Original Research

Tubular pure copper grain refining by tube cyclic extrusion–compression (TCEC) as a severe plastic deformation technique

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Abstract

Tube Cyclic Extrusion–Compression (TCEC) method is a novel severe plastic deformation technique developed for grain refining of cylindrical tubes to ultrafine grained (UFG)/nanostructured ones. In this method, tubes are fully constrained and deformed between chamber and mandrel with a small neck zone. The principle of TCEC technique which was adopted to impose severe plastic strains to the tubular materials was explained. Also, the deformation and grain fragmentation mechanism during TCEC was analyzed. The material deformation characteristics during TCEC were numerically simulated by FE code of ABAQUS/Explicit. The FEM results demonstrated that TCEC technique was able to impose extremely high plastic strains. The TCEC method was successfully applied to a commercially pure copper (99.99%) and significant grain refinement was achieved. TEM observation demonstrated the refinement of grains from the initial size of 45 μm to 200–350 nm after four processing cycles of TCEC. Microhardness measurements were carried out across the thickness of the initial and processed tubes. The results show good homogeneity of hardness distribution and an increase to 102 Hv from initial value of 55 Hv after four TCEC cycles. Mechanical properties of the specimens were extracted from tensile tests. The obtained results documented notable increase in the yield and ultimate strengths, whereas the uniform and total elongations decreased. Fracture surfaces after tensile tests were investigated by scanning electron microscopy (SEM), and the observed morphology indicates ductile fracture mode after four cycles of TCEC.

Keywords: Tube Cyclic Extrusion–Compression; Severe plastic deformation; Pure copper; Nano crystalline; Mechanical properties

1. Introduction

Over the last decades, ultrafine grained (UFG) and nanocrystalline (NC) materials have been well recognized as materials with superior and unique mechanical and physical properties. Accordingly, many investigations have been devoted to the severe plastic deformation (SPD) processing of UFG and NC materials [1,2]. The most commonly used SPD methods are Equal Channel Angular Pressing (ECAP) [3], Cyclic Extrusion Compression (CEC) [4]; Accumulative Roll Bonding (ARB) [5]; and High Pressure Torsion (HPT) [6] which have been developed for processing of bulk and sheet materials. Nowadays, demands for UFG and NG tubes necessitate development of SPD techniques for tubes. High Pressure Tube Twisting (HPTT) [7], Accumulative Spin-Bonding (ASB) [8], Tube Channel Pressing (TCP) [9], Parallel Tubular Channel Angular Pressing (PTCAP) [10], Tube High-Pressure Shearing (t-HPS) [11], Repetitive Tube Expansion and Shrinking (RTES) [12], Tube Cyclic Extrusion–Compression (TCEC) [13] and Tube Cyclic Expansion–Extrusion (TCEE) [14] are the SPD methods especially developed for tubular materials.

High-strength and high-conductive materials are the primary need of nowadays electronic industries. Two main strategies for strengthening of engineering metals are (i) through grain refinement and (ii) via adding alloying elements. Despite the alloying approach is more efficient and copper alloys have high strength and are widely used in these industries, their electrical conductivity is inherently lower than pure copper

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because of the alloying atoms. On the other hand, previous researches [15,16] have revealed that severe plastic deformation slightly reduces the electrical conductivity due to the accumulation of lattice defects during severe plastic deformation. But, it increases the strength of pure copper to the range of strength in alloyed ones [16]. Therefore, processing UFG and NC commercial pure copper by severe plastic deformation has been drawn great attentions in last decade [17]. In this regard, in the present study, for the first time, ultrafine grained and nano-crystalline tubes of pure copper were processed by TCEC as a novel SPD technique introduced by the authors [14]. Tubular pure copper was TCEC processed up to four cycles, and the capability of TCEC in grain refinement of pure copper was examined via evaluation of microstructure evolution, variation of mechanical properties and tensile fracture surface morphology.

2. TCEC principles

Schematic illustration of the TCEC method is shown in Fig. 1(a). In the TCEC method, tube is fully constrained from all sides by placing it between mandrel and chamber and then fastening of two end caps. A short thicker cross section devised on the mandrel provides a small neck zone between the mandrel and chamber as shown in Fig. 1(a). The enclosed tube can be processed by pressing the mandrel downward to where all the cross sections of the tubular specimen pass the neck zone indicated in Fig. 1(a). TCEC is a cyclic process in which the cross section of the tube is first extruded from the neck zone and subsequently compressed to initial thickness after passing the neck zone. In order to accumulate severe plastic strains in the tubular material, symmetric design of TCEC processing tools allows repetition of the process by die upside down rotation after each cycle. Fig. 1(b) shows the designed and fabricated tools for the TCEC process. The total accumulated strain after N cycles of TCEC process (εTN) can be obtained using Eq. (1) [13] which was derived from the geometry of tools in Fig. 1(a). The values of total accumulated strain after TCEC processing with one to four cycles are 1.92, 3.84, 5.8 and 7.7, respectively.

\[ \varepsilon_{TN} = N \varepsilon_{t} = 2N \times \left[ \ln \left( \frac{R^2 - r^2}{R_0^2 - r_0^2} \right) + \frac{4}{3} \cot \left( \frac{\phi_2}{2} \right) \right] \] (1)

3. FEM and experimental procedures

The numerical studies of the TCEC process and the deformation during this process were carried out using commercial FE code (Abaqus/Explicit; Simulia). The process was simulated in the 2D axisymmetric model, and the section of the tubular specimen was meshed using axisymmetric four node elements (CAX4R). The geometrical dimensions and mechanical properties of the tubes in the simulation were considered to be the same as those of the experiment. This permits the comparison of the simulation results with experimentally obtained ones. The isotropic rule has been used as plasticity hardening rule of the material behavior. An automatic remeshing (adaptive meshing) method was employed in the simulations to accommodate the imposed large strains and heighten the accuracy of the results. The arbitrary Lagrangian–Eulerian (ALE) adaptive meshing maintains a high-quality mesh under SPD by allowing the mesh to move independently with respect to the underlying material. The material density artificially increased using mass scaling option in order to reduce the computation cost. Errors in the results due to the increased inertia forces was avoided by monitoring of the ratio of the kinetic energy to the internal energy to be quite negligible. This confirms the quasi-static response of the explicit method with applied step time and mass scale. The chamber and the mandrel were modeled as analytical rigid parts. The contact between the tools and the specimen was modeled using the penalty scheme and Coulomb friction with friction coefficient of 0.05.

A series of TCEC experiments were performed to investigate the capabilities of this novel technique in grain refinement. Commercial copper (purity 99.98%) was used in this study. Tubular samples with outer diameter of 20 mm, inner diameter of 15 mm (2.5 mm in thickness) and length of 60 mm were machined from as-received material. A fully recrystallized homogeneous microstructure was obtained by annealing at 600 °C for 2 h. All components of the TCEC tools were fabricated from tool steel and hardened to 55 HRC (Fig. 1b). The major parameters of the TCEC tools shown in Fig. 1a are \( R_0 = R = 10 \text{ mm}, \ r = 8.75 \text{ mm}, \ r_0 = 7.5 \text{ mm}, \ L = 1 \text{ mm} \) and \( \alpha = 30° (\Phi_1 = \Phi_2 = 162°) \). The TCEC experiments were carried
out under a constant pressing speed of 5 mm/min at ambient temperature. Friction on contact surfaces was reduced using MoS2 spray as lubricator. The processed samples were cut perpendicular to the tube axis and the surfaces were subsequently prepared by standard metallographic techniques. Optical Microscopy (OM) and Transmission Electron Microscopy (TEM) were employed to evaluate microstructure and grain refinement phenomenon. The microhardness of the tubes was measured by Vickers hardness machine in a load of 100 gf for a dwell time of 15 s. The mechanical properties of the initial and processed tubes were investigated using tensile tests of samples having dog-bone geometry with the gauge length parallel to the tube axis. The tensile fracture surface of the samples was observed by scanning electron microscopy (SEM).

4. Results and discussion

4.1. FEM

The microstructure evolution during severe plastic deformation is directly related to the accumulated effective strain and its distribution. Therefore, the numerical analysis of the strain accumulation and distribution is of significance \[18,19\]. The distribution of equivalent plastic strain (EPS) for the TCEC processed tubes after one to four cycles are shown in Fig. 2. As can be seen, the results indicate good homogeneity of EPS values across the thickness and in the whole of the processed tubes. Fig. 3 shows the distribution curves of the EPS values across the thickness of the tubular specimens. The average equivalent plastic strains of 2.1, 4, 6 and 7.9 were achieved after one to four cycles of TCEC, respectively.

Fig. 4 illustrates simplified triangular slip line field of the deformation during extrusion half cycle of the TCEC process. Among the three specified deformation regions in Fig. 4(a), region 2 where the material undertakes continuous plastic strains is the main deformation region. In Fig. 4(a), \( v_1 \), \( v_2 \), \( v_3 \equiv (T/T) v_1 \) are respectively the velocity of material in regions 1–3. Also, \( E_1 \) and \( E_2 \) are the beginning and end surfaces of the main deformation zone. As can be seen in Fig. 4(a), due to the material flow continuity during deformation, the normal components of the material velocities for an arbitrary point on the entry surface (\( E_1 \)) and the corresponding point on the exit surface (\( E_2 \)) must be equal \( (v_{1n} = v_{en} \text{ and } v_{3n} = v_{en}) \). Nonetheless, the values of tangential velocities on the entry and exit surfaces (\( E_1 \) and \( E_2 \)) might be different \( (v_{1t} \neq v_{et} \text{ and } v_{3t} \neq v_{et}) \). This difference, in turn, leads to velocity discontinuities on the \( E_1 \) and \( E_2 \) surfaces. The velocity discontinuity brings about shear stresses along the boundary surfaces \[20\] as shown in Fig. 4(b). The resultant shear stresses are the main causes of the grain fragmentation during TCEC processing. This analytical analysis of deformation was also investigated in the FE simulations. Fig. 4(c) demonstrates the contour of plastic shear strains during TCEC processing. Fig. 4(d) shows iso-surface contour of the resulted shear strains during the TCEC method. As shown in Fig. 4(d), there is good agreement between the numerical and analytical analysis.

4.2. Experiment

4.2.1. Microstructure evolution

Fig. 5 shows the fully recrystallized homogeneous microstructure of the initial material with a mean grain size about 45 μm.

![Fig. 2. TCEC processed specimens (a) during first cycle and after (b) first cycle, (c) second cycle, (d) third cycle, (e) fourth cycle.](image-url)
In order to investigate the microstructural evolutions after TCEC processing through one to four cycles, the evolved microstructures were observed by OM and the micrographs are shown in Fig. 6. As illustrated in Fig. 6(a), the microstructure of the specimen after one TCEC cycle was substantially evolved and is notably different from the initial microstructure of the as annealed copper. The microstructure evolution and grain refinement in the processed material is somewhat indiscernible after the second cycle of TCEC. Nonetheless, it is readily seen that processing with higher cycle numbers of TCEC results in equiaxed grains with sub-micrometer sizes (Fig. 6(b–d)). In order to further examine the grain refinement level after TCEC processing, the microstructure of the four cycle processed specimen was also observed using TEM. The TEM micrograph and the selected area diffraction (SAD) pattern of the specimen after processing with four TCEC cycles is shown in Fig. 6(e). It has been well demonstrated that the materials with medium to relatively high stacking fault energies (SFE) like Cu experience refinement of coarse grains via dislocation activities upon continued severe plastic deformation. The grain refinement phenomenon in copper due to TCEC processing includes the following steps [21]. Material straining in its initial steps manipulates dislocations in the
structure. The relatively high mobility of the dislocations results in tangling of the dislocations and consequently formation of dislocation cells. Therefore, the structure transforms to a structure containing high and low dislocation density regions. Further straining increases the density of dislocations in the cell walls and gradually transforms these dislocation walls to the low angle boundaries which subdivide the coarse grains into an intra-granular refined sub-grains. When the deformed material crosses the velocity discontinuity surfaces, the formed subgrains undertake the tangential (shear) stresses at the subgrain boundary level and the various subgrains experience different rotations during severe plastic deformation. That is to say, different intra-granular sub-grains within a grain tends to rotate towards different stable end orientations [22]. Accordingly, the imposed shear stresses to the material on the velocity discontinuity surfaces disorient the sub-grains and gradually transform low angle dislocation walls to the conventional high angle grain boundaries. This process gradually fragments original coarse grains and refines the microstructure to ultrafine grained and nano-crystalline one. The UFG and NC material is the consequence of the gradual transformation of the preceding bimodal substructure including cells and cell walls. Severe plastic deformation progressively increases dislocation density in the subgrain walls. Shearing and sliding of the sub-grain boundaries occur because of dislocations with Burgers vectors lying in the boundary plane; on the other hand, other dislocations increase boundary misorientation in accordance with their combined Burgers vectors [23]. Therefore, the strain accumulation increases misorientation of the sub-boundary to where the conventional high angle boundaries at large strains are formed. As expected, Fig. 6(e) demonstrates that after processing with four cycles of TCEC, the initial microstructure with grain size of 45 μm was remarkably refined and grain size of 200–350 nm was achieved. It is worth mentioning that the dispersed selected area diffraction (SAD) pattern of the processed copper in Fig. 6(e) demonstrates increase in the number of high angle boundaries [24,25].
4.2.2. Mechanical properties

Microhardness variations after TCEC cycles were documented in diagrams of Fig. 7. Fig. 7(a) illustrates bar diagram of the microhardness values at the cross section of the tubular specimens. As shown in Fig. 7, the values of microhardness are remarkably affected by the number of TCEC cycles especially after the first and second cycles. The microhardness values increase with increase in the number of processing cycles as a result of grain refinement. The increase of microhardness values is due to the increase of boundary and boundary misorientation, formation of subgrains and dynamic recrystallization during severe plastic deformation [26,27]. As can be seen in Fig. 7 diagrams, with increase in the number of TCEC cycles the microhardness values gradually lie close to a saturation level. In other words, at early stage of straining with the TCEC process, the hardness increases by increase in the amount of the accumulated strain but gradually levels off and approaches to a steady-state level. Dynamic recovery phenomenon is the main cause of this trend [28]. Dynamic recovery prohibits the steady rate of microstructure refinement and grain size reduction. Dynamic recovery annihilates dislocations and its rate is proportional to the dislocation density in the deformed material. Therefore, further straining increases the rate of dynamic recovery and consequently results in decrease in the rate of grain refinement at large strains. The results of microhardness measurement across the thickness of the tubes are shown in Fig. 7(b). These results demonstrate good homogeneity of microhardness distribution through thickness but the close examination reveals that the values of microhardness slightly increased from outer surface of the tubes toward the inner surface in the early cycles while the microhardness homogeneity across tube's thickness increased by increase in the number of TCEC cycles. Tubular specimen’s initial microhardness of 55 Hv increased to 80, 93, 99 and 102 Hv after one to four cycles of TCEC process, respectively. The peripheral microhardness of the processed specimens was also measured in the middle of the tubes thickness and the results are shown in Fig. 7(c). There is good homogeneity in the peripheral microhardness values.

Fig. 8(a) shows engineering stress–strain curves of initial pure copper material and the TCEC processed tubes by one to four TCEC cycles. As illustrated in Fig. 8(a), after processing by TCEC, yield strength (YS) and ultimate strength (US) notably increased, while the total elongation decreased. The major characteristics of the stress–strain curves are summarized in the diagrams of Fig. 8(b), which documents the variations of yield and ultimate strength, elongation to failure and uniform elongation. Fig. 8(b) demonstrates that the yield and ultimate strengths increase with increase in the number of TCEC cycles, and at higher strains gradually level off and lie close to saturation values [28,29]. This trend is analogous to the microhardness variations illustrated in Fig. 7. Fig. 8(b) also
illustrates the variation of elongation to failure versus the numbers of TCEC cycle. The total elongation to failure decreases with increase in the number of TCEC cycles while it demonstrates slight increase at the forth processing cycle (equivalent strain of ~8). Dynamic recovery and formation of dynamically recrystallized grains are the main reason for approaching to a saturation level for yield and ultimate strengths and at the same time slight enhancement in the total elongation value. It is worth mentioning that after processing by TCEC, the uniform strain (the strain hardening region) in the stress–strain curves was significantly reduced and is very limited. That is to say, the main plastic deformation during tensile tests occurs in the instable plastic zone [30] and necking is seen in initial stages of the tensile deformation. Nonetheless, an increase in the value of uniform strain after processing by four TCEC cycles was recorded.

4.2.3. Fracture surface

Fig. 9 shows the fracture surface morphology of the tensile samples in the as-annealed state and after processing by four TCEC cycles. It was observed that all the tensile samples exhibit necking, and there are some slip bands on the surface of the tensile samples. As explained in Section 4.2.2, the uniform elongation of TCEC processed specimens is very limited. This leads to very little work hardening and consequently immediate necking in tensile tests. The SEM observations demonstrate a typical ductile fracture with a large number of dimples in the fracture surface of the tensile samples. The dimples in the fracture surface of the annealed specimen are chiefly deep and large in diameter, as illustrated in Fig. 9(a). It is worth mentioning that there are also some shallow and small dimples among the large ones. After TCEC processing, the great reduction in the uniform elongation and work hardening ability of the material results in smaller and shallower dimples due to not having enough time for nucleated dimples to grow and assemble with other dimples. Fig. 9(b) illustrates the fracture surface of the four cycle processed specimen including very small and shallow dimples. The decrease in the dimple size is attributed to the grain refinement in the structure of the TCEC processed tubes [31,32].

5. Conclusions

For the first time, Tube Cyclic Extrusion–Compression (TCEC) as a novel SPD technique for tubes was utilized for processing ultrafine grained and nano-crystalline tubular pure copper. Tubular specimens of pure copper were processed by one to four TCEC cycles and the capability of TCEC in grain refining of commercially pure copper was investigated by finite element simulation and experimental observations and measurements. The principle of TCEC was explained, and the grain fragmentation mechanism during TCEC was analyzed. TCEC of pure copper was numerically simulated by FE code of ABAQUS/Explicit and the main characteristics of deformation during TCEC were extracted, which indicate that TCEC technique imposes extremely high plastic strains (~2) to the material after each cycle. The experimental results demonstrate
that the tubular specimens of pure copper were successfully processed by one to four cycles of TCEC. The microhardness measurements indicate that the initial microhardness of 55 Hv in the as-annealed state increased to 102 Hv after application of four TCEC cycles. In addition, there is a good homogeneity in the peripheral microhardness and the microhardness across thickness of the processed tubes. Optical observations documented remarkable grain refinement after TCEC processing. TEM micrograph of the specimen processed by four cycles of TCEC demonstrates refinement of microstructure to 200–350 nm from the initial grain size of 45 μm. Mechanical properties of the initial and TCEC processed specimens were extracted from the tensile tests. The results demonstrate significant increase in yield and ultimate strengths and remarkable decrease in the uniform and total elongations. SEM observations of the tensile fracture surfaces indicate ductile fracture mode in the processed pure copper after application of four TCEC cycles.

References