

## Low-temperature volume radiation annealing of cold-worked bands of Al-Li-Cu-Mg alloy by 20-40 keV Ar<sup>+</sup> ion

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**Abstract.** The processes of radiation-dynamic nature (in contrast to the thermally-activated processes) in the course of short-term irradiation of 1 mm thick bands of cold-worked aluminum alloy 1441 (of system Al-Li-Cu-Mg) with Ar<sup>+</sup> 20-40 keV were studied. An effect of in-the-bulk (throughout the whole of metal bands thickness) low-temperature radiation annealing of the named alloy, multiply accelerated as compared with common thermal annealing processes was registered (with projected ranges of ions of considered energies definitely not exceeding 0.1 μm). The processes of recrystallization and intermetallic structure changes (occurring within a few seconds of Ar<sup>+</sup> irradiation) have the common features as well as the differences in comparison with the results of two hour standard thermal annealing.

### 1. Introduction

Utilization of accelerated ion beams with energies ranging from units to hundreds of keV has given rise to development of a number of innovative technologies currently finding broad application in treatment of structural materials with a view to obtain properties unattainable through application of traditional methods of treatment.

Unfortunately, the projected ranges of the above energy ions in material measure from a few tens to hundreds of nanometers only. The modified zone of such depth is definitely inadequate for the majority of technical applications.

Use of ions with energies of several tens to hundreds of MeV, whilst increasing the ion affected zone to several tens of micrometers [1, 2], however leads to significant complication and rise in the cost of the process. The same relates to attempts to combine different methods of coatings deposition and ion irradiation.

*Use of powerful continuous low-energy [3] and nanosecond pulse power ion beams [4, 5, 6], also high-dose implantation (with generation of dislocations migrating deep into the material) [7, 8], allows the modified zone depth to be extended to several tens of nanometers.*

However, for the purposes of many technical applications, it is more desirable to increase the depth of accelerated ions modifying action on the structure and properties of materials by at least 1-2 orders of magnitude, which would enable modification of *submillimeter* and *millimeter* layers of materials at their surface irradiation.



The record depth of action on materials may be reached at the expense of “*radiation-dynamic effects*” [9, 10]. Nanodomains of dense cascades of atomic displacements present the zones of explosive energy release with formation of thermal spikes. The temperature of such domains, with the time of thermalization of the order of  $10^{-12}$  s, reaches 5000-6000 K and higher [11, 12]. Their fast expansion causes emission of post-cascade shock waves [13-15]. In thermodynamically *stable* media such waves are damped out quickly.

Phenomenological and hydrodynamic description of such processes in *metastable* media with high stored energy [9] indicates the possibility of the undamped mode of post-cascade waves propagation initiating structural-phase transformations at their front. At that, the registered depth of impact is by far ( $10^3$ - $10^5$  times) [9, 16, 17] greater than the projected ranges of heavy accelerated ions. This enables modification of *submillimeter* and *millimeter* subsurface layers of materials.

In the present work, the existence of a radiation-dynamic component in accelerated ions action on material is taken advantage of in recovery of ductility of sheets (1 mm thick) of cold-worked aluminum alloy 1441 of Al – 2.0 Li – 1.7 Cu – 1.0 Mg – 0.11 Zr system. As a result of the cold plastic deformation up to ~70% the alloy completely loses its plasticity, rendering its further cold rolling impossible without intermediate annealing.

## 2. Experimental

Samples were irradiated in ion implanter ILM-1 outfitted with technological low-pressure hollow-cathode glow discharge *ion source* PULSAR-1M [18] generating round cross section gas-ion beams ( $S \sim 100 \text{ cm}^2$ ,  $E = 10\text{-}50 \text{ keV}$ ,  $j = 50\text{-}500 \text{ }\mu\text{A/cm}^2$ ).

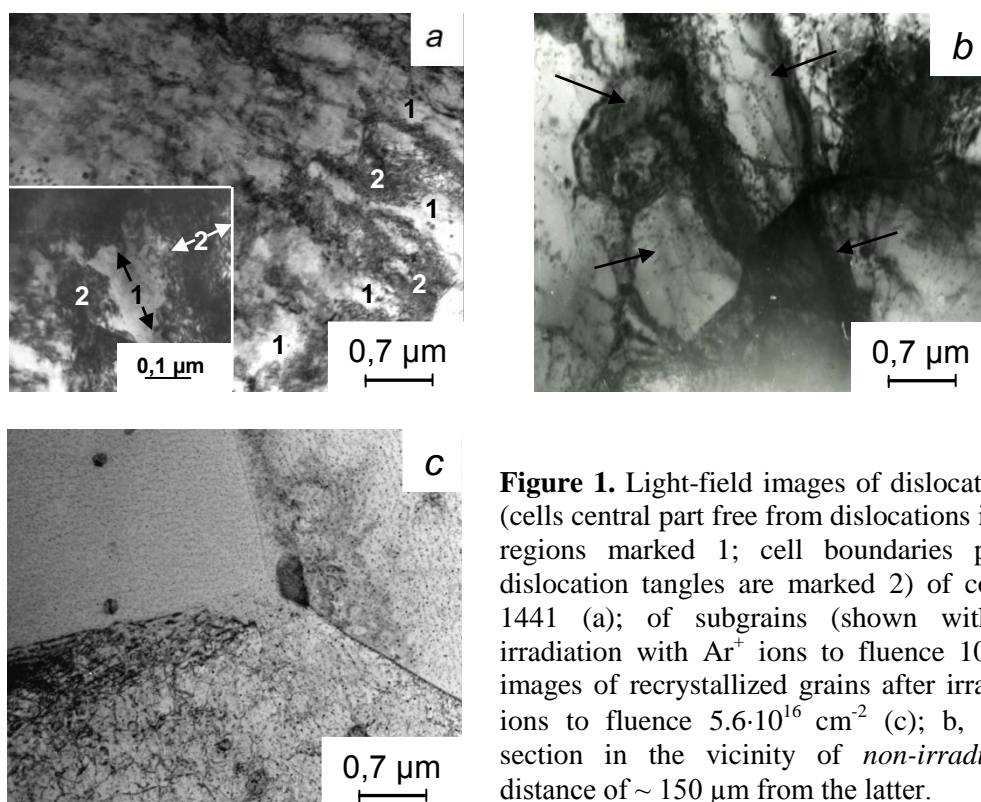
Mechanical tests and structural study were carried out on uniaxial tensile test samples (GOST 1497-84). There were four tensile samples irradiated simultaneously, to one of the samples a thin chromel-alumel thermocouple was welded (with laser welder "Quantum 15") for temperature monitoring. The initial and irradiated samples microstructure was studied under optical microscope Neophot-21 and electronic microscope JEM-200 CX. The irradiated samples structure was studied by these methods across two sections, parallel to the irradiated surface and square to it (cross-section). This allowed us to establish that structural changes and phase transformations initiated by ion irradiation occur throughout the bulk of alloy strips *1 mm thick*: this is more than  $10^4$  times greater than the implanted ions projected ranges which certainly not exceeding  $0.1 \text{ }\mu\text{m}$  [19].

## 3. Details of exposure, measurements and results

*Cold-worked alloy 1441* features higher strength properties and lower ductility:  $\sigma_B = 316 \text{ MPa}$ ,  $\sigma_{0.2} = 296 \text{ MPa}$ ,  $\delta = 3 \text{ \%}$ . In the initial state, this alloy microstructure features a characteristic structure, which points to the presence of elongated  $\sim 2 \text{ }\mu\text{m}$  thin grains. A non-uniform dislocation cell structure is observed in the bulk of the grain (Figure 1 a). There are viewed intermetallics of crystallization origin  $\text{Al}_3\text{Fe}_2\text{Si}$ , on the average, up to  $2 \text{ }\mu\text{m}$  in size, equiaxial-shape  $\beta'$ -phase ( $\text{Al}_3\text{Zr}$ ) particles 40-60 nm in diameter, and fine-disperse  $\text{Al}_3\text{Li}$ -phase particles less than  $1 \text{ nm}$  in diameter precipitated as a result of natural ageing process at room temperature.

*Annealing* ( $T = 370^\circ\text{C}$  during 2 h) leads to an abrupt drop of strength characteristics and increase in ductility of alloy 1441:  $\sigma_B = 245 \text{ MPa}$ ,  $\sigma_{0.2} = 134 \text{ MPa}$ ,  $\delta = 20 \text{ \%}$ , taking place at the expense of formation of recrystallized structure with a grain size of  $3\text{-}8 \text{ }\mu\text{m}$ . Besides, phase  $\text{Al}_3\text{Fe}_2\text{Si}$  and  $\text{Al}_3\text{Zr}$  particles considerably decrease in number with decrease in their size to 50 and 20-30 nm, respectively.

Alloy 1441 *irradiation* to relatively low  $\text{Ar}^+$  ion fluences ( $E = 20\text{-}40 \text{ keV}$ ,  $j = 100\text{-}400 \text{ }\mu\text{A/cm}^2$ ,  $D = 10^{15}\text{-}10^{16} \text{ cm}^{-2}$ , with the respective irradiation times  $\tau = 1\text{-}10 \text{ s}$  and temperature of sample heating with a beam  $T < 40\text{-}130^\circ\text{C}$ ) causes sample bulk structural transformation from the initial cell structure to a subgrain, similar to polygonal, structure (Figure 1 b). This leads to up to 6 % increase in relative elongation at constant strength characteristics.



**Figure 1.** Light-field images of dislocation cell structure (cells central part free from dislocations is viewed as light regions marked 1; cell boundaries presenting dense dislocation tangles are marked 2) of cold-worked alloy 1441 (a); of subgrains (shown with arrows) after irradiation with  $\text{Ar}^+$  ions to fluence  $10^{15} \text{ cm}^{-2}$  (b), and images of recrystallized grains after irradiation with  $\text{Ar}^+$  ions to fluence  $5.6 \cdot 10^{16} \text{ cm}^{-2}$  (c); b, c – longitudinal section in the vicinity of *non-irradiated* surface at distance of  $\sim 150 \mu\text{m}$  from the latter.

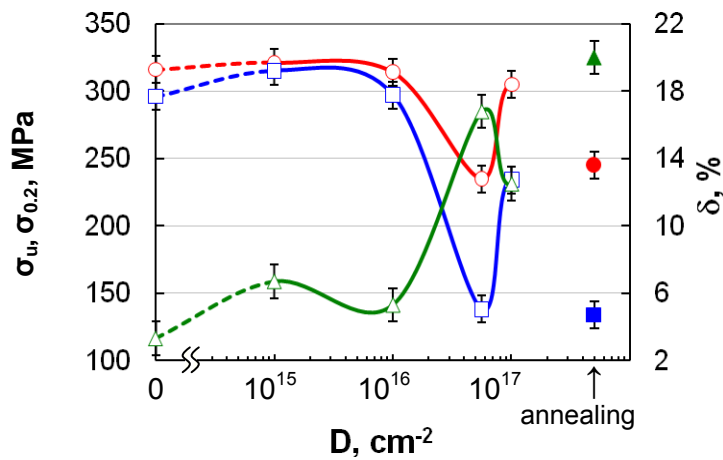
At fluences of  $\sim (2-5) \cdot 10^{16} \text{ cm}^{-2}$  (irradiation time  $\tau \sim 7 \text{ s}$ ,  $T = 330^\circ\text{C}$  at  $E = 40 \text{ keV}$ ,  $j = 400 \mu\text{A/cm}^2$ ,  $D = 1.75 \cdot 10^{16} \text{ cm}^{-2}$  and  $\tau \sim 60 \text{ s}$ ,  $T = 260^\circ\text{C}$  at  $E = 20 \text{ keV}$ ,  $j = 150 \mu\text{A/cm}^2$ ,  $D = 5.6 \cdot 10^{16} \text{ cm}^{-2}$ ), formation of recrystallized grains (Figure 1 c) 12-20  $\mu\text{m}$  in size and fine-disperse equiaxial grains  $\sim 2-5 \mu\text{m}$  in diameter takes place. This leads to significant softening of the alloy and, simultaneously, to increase in its ductility:  $\sigma_b = 218-235 \text{ MPa}$ ,  $\sigma_{0.2} = 130-138 \text{ MPa}$ ,  $\delta = 17-18\%$  (minor differences may occur due to conditions of irradiation). At that, changes in the mechanical properties, also in the dislocation and grain structure, are generally similar to those resulting from alloy 1441 furnace annealing at  $T = 370-400^\circ\text{C}$  during 2-3 h.

With further fluence increase from  $5 \cdot 10^{16} \text{ cm}^{-2}$  to  $\sim 10^{17} \text{ cm}^{-2}$ , *dissolution* of phase  $\beta'$  ( $\text{Al}_3\text{Zr}$ ) as well as *decrease in size* and *partial dissolution* of  $\text{Al}_8\text{Fe}_2\text{Si}$  intermetallides take place throughout the bulk of the alloy. It should be noted that particles of  $\beta'$ -phase ( $\text{Al}_3\text{Zr}$ ) of crystallization origin are preserved even after a two-hour furnace *annealing* at  $T \sim 370^\circ\text{C}$ , while *irradiation* during only units (or several tens) of seconds (in all cases, at  $T < 370^\circ\text{C}$ ) leads to dissolution of the above particles in the bulk of 1 mm thick samples.

Besides, irradiation to fluences of  $\sim 5 \cdot 10^{16} \text{ cm}^{-2}$  and higher leads to formation of disperse particles of a new plate-like phase  $S_1$  ( $\text{Al}_2\text{LiMg}$ ) and particles of phase  $\theta'$  ( $\text{Al}_2\text{Cu}$ ).

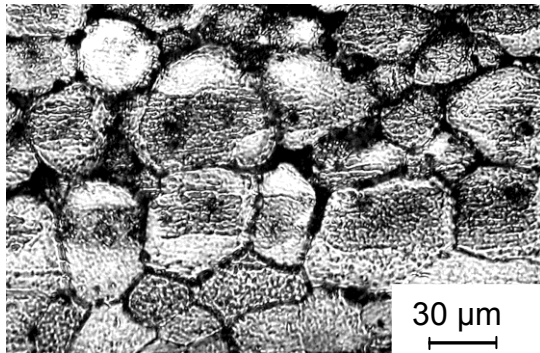
Increase in fluence up to  $10^{17} \text{ cm}^{-2}$  following the mentioned *stage of softening*, again, *leads to increase* in strength characteristics:  $\sigma_b = 315 \text{ MPa}$ ,  $\sigma_{0.2} = 204 \text{ MPa}$  ( $E = 20 \text{ keV}$ ,  $j = 150 \mu\text{A/cm}^2$ ), with some drop of  $\delta$  to  $\sim 13\%$  (Figure 2), in spite of the observed noticeable growth of grain average size up to  $30 \mu\text{m}$  (Figure 3) in the bulk of the irradiated samples. A similar change in properties is also observed at other ion beam parameters:  $E = 40 \text{ keV}$ ,  $j = 200$  and  $400 \mu\text{A/cm}^2$ .

The nonmonotonic nature of mechanical properties change in alloy 1441 under increased fluences (see Figure 2) is explained by simultaneous occurrence under irradiation of *two competing processes*: *recrystallization with formation of new grains*, on the one hand, and *solid solution decomposition* with formation of fine-disperse particles of strengthening phases  $\theta'$  ( $\text{Al}_2\text{Cu}$ ) and  $S_1$  ( $\text{Al}_2\text{LiMg}$ ), on the other.



**Figure 2.** Dependence of alloy 1441 mechanical properties on Ar<sup>+</sup> ions fluence, E = 20 keV, j = 150 μA/cm<sup>2</sup>.

The said processes take place throughout the bulk of 1 mm thick samples (Figure 3) in the course of their short-term heating with an ion beam to temperatures close to the temperatures of common furnace annealing (with no long-term exposure to such temperatures), or at lower temperatures, e.g., not higher than 260°C, in case of irradiation to fluences of  $5.6 \cdot 10^{16} \text{ cm}^{-2}$  at  $j = 150 \text{ μA/cm}^2$ , which is 110°C lower than at intermediate furnace annealing.



**Figure 3.** Microstructure of alloy 1441 irradiated with Ar<sup>+</sup> ions, E = 20 keV, j = 150 μA/cm<sup>2</sup>, D = 10<sup>17</sup> cm<sup>-2</sup>, T < 370°C, in sample cross-section (near the *non-irradiated* side of the sample; the structure of regions in the vicinity of the irradiated side and in the sample central part is similar to that shown).

Short-term (8 s) heating of alloy 1441 samples to 130°C in furnace at a rate similar to that observed under ion irradiation to a fluence of  $10^{16} \text{ cm}^{-2}$  (E = 30 keV, j = 200 μA/cm<sup>2</sup>, τ ~ 8 s, T = 130°C) exerts no influence on the structural state of cold-worked alloys. Samples preserve their cell structure. This positively points to a non-thermal nature of structural transformation in the bulk of alloy 1441 under low fluences ( $10^{15}$ - $10^{16} \text{ cm}^{-2}$ ). It should be noted that at higher fluences (and, accordingly, at a substantial heating of the samples with ion beam exposure), thermally-activated processes get superimposed on the radiation-induced one.

As noted in the Introduction, in *metastable media* with higher stored energy, as is the case with cold-worked alloys, the conditions of undamped propagation of post-cascade waves may be realized, with such waves initiating structural and phase transformations at their front. This relates to the processes of single atoms rearrangement, dislocations unlocking, cooperative atomic restructuring in solids etc. It was shown [9], that practically in all metastable media, even in those with minimum stored energy ΔF of the order of hundredth, or even thousandth, fractions of eV (on the average, per atom of a medium), solitary waves of sufficiently high amplitude ε (ε ≥ Δf; where Δf is of any process energy barrier; normally, Δf < 0.2-0.3 eV) may propagate without damping, causing such media rearrangement.

Energy density in a *self-propagating* (undamped) solitary wave in steady-state conditions is such that the rate of this energy dissipation in a medium is exactly equal to the rate of energy release at the expense of structural rearrangements at its front.

#### 4. Conclusions

Radiation annealing presents a new approach to recovery of ductility of cold-worked alloys. It takes place at lower temperatures and with multiple acceleration of the rate of the process. The energy requirement in treatment is 2-3 times lower than that in furnace annealing [20]. There exist acceptable engineering solutions to the problems related to necessity of implementing the process in vacuum. It was shown [21] that radiation annealing may run in the course of the sheets and sections moving process (relative to the ion source). The difference in structural changes, mostly at final stages of *thermal* and *radiation* annealing, provides additional opportunities for control of structure and properties of the metal.

#### Acknowledgments

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