP processing, dislocation density increases rapidly with increasing strain. High-density dislocations form cell walls that later evolve into low angle grain boundaries, separating subgrains. Misorientation of neighboring grains becomes more significant at higher strain, and eventually high angle grain boundaries become dominant. The evolution of average grain size vs. extrusion strain is shown in Fig. 6. The Von Mises extrusion strain γ is calculated by Segal as [9]:

$$\gamma = (2/\sqrt{3})N\cot(\phi/2), \tag{1}$$

where N is the number of passes and ϕ is the angle between the inlet and exit channels of the die. As shown in Fig. 6, a higher strain leads to a smaller average grain size, and TEM studies show more grains are separated by predominantly high-angle grain boundaries.

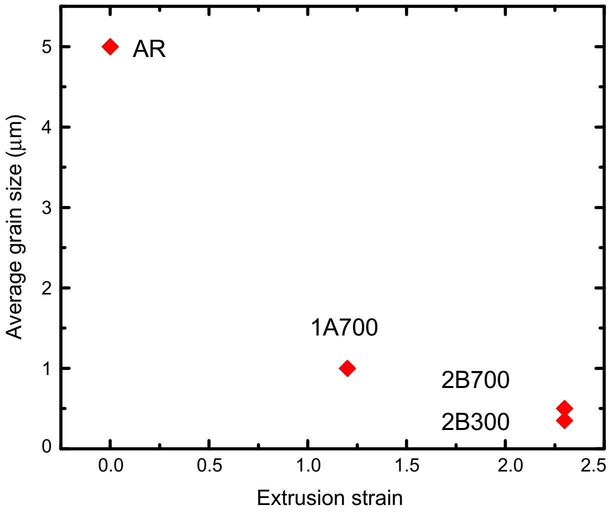

Fig. 6. Evolution of the average grain size with extrusion strain. The average grain size decreases continuously with increasing extrusion strain.

The microstructure of the T91 alloy was successfully refined, by an order of magnitude, from a few microns down to ~350 nm. The authors are attempting to further refine the microstructure by increasing the extrusion strain.

Another approach to refine microstructure is to reduce processing temperature and increase strain rate. During ECAP extrusion processing, the dislocation generation rate and recombination rate (recovery) are a function of temperature. Extrusion at lower temperature will reduce the recovery rate and thus effectively increase the overall dislocation density which later leads to the formation of high angle grain boundaries. The present study shows that a slightly smaller average grain size was achieved in specimens processed at 300 °C comparing to that processed at 700 °C. Theoretically, room temperature processing should reduce the average grain size even further. Practically, extrusion temperature and speed is limited by the flow stress, remaining ductility, and strain sensitivity of the material. The processing temperature is limited to 300 °C in this study to achieve uniform flow (extrusion) of T91 alloys.

The hardening of SPD processed T91 originates mainly from grain refinement, as the predominant phase remains ferrite throughout deformation. A linear relationship in the hardness vs. $d^{-1/2}$ plot indicates that hardening is due to dislocation pile-ups down to submicron length scales within grains as described by the empirical Hall–Petch model [10,11]. A linear fit of the as-processed hardness values was obtained as shown in Fig. 4. When multiplied by 1/3 to obtain the approximately equivalent yield strength values, the slope of this line is 430 MPa $\mu m^{1/2}$, comparable to those in other iron alloys in the literature [12–14]. To understand the influence of annealing on hardness evolution, the hardnesses of annealed 2B300 specimens are compared to those of as-received and processed T91 specimens, as shown in Fig. 4 by the solid diamond symbols. Again, softening due to grain coarsening is confirmed in annealed T91 specimens. However, the hardnesses of annealed T91 specimens clearly fall underneath the linear fit of as-processed specimens, indicating the existence of another contributing factor to the observed softening. Comparing the microstructure of annealed specimens (Fig. 3) to that of as-processed ones (Fig. 2(c) and (d)), indicates that, in addition to grain coarsening, the dislocation density within grains decreases significantly after annealing. It is widely believed that increased dislocation density by extrusion also contributes to hardening. However, the contribution of dislocations to hardening typically comes secondary compared to grain size induced strengthening. This is consistent with the moderate hardness drop, by a few hundred MPa, in extruded T91 after annealing up to 700 °C as shown in Fig. 4. Additionally it is noticed that the slope of the linear fit (indicated by dash line in Fig. 4) for annealed T91 specimens is similar to that before annealing, indicating that the strength of the grain boundaries to prevent the glide of dislocation remains constant.

The current study shows that ECAP of T91 steel can effectively strengthen the alloy by significantly refining the microstructure of ferrites. Further, the microstructure remains in the UFG regime up to nearly 600 °C, making it suitable for use as a structural reactor steel in several applications, pending long-term study. Further studies will be performed to improve the thermal stability of these refined microstructures to even higher temperatures. Additionally, ion irradiation studies are underway to quantify any change in the radiation tolerance of T91 after ECAP processing.

5. Summary

ECAP processing of T91 alloys with a ferritic structure effectively refines the microstructure (grain size) by approximately an order of magnitude, from a few microns to ~350 nm, and strengthens the alloy by up to 70%. Results show that strengthening originates mainly from grain refinement, and moderately from the increase of dislocation density. The microstructure and mechanical strength of ferritic T91 alloys with ultra fine grains are thermally stable up to 500 °C for 10 h, and show little degradation after 10 h of annealing at 600 °C.

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