ULTRAFINE GRAINED ALLOYS PRODUCED BY SEVERE PLASTIC DEFORMATION: ISSUES ON MICROSTRUCTURAL CONTROL AND MECHANICAL BEHAVIOUR*

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Abstract

A review is presented about recent results obtained on properties of ultrafine grained structural alloys and pure metals processed by severe plastic deformation. Carbon steels, aluminium alloys and pure nickel were investigated after processing by Equal Channel Angular Pressing. Data on microstructural evolution, aging kinetics, mechanical properties and recrystallization texture are discussed to draw a picture of advantages and drawbacks of such ultrafine materials.

Riassunto

L'articolo presenta alcuni recenti risultati sulle proprietà di leghe e metalli puri a struttura ultrafine ottenuti per deformazione plastica severa. L'affinamento è stato ottenuto su acciai al carbonio, leghe di alluminio e nichel puro mediante tecnica Equal Channel Angular Pressing. Vengono presentati e discussi i risultati sull'evoluzione microstrutturale, sulla cinetica di invecchiamento, sulle proprietà meccaniche e sulla tessitura sviluppata dopo ricristallizzazione dei diversi materiali per delineare un quadro generale su vantaggi e svantaggi caratteristici dei metalli a struttura ultrafine.

INTRODUCTION

Severe plastic deformation (SPD) is currently one of the most promising methods for processing ultrafine microstructures. Ultrafine-grained materials generally offer high strength coupled with reasonably good fracture toughness and often superplastic properties at moderate temperatures and at high strain rates [1-6]. Especially, the low-strength commercially pure and low-alloyed metals can be dramatically enhanced by extensive grain refinement, thus offering new opportunities to exploit some attractive physical properties in metals having a reasonably high strength for structural applications.

Fundamental research works on grain refinement by intense plasticity published in recent years focussed on the mechanisms that effectively reduce grain size down to submicrometer size in metals. Some general features appear to hold for policrystalline fcc and bcc metals having medium and high stacking fault energies such as Al, Cu, Ni and Fe deformed in the α-phase temperature field. Interestingly, these aspects are observed both in cold and hot deformation conditions [7-10].

For low to medium strain levels, cell blocks initially develop. These microstructural features have high-angle grain boundaries (HAGBs) often parallel each other and separate regions within which the dislocations are not randomly distributed but accumulate into cell boundaries with low-angle misorientation. The cell boundary orientation initially follows specific crystallographic planes related to slip directions but with increasing strain, it becomes oriented along macroscopic planes or assumes a fibrous arrangements depending on the specific macroscopic plastic flow induced by the deformation process. At still higher strains the cell block spacing decreases while the misorientation angle both across blocks and across single cells increases. This so-called process of grain subdivision is such that single cells or groups of cells (namely portion of grains) can rotate toward different orientation to create a set of new small equiaxed grains with HAGBs, thus generating an ultrafine structure.

It was also stated that grain subdivision is markedly accelerated if localized shearing or local migration of the boundaries can occur (i.e. local HAGB contact normal to the elongation direction assisted

*This paper was presented at the "nd International Conference & Exhibition in Metallurgical Process Technology", Riva del Garda 19-21 Sett. 2004 by high temperature diffusion). Significant structural changes can also occur if the strain path is changed (e.g. by cross-rolling) or if non-deformable second-phase particles can cause inhomogeneous deformation [7,8].

The above described mechanism, observed in many deformed metals and governed by a continuous reduction of cell block size with increasing strain followed by grain subdivision, has been a key factor stimulating a large number of research programmes in recent years. The evolution of materials subjected to ultra-high strains (severe plastic deformation – SPD) is currently one of the most attractive research subject owing to the opportunity of generating ultrafine or even nanostructured bulk metals and alloys. Several studies are focussed on conventional processes and straining modes (e.g. rolling and plain strain conditions) [11-13] with the aim of achieving a substantial grain refinement in commercial products. However, most of the methods for the synthesis of ultrafine materials rely on laboratory processes by which almost unlimited strains can be introduced in ductile metals such as equal channel angular pressing (ECAP), accumulative roll bonding (ARB) and high pressure torsion straining (HPTS) [1,3,14,15].

Ultrafine and nanostructured bulk metals possess a number of peculiar properties still requiring deeper investigation to be fully understood. The present paper is aimed at presenting some activities within this field, that are currently in progress at the research groups of the authors. Results on Al alloys, low-C steels and pure Ni deformed by ECAP will be presented giving particular emphasis on the properties achieved by refining the materials to the ultrafine grain scale.

EXPERIMENTAL PROCEDURES FOR ECAP PROCESSING

The facility for ECAP processing the samples at room temperature is based on a system of two symmetrical half die machined from two blocks of H11 tool steel, heat treated to achieve a nominal hardness of 45 HRC. Cylindrical channels having a diameter of 10 mm were machined with a configuration characterised by angle values of ϕ=90° (channel intersection angle) and ψ = 20° (external curvature angle). In Figure 1 the geometry of the two halves of the die are depicted. Previous practical experiences had shown that a calibrated length of 10 mm after the corner followed by an enlargement of the exit channel could reduce friction and thus lower the pressing load. With the aim of further reducing friction and increasing wear properties, once the channels had been machined and heat treated, the die was ion nitrided.

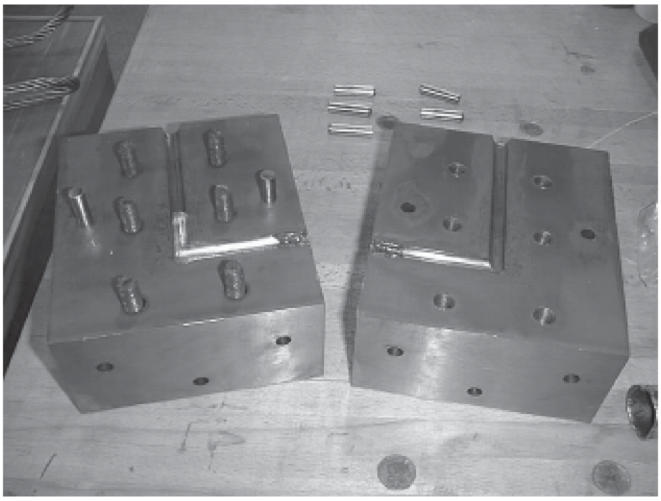

Fig. 1: Geometry of the two halves of the die used for ECAP processing the materials at room temperature. A sample of an interrupted test is shown at the region of intersection of the channels.

The two half dies are assembled and held together with 6 bolts having a diameter of 20 mm. The outer dimensions of the assembled die are approximately 160x160x185 mm^3. A plunger was obtained by the design of a dedicated fixturing device to be installed on the load cell, so as to record plunger load and displacement during pressing. A centering device is also used to precisely center the die with respect to the plunger, before each testing session.

The samples for ECAP testing are coated with a lubricant based on molybdenum disulphide. Several specimens are pressed consecutively and repeatedly to accumulate the desired plastic deformation. Samples processed with the so called route C (rotation by 180° of the sample between each pressing) were generally produced [16]. For alloys requiring high-temperature pressing, a second die was designed with an angle configuration of ϕ = 110° and ψ = 20°. This die is equipped with four electric resistance heaters surrounding the entry channel and the corner between the channels. The heating system is controlled by a termocouple located in the die close to the corner and allows to maintain stable testing temperatures up to 400°C. A cooling circuit was also set for both the centering device placed beneath the die and for the plunger installed on the load cell.

The theoretical deformation accumulated by passing through the die can be estimated by the Iwahashi equation that considers ideal conditions of simple shear and homogeneous deformation over the cross section [17], being N the number of passes and ϕ and ψ the above defined angles.

$$\varepsilon_N = \frac{N}{\sqrt{3}}\left(2\,co\left(\frac{\Phi}{2}+\frac{\Psi}{2}\right)+\Psi\,cos\,ec\left(\frac{\Phi}{2}+\frac{\Psi}{2}\right)\right) \qquad (1)$$

According to equation (1), the theoretical strain experienced by the specimens pressed into the dies described above corresponds to 1,05 and 0,63 for each pass, for $\phi = 90°$ and $\psi = 120°$, respectively.

RESULTS AND DISCUSSION

Aluminium alloys

Commercially available extruded bars of a 6082 Al-Mg-Si alloy having a diameter of 10 mm were investigated. Samples of 100 mm length were cut from the bars and subjected to thermal treatments. Two different tempers were selected for the present investigation. A first set of samples was solution treated (530°C for 2 hours and water quenching) whereas a second set of samples was studied in the annealed condition (530°C for 2 hours and slow furnace cooling).

In figure 1 a set of TEM micrographs showing the evolution of the grain structure at increasing strain (i.e. number of passes) is given [18]. The original grain size of the solution treated alloy was of the order of 10 mm. After the first ECAP pass, a set of parallel bands of cells a few hundreds of nanometers in width formed. By increasing the number of passes, the sub-boundary misalignment across single cells increased (as inferred by SA diffraction pattern analysis, not shown here). Eventually, grain subdivision and further increase of cells misalignment led to an ultrafine equiaxed high-angle grain structure, with an average size of 300 nm. The observed structural refining process is in full agreement with established general

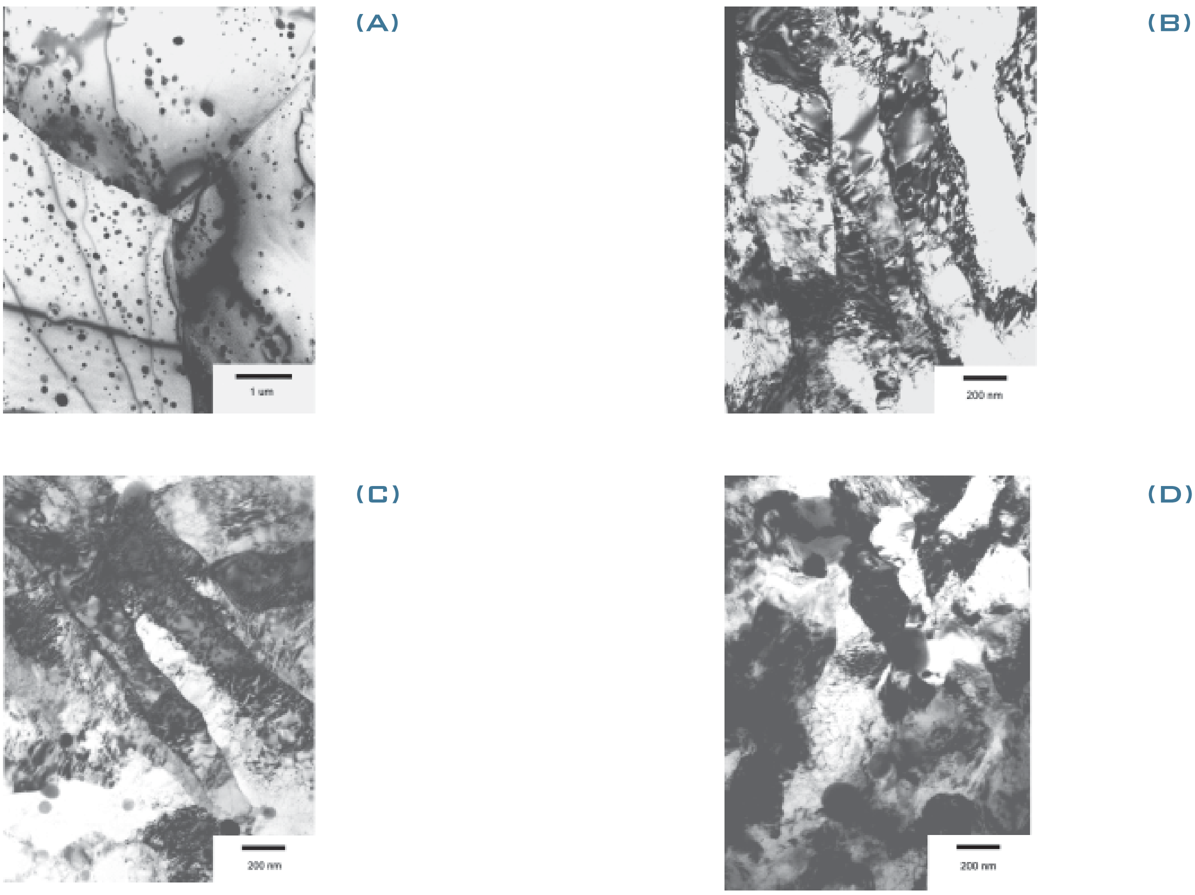

Fig. 2: Grain structure evolution of the the 6082 alloy as a function of ECAP passes: (a) solution treated sample; (b) 1 pass; (c) 2 passes; (d) 6 passes [18].

theories on severe plastic deformation and match other literature results on pure Al and Al alloys tested at room and at moderately high temperatures [14,19-21].

One of the main issues studied by the present authors is the aging behaviour of severely deformed Al 6082 alloy, with particular attention to precipitate evolution and transformation kinetics. DSC analyses on solution treated and ECAP processed samples showed marked differences in endo-and exothermic peak positions and shapes with respect to the unprocessed alloy. The peaks for metastable strengthening β" and β' precipitates were markedly anticipated and the formation of stable β phase was partially suppressed in the alloys processed to 4 and 6 ECAP passes, presumably due to anticipated precipitation of Si-rich particles [18].

Post-ECAP aging behaviour was investigated by isothermal treatments at 130, 160 and 180°C. An estimate of the alloy strength evolution during aging was performed by microhardness profiles, as shown in figure 3.

It was demonstrated that even a single ECAP pass significantly accelerates the aging kinetics, especially at the lowest aging temperature of 130°C (see figure 3a). For the alloy processed to six ECAP passes, the aging kinetics at 160 and 180°C became so fast that a peak of hardness was hardly visible in the aging curves. It was suggested that

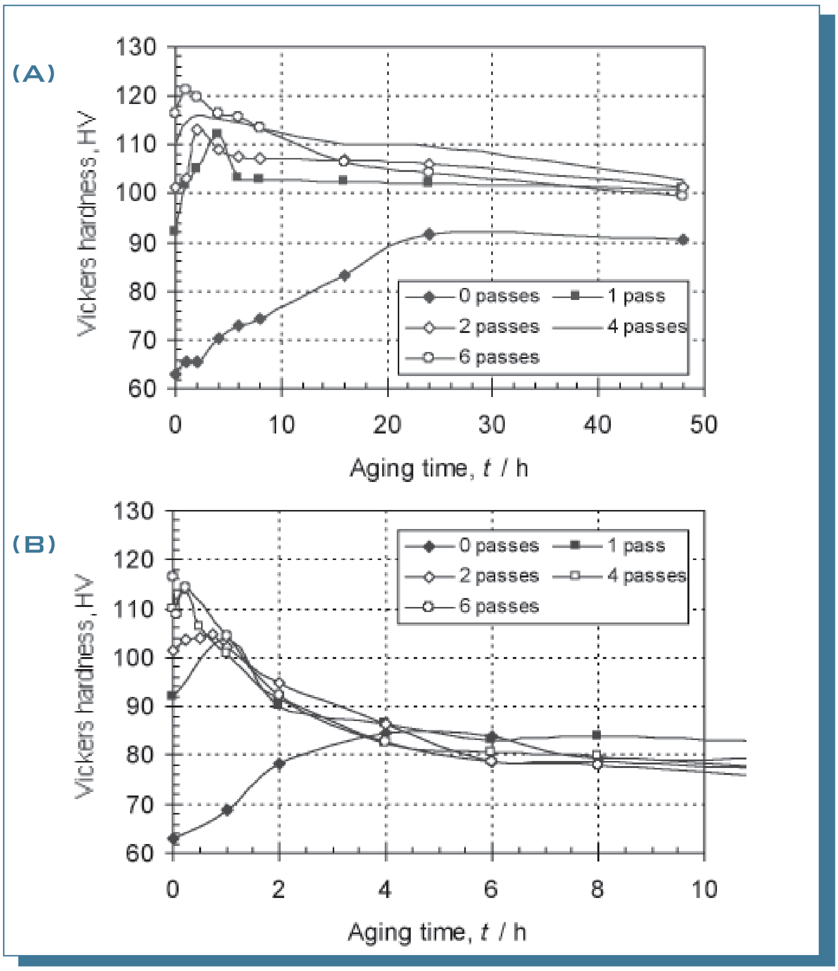

Fig. 3: Aging behaviour at 130°C (a) and 180°C (b) of the solution treated and ECAP processed 6082 alloy.

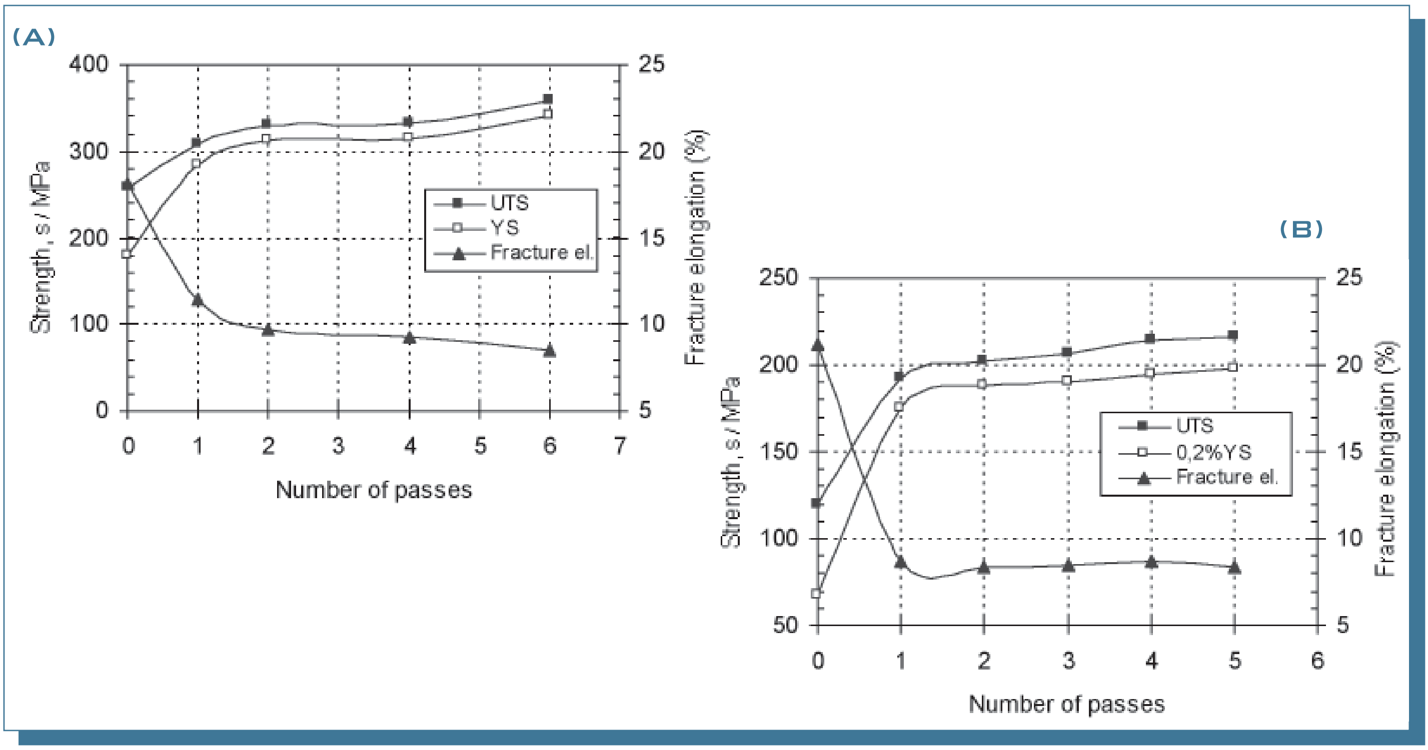

Fig. 4: Tensile properties of the 6082 alloy. (a) solution treated, ECAP processed and aged; (b) annealed and ECAP processed.

rapid overaging phenomena as well as restoration of the severely deformed structure contribute to this process at 160 and 180°C.

Aging at 130°C of the SPD processed alloy revealed to be more effective by virtue of the comparatively slower kinetics and of the limited overaging effects, suggesting a reasonably high structure stability at this temperature. From figure 3a it can also be noticed that the precipitate strengthening generated after post-ECAP aging (i.e. the increase in hardness from aging time 0 to peak hardness time) becomes of lower importance by increasing the amount of severe plastic deformation experienced by the sample [18]. On the basis of the above data, the aging times corresponding to peak hardness at 130°C were established and adopted to age ECAP processed billets for mechanical property evaluation.

Figure 4 is a summary of the mechanical properties achieved in the above described solution treated, ECAP processed and post-ECAP aged samples. For comparison purposes, fully annealed and ECAP processed samples were also considered in figure 4b. Tensile tests were carried out at room temperature, at an initial engineering strain rate of $6{,}7\cdot10^{-4}s^{-1}$. The tensile specimens were machined from the processed billets with a gauge length of 30 mm and a diameter of 6 mm.

From the tensile data it is inferred that for both temper conditions, ECAP processed materials underwent a dramatic increase in strength by the first pressing. After the second pass, a tendency toward saturation of properties readily occurs. As expected, fracture elongation dropped significantly on the first ECAP pressing but ductility still kept at a satisfactory level, slightly below 10%, even at the sixth ECAP pass.

Nickel

The effect of SPD on pure Ni was investigated in order to evaluate the possibility of inducing a preferred orientation in deformed and subsequently annealed samples. Ni is considered as a medium staking fault energy metal therefore, under severe deformation conditions, a grain refining behaviour similar to Al is expected [22]. Interest on texture evolution of deformed and recrystallized Ni is recently growing due to the use of pure Ni and his non-magnetic alloys as a substrate for superconductor materials [23-25]. Published researches allowed demonstrating that recrystallization texture of heavily cold rolled Ni can be controlled to a large extent, obtaining even a strongly developed cubic texture [26-27]. In fact, fcc metals can form a very sharp biaxially cube orientation by a recrystallization heat treatment after heavy cold rolling to strains exceeding 90%. This production route is called RABITS procedure (Rolling Assisted Biaxially Textured Substrate) and it has been successfully applied for the production of II generation HT_C superconductors.

Electrolytic Ni was Plasma Arc Melted (PAM) to produce bar shaped samples (prisms having a length of 250mm and a 45x35mm section) that were cold rolled to a diameter of 12 mm. The samples were then machined to a size suitable for ECAP pressing and finally annealed at 550°C for 2 hours. In figure 5 the representative microstructure of the samples is depicted. It can be observed that the structure is made up of rather coarse equiaxed grains homogeneous in size (110 μm).

ECAP tests were performed at 200°C to reduce the load required for pressing and to increase plasticity of the samples. The die with channels intersecting at 110° was used, selecting a plunger pressing speed of 0,5 mm/min. The samples were pressed following either route C (rotation by 180° of the sample between each pressing) or route A (no rotation of the sample between each pressing). In the former case, billet failure occurred by diffused shear cracking after 4 passes whereas, in the latter case, the billets were pressed for up to 7 passes without any sign of damage.

For a constant strain path such as that induced by route A, the shear strain simply builds up in one plane. In route C, the 180° rotation results in a reversion of the shear strain every alternate cycle, leading to a fully redundant strain after an even number of extrusions [19-21]. It is therefore expected that the samples pressed according to route A should develop a progressive elongation of grains or cell blocks most effectively. This would possibly lead to a more defined texture of the grains. On the contrary, route C theoretically restores the grain shape on every reversion of strain (two passes). However, it was also demonstrated that by route C the development of LAGB cells into small equiaxed grains occurs more rapidly than by route A [19].

Billets processed to increasing number of ECAP passes were annealed at 700°C for 1,5 hours to promote recrystallisation. Samples for microstructural and texture analyses were then obtained by cutting the billets along a plane normal to billet axis.

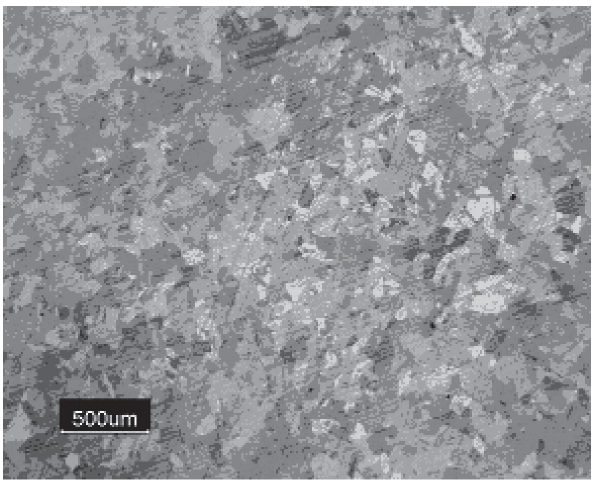

Fig. 5: Optical micrograph of the annealed Ni samples.

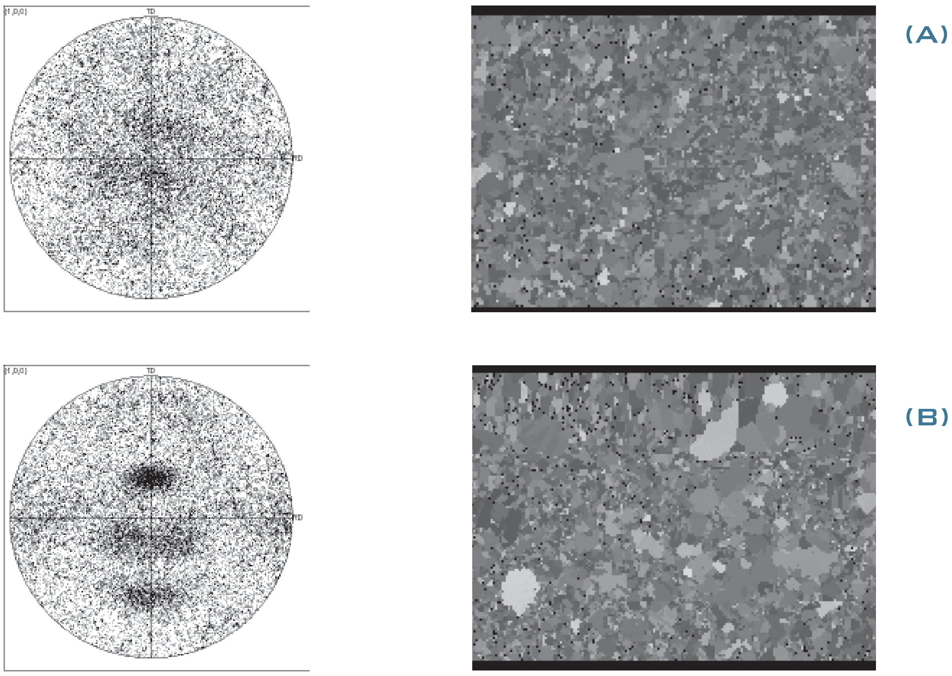

Fig. 6: Polar figures and the orientation maps of ECAP samples processed to four passes and annealed; (a) route C; (b) route A.

EBSD (Electron Back Scattered Diffraction) data are presented here to depict the effectiveness of SPD in modifying the recrystallisation grain structure and texture of pure Ni. Figure 6a shows the polar figure and the grain orientation map of a sample processed according to route C to four passes and then annealed. Figure 6b is the corresponding figure for a similar sample deformed according to route A.

From the preliminary data presented in this study, it can be supposed that route A pressing to four passes would be less effective in refining the Ni structure and/or keeping a small size of grains after annealing. However, it is also noticed from figure 6b that route A structure is composed by a number of coarse recrystallised grains that show a texturing effect, even if not well developed. Deeper investigations are currently in progress to better elucidate this behaviour and to explore different processing parameters in order to sharpen the observed differences in texture evolution.

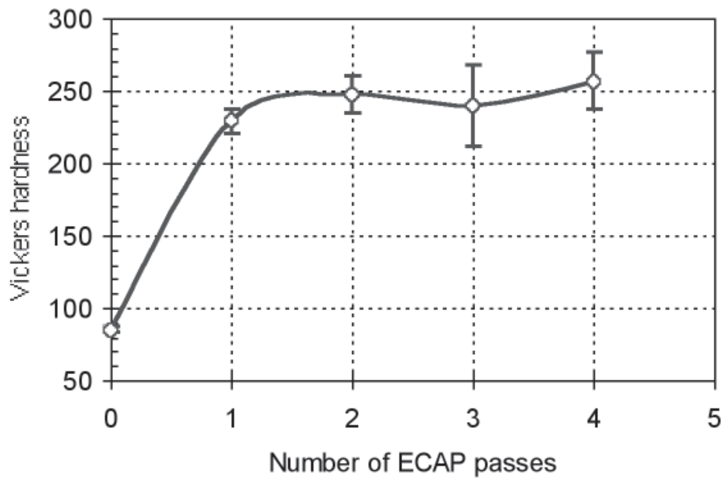

Fig. 7: Microhardness evolution of the 0,05%C steel as a function of number of pressings at 300°C.

Carbon steels

A plain low carbon steel containing 0,05%C, 0,27%Mn and 0,04%Si was investigated with the aim of evaluating the possibility of effectively refining ferrous alloys by ECAP.

Cylinders of diameters 10 mm were cut from the as delivered drawn bars and fully annealed at 900°C for 1,5 hours. ECAP tests were carried out at 300°C with the 110° angle die and a plunger pressing speed of 3 mm/min.

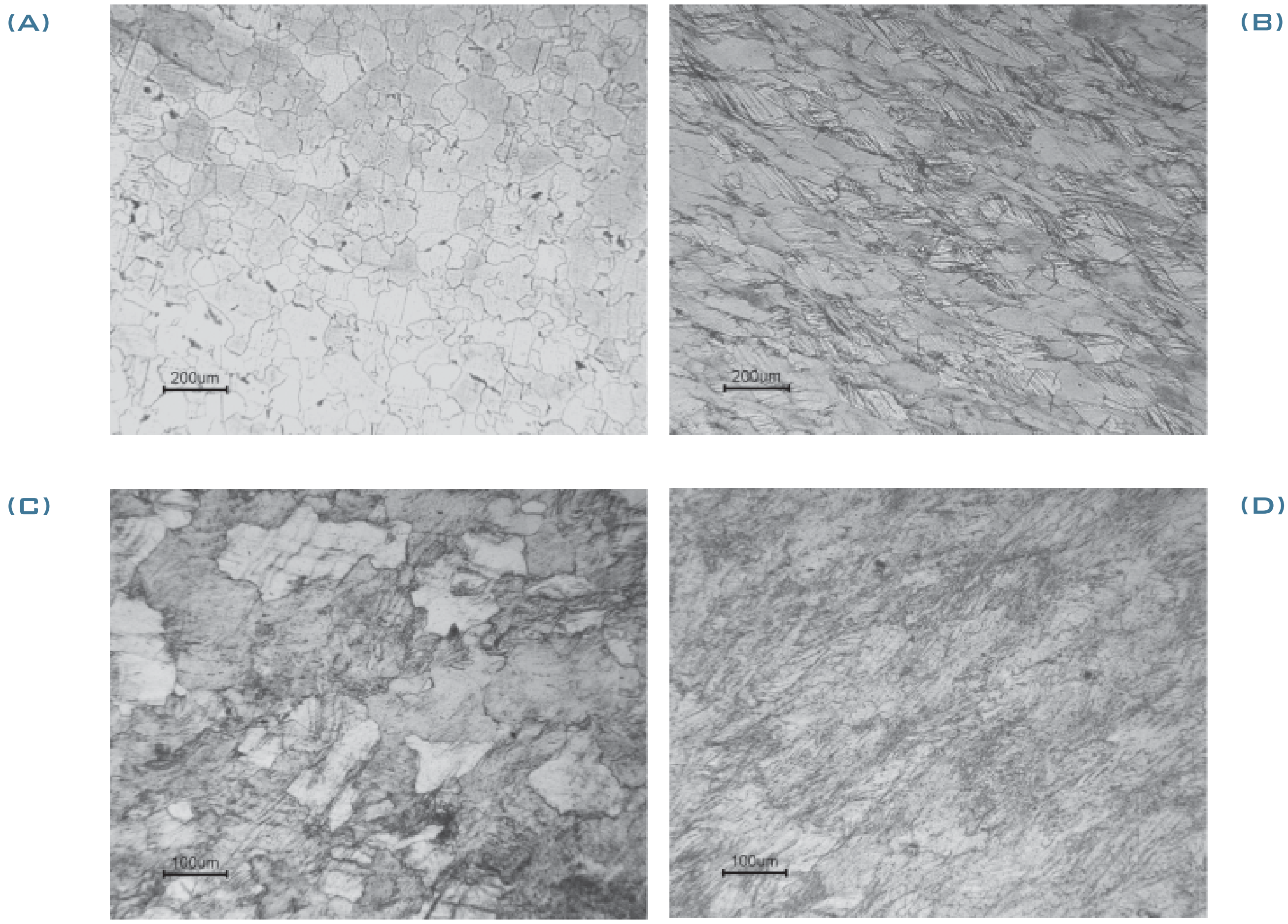

Fig. 8: Optical micrographs of the 0,05%C steel pressed at 300¡C by ECAP. (a) annealed structure not deformed; (b) 1 pass; (c) 2 passes; (d) 4 passes.

By adopting route C, it was possible to produce samples pressed up to four times before extensive cracking occurred. Microstructural analyses by optical and SEM microscopy as well as by EBSD were then carried out to elucidate some features about SPD of low carbon steels.

Vickers microhardness testing on longitudinally sectioned billets was also adopted as a simple means of evaluating mechanical property rise as a function of ECAP pressings. Figure 7 depicts the average measured data with their experimental scatter. A trend similar to that already reported for the strength of the 6082 Al alloy is obtained. Also for the severely deformed steel, a steep rise in hardness is observed after the first pressing, with a subsequent near-saturation of properties with increasing number of ECAP passes.

In figure 8 typical optical micrographs of the steel structure are collected as a function of strain experienced by ECAP. The original coarse equiaxed ferritic grain structure of the as annealed steel (figure 8a), after the first pressing appears to be extensively deformed into parallel bands of grains (figure 8b) roughly oriented along the macroscopic shear direction induced by the die geometry. Traces of slip planes inside the grains are mostly visible in particular grains, due to the possible effect of the metallographic etchant.

Inspection of the structure deformed by two passes (figure 8c) confirms that an approximately equiaxed grain structure is restored owing to the reversion of strain. However the contour of the grains does not appear to be well defined as in the original structure. In addition, the traces of crossed slip observed in the coarser grains suggest that during the second pressing with inverse strain direction, the steel does not exactly follow the original microstructural strain path, but new slip systems became activated. After three and four pressings (figure 8d) the structure develops fine features not readily resolvable at the optical microscope. The original grain contours were not preferentially etched and only faint races of the complex plastic flow are inferable.

More accurate SEM observations revealed that the brittle Fe_3C phase contained in the pearlite islands and as grain boundary cementite underwent

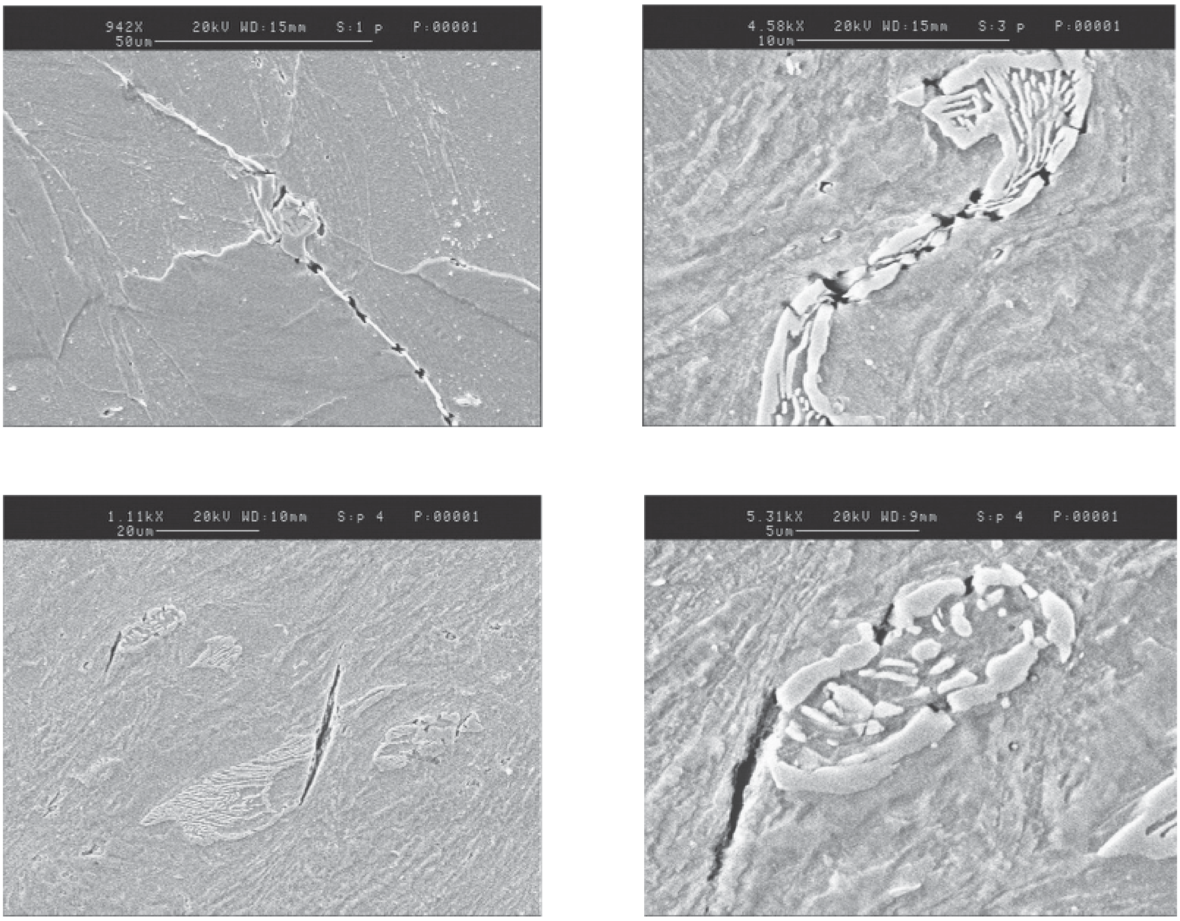

Fig. 9: SEM micrographs showing microstructural details of the ECAP pressed samples. (a) 1 pass; (b) 3 passes; (c and d) four passes.

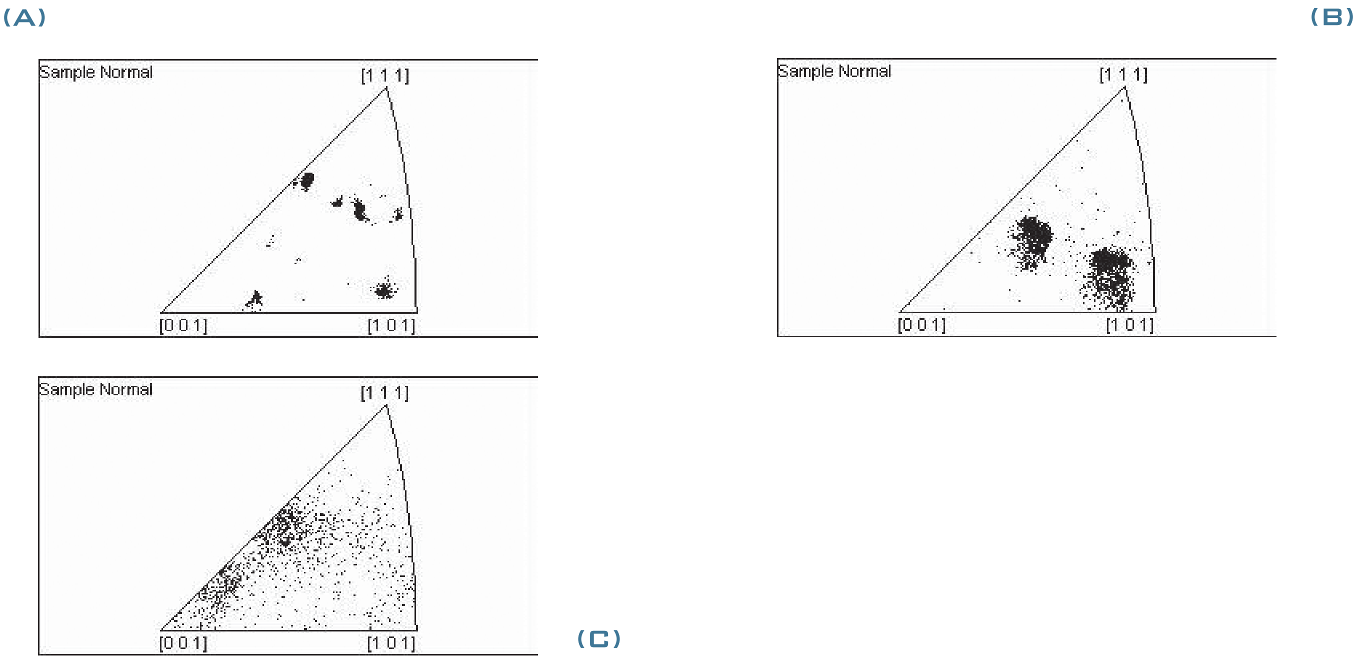

Fig. 10: Inverse polar figures of a selected area covering only a few grains of the 0,05%C steel processed at 300°C. (a) original texture; (b) after 1 pressing; (c) after four pressings.

diffused cracking during severe plastic deformation, as shown in figure 9. However, the strong hydrostatic compression stress component associated to ECAP pressing is believed to prevent such cracks to propagate inside the ferrite phase, at least up to a defined degree of imposed strain that corresponded to about three pressings (equivalent to a total strain of 1,89) in the present case. From figure 9c and 9d, it clearly appears that after this threshold, cracks tend to propagate into the ductile ferritic matrix starting from the above described brittle regions.
High-angle grains and low-angle cell development in the severely deformed steel is currently under investgation by coupled EBSD and TEM analyses. In figure 10 a first relevant result is reported in terms of modification of the inverse polar figures pertaining to a limited number of representative grains in the structure. In the as annealed structure analysed at a magnification of the order of 2000X, a few grain are selected and represented by well defined spots in the inverse polar figure due to uniform crystallographic orientations within the single grains (figure 10a). With increasing straining (figures 10b and 10c), it can be clearly noticed that the extension of the spots markedly increases due to growing misorientation progressively accumulating within the single grains (or cell blocks). After four pressings, the structure is supposed to consist of small HAGB subunits having a submicrometer size. Such structure cannot be easily resolvable by the EBSD system fitted to a conventional SEM and gives rise to a broad scatter in the orientation maps.

CONCLUSIONS

A series of experimental results about Al alloys, steel and pure Ni has been presented to highlight some promising issues of severe plastic deformation. The technique of ECAP, as for several other laboratory processes developed to produce significantly large strains in metals, revealed to be a valuable tool for fundamental research on structural and mechanical properties evolution as a function of process conditions.
It is well accepted that general theories hold for structure development in metals subjected to large deformations both at room and at moderately high temperatures. Through these theories it is possible to envisage processing conditions leading to extensive structure refinement, down to submicrometer grain size levels, into the field of nanostructured metals.
At this stage, efforts should be addressed to upgrade this knowledge toward the processing of commercial wrought products such as plates and rods. A first step would consist in translating the straining conditions suitable to effctively refine metallic alloys to conventional rolling and drawing processes, even considering the intrinsic limitation related to the maximum strain bearable by the materials before failure during processing.
A second line of research is the adoption of modified processes, based on those currently used in research laboratories, well suitable to impart severe deformation to metals without damage. It must be recalled that such processes should possess high productivity and production flexibility to successfully face the market competition. This latter will probably be the main point that will state the real development chances of ultrafine metallic alloys.

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