MODELING OF THE NANOSTRUCTURING MECHANISMS IN SEVERE PLASTIC DEFORMATION OF THE CRYSTALLINE MATERIALS BASED ON THE LIVE CELL MIMICS

Project presentation

Recent developments of the severe plastic deformation processes that allow to obtain parts or workmaterials with improved mechanical properties such wear resistance, magnetism, ductility, thye present poject proposes to verify one hypothesis elaborated by the research team. This is as following "the deformation mechanism that determine nanostructuring of the coarse grain until is reaching nanostructuring grains (lower then 30 nm) could be explained by the analogy with the geometrical evolution of the proteins chains inside the cell". Mathematical model of the protein movement inside the cell, particulary actin filaments will be used in the dynamic molecular simulation of the dislocation trails inside the nanocrystaline materials undergo to severe plastic deformation conditions.

There are presented as follows:

i) **<u>State-of-the-art of the researches results in the field of the project</u> presented in the high level journals as:** in [*Science*, impact factor/2006 (FI) - 30,028; *Nature Materials*, FI - 19,194)] Mat. Sci. Eng. R-Rep (FI 10.52), Advanced Engineering Materials (FI 1,402), Physical Review Letters (FI 6.297), Journal of Computational and theoretical Nanoscience (FI 2.90), Journal of Nanoparticle Research (FI 2.156), as well as presented within the reports of some important research laboratories in the field: Laboratoire de metallurgie mecanique – Ecole Politechnique Federale de Lausanne, Elvetia, Department of Civil and Environmental Engineering, Massachusetts Institute of Technology, SUA, Laboratoire de Proprietes Mecanique et Thermodinamique – LPMTM-CNRS, Franta, The Institute of Physical and Chemical Research – RIKEN, Japonia, School of Materials Science and Engineering, Shenyang University of Technology, China;

ii)

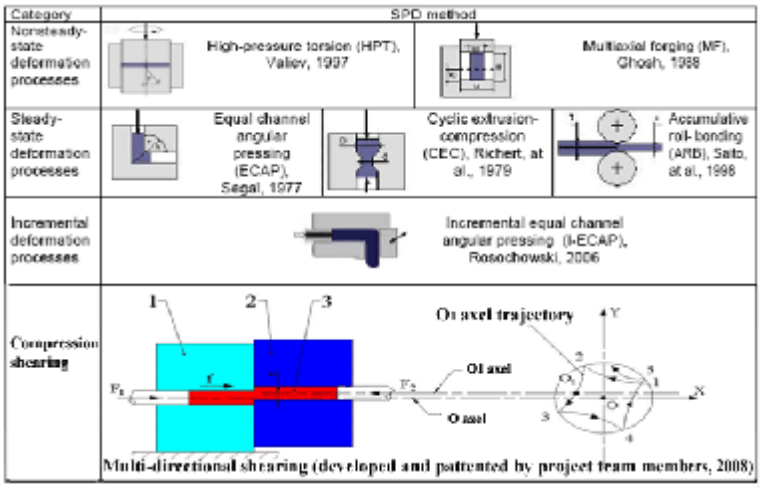
ii) A summary of the preliminary researches carried out by the research team that initiated this research

<u>i) State-of-the-art of the researches results in the field of the project:</u>

By using metal forming, it is possible to fabricate only so-called ultrafine grained metals, not even touching desirable nanometals with nanocrystalline structure. *New findings on the mechanism of plastic deformation* of nanometals could *open the gate for metal forming* in this range. Metal forming processes arrived to the frontier between top-down (materials transformation) and bottom-up (materials synthetization), which means nanotechnology.

1. Severe plastic deformation processes: Severe plastic deformation (SPD) processes (table 1) are the next stept in achieving nanostructuring of any metal that appeares recent as a new task because the size of the grains obtained in the parts - ultrafine grained (UFG) gives the metals unusual combination of mechanical behaviour such high strength and good ductility. Researches was oriented to pass forward to developing new technologies of severe plastic deformation that conduct to nanocrystalline structure (NC) that brings new mechanical properties to the materials such magnetism and will allow to deform pure metals. To control these properties resulted during manufacturing of the metalic materials, a fundamental description of their mechanical behaviour according to the material structure is required. The relation between mechanical behaviour and material structure is the deformation mechanism that involves subdividing the original coarse grains into much smaller domains (subgrains) by various systems of shear bands followed by subgrain rotations [1]. To be effective, this description must be based on a solid understanding of operative deformation mechanisms [7,8].

Table 1 – Some of the severe plastic deformation processes classified on the basis of the speed of the deformation



Remark no. 1 – Severe plastic deformation are manufacturing processes that can lead to ultrafine grain formation and even nanostructures in high pressure condition and large deformation paths. From the existing SPD processes, steady-state ECAP process is candidat for obtaining ***bulk nanostructured*** parts with ***isotropic properties***. Despite the new developed SPD processes, ***ECAP*** is already studied by finite element simulation and experiment and the relation between the influence of the process control parameters (speed, pressure, extrusion angles) on the grain size is already known.

2. The physiscs of the deformation mechanism. Hypothesis. The above-presented SPD processes produce UFG between the regions I-II. As is shown in figure 1, region I correspond to the traditional metal forming processes, where the deformation mechanism is mainly explained by generation of the Frank-Reed dislocations and the relation between mechanical behaviour and grain size – known as constitutive laws is very well fitted by various models and laws (Lemaitre-Chaboche, Swift, Voce, Teodosiu-Hu). Region I is governed by the well-known Hall-Petch effect of grain size (dg), whereby decreased grain size results in increased yield strength. Region II, from 1000 nm to 30 nm, spanning grain sizes from ~1 micron down to ~20 nm, includes phenomena such as localization of shear, decreased strength and increased ductility. Deformation mechanims are caused by disordered grain boundaries that begin to dominate the mechanical behaviour (ultrafine-grained polycrystalline metals). Region III, encompassing grain sizes less than 20 nm, exhibit a reverse Hall-Petch effect, where strength actually increases in proportion to grain size, and deformation is dominated by grain boundary activity. Finally, Region IV includes amorphous metals which exhibit zero strain-hardening and are essentially brittle solids [13]. This region that correspond to nanostructured materials fomed by nanocristals. Nano-crystalline phases have become one focus of attention to scientists because the Hall-Petch relation, when extrapolated to nanometer range grain sizes, predicts extremely hard materials. However, experimental evidence suggests that in the nanometer range the Hall-Petch relation apparently fails to describe the observations; a new regime appears, possibly controlled by grain boundary plasticity, whose quantitative description is still controversial [l-5].

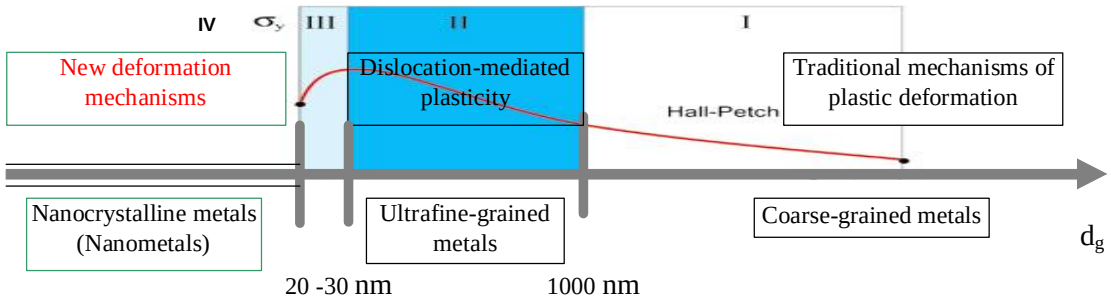


Fig. 1. Classification of polycrystalline metals according to grain size [12]

Remark no. 2 – The ***deformation mechanism*** in Region III and IV are not fully explained nowadys. The recent researches underlined complex phenomena that have a concurrent action and their explanation is possible only using ***stochastic analysis***.

3. Pecularities of the deformation mechanism in the regions III and IV:

Dislocation motion deformation mechanism -In coarse-grained polycrystalline metals, plasticity is carried by dislocations generated by sources within the grains. Such dislocations propagate and interact with preexisting structures and also with each other, and can partly annihilate each other. Based on the concept of dislocation pile-ups at grain boundaries, the Hall-Petch relationship predicts an increase in the yield stress σo (also strength and hardness) with decreasing grain size dg (σi is a friction stress opposing dislocation motion and k is a constant). $\sigma_o=\sigma_i+k / dg$ (1).

This relation loses its validity for grain sizes smaller than 10 – 30 nm, the relationship (1) [6], when the deformation mechanism is based on partial dislocation composed of twinning and grain bounadry sliding. *Twinning* becomes a preferred deformation mode in aluminum with a grain size on the order of 10 nm, which is consistent with the HRTEM observations of deformation twins in the grains with sizes of 10 to 20 nm. The model also provides a physical explanation of the preferential generation of partial dislocations, which results in the formation of stacking faults and deformation twins in nanocrystalline grains as suggested by computer simulations (*6–8*, *12*, *24*). *Grain boundary sliding*, density dislocation and grain boundary difussion become important deformation mechanisms below 5 nm but, at this scale, the precise deformation is still uncertain. At this scale, where in situ TEM analysis or HRTEM are impossible to be done, some authors carried out the stochastic analysis that consist in modeling using dynamic molecular method. Because of the small sizes of grains, dislocations cannot be generated, Frank-Read sources are too large to fit within grain, or because dislocations are energetically very expensive under very small geometrical confinement. This leads to a deformation mechanism based on motion of partial dislocations.

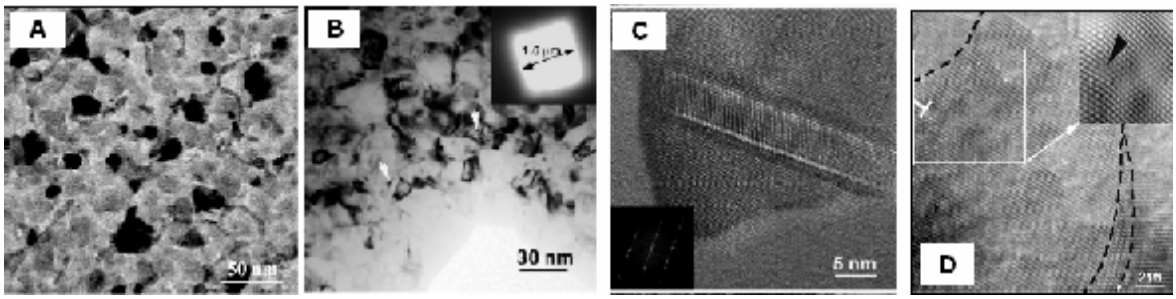


Fig. 2 Deformation mechanisms underlined by the experiment in Regions III-IV

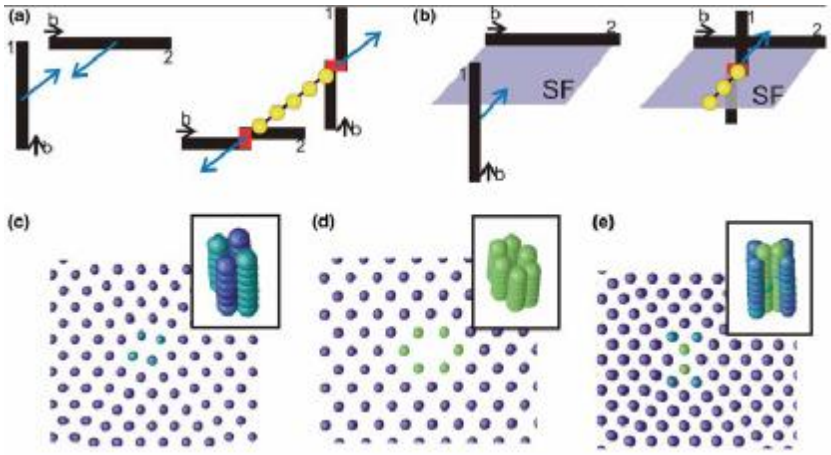
A - Shan[2004] Microstructure of as-prepared nanocrystalline aluminum. (A) Bright-field TEM micrographsh owing nanocrystalline grains with sizes ranging from 10 to 35 nm. No dislocations or deformation twins can be seen.
B – Chen [2003] TEM micrograph of deformation twins around an indent in nanocrystalline aluminum. The inset shows the indent with the fourfold geometry.

C - HEM micrograph showing a deformation twin in (A) with parallel boundaries. This atomic resolution image corresponds
to the [110] direction and illustrates the mirror symmetry between the twin and the matrix. The morphological feature of the grain boundary at the twin band indicates that the twinning results in plastic deformation.

D- Shan[2004] - A typical HRTEM image of a thin area formed by deformation. A dislocation (white T) is trapped inside a grain close to the grain bouandary (GB) (delineated by black dashed line). The inverse Fourier-filtered image (**inset**) from inside the white box shows the dislocation (black arrowhead) with more clarity.

Remark no. 3 – The principal actor of the ***deformation mechanism*** at the nanoscale (below 8-5 nm) is represented by the grains interfaces that are composed by atoms. At this scale, the nanostructure is ***cvasi-amorphous*** that suggests elastic antropy and difussion phenomena. These properties are proper to rubber or ***hyperelastic materials***. The way in which the atoms of each boundary grain communicates with the others is based on the fundamental mechanisms common to all natural materials.

4. Incercari de modelare a mecanismelor de nanostructurare: Molecular dynamics (MD) simulations have been used to study the atomic-scale processes that occur during plastic deformation of polycrystalline aggregates of nanocrystalline grains because at this scale no experimental model is able to catch the real deformation mechanism. The simulations suggest that nanocrystalline metals accommodate externally applied loading, by means of grain boundary sliding and the emission of partial dislocations that run across the grains. The MD simulations also suggest that grain boundary sliding and partial dislocation emission are triggered by atomic shuffing and free volume migration in the grain boundaries. These dynamic atomic-scale processes would be nearly impossible to image with TEM due to the instant time associated with their occurrence. Sharpening of the diffraction peaks, as a result of plastic deformation, is one indication of structural rearrangement in the grain boundaries.
[Buehler] reported that using dynamic molecular of a 500 million-atom simulation of work hardening in a model system for ductile solid, the dislocations exhibit three fundamental mechanisms: (1) dilocation cutting processes, jog formation and generation of trails of point defects (that represents the discovery of a new class of point defects which could play an important role in situation when partial dislocations dominate plasticity), (2) activation of secondary sleep systems by Frank-Read and cross-slip mechanisms, and (3) formation of sessile dislocations such as Lomer –Cottrell locks. Buehler obtained the representation of the dislocation in Cu deformed with 25% equivalent strain (figure 4). Representation of the mentioned mechanisms are presented in figure 3.



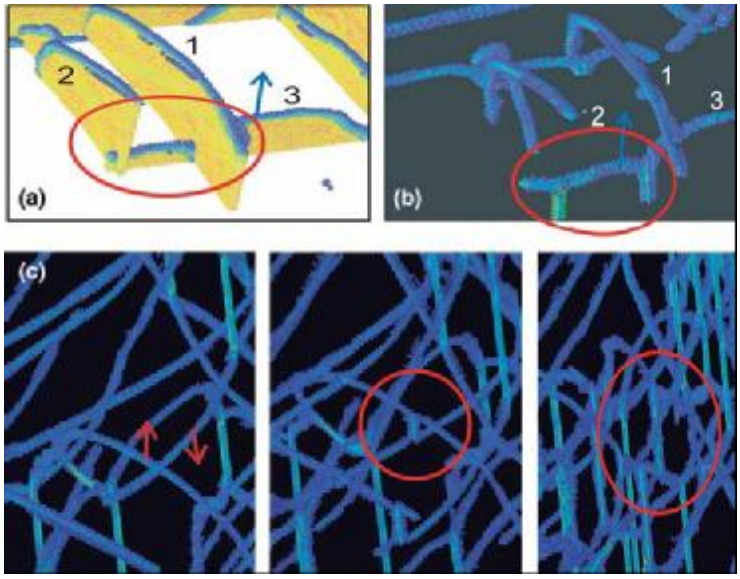


Fig. 4 - Generation of trails of point defects in early stages of the simulation. Dislocation number 1 and number 2 leave a stacking fault plane, which is subsequently cut by dislocation number 3. Therefore, two trails of partial point defects are generated resulting in bowing of dislocation number 3 (see the mechanism shown in Fig. 3(b)). Subplot (**a**) shows a centrosymmetry analysis [25] where the stacking fault planes are drawn yellow; subplot (**b**) shows an energy analysis of the same region where the stacking fault planes are not shown. Panel (**c**) shows the reaction of the two dislocation clouds originating from opposing crack tip causing generation of numerous point defects

Due to the Burgers vectors and the dislocation line orientation, when the dislocation clouds meet straight ahead of the crack, the reactions are very similar to those observed in the previous stages when dislocations of the same clouds cut each other's stacking fault and thus, trails of partial point defects are generated. In Fig. 4(c), the significant effect of jog dragging on the motion of the dislocations is clearly observed. The elastic interaction of dislocations repelling each other causes a decrease in the dislocation velocity. Due to the crystal orientation and the Burgers vector of the dislocations, only trails of partial point defects as well as interstitials can be generated from the cutting processes on the primary glide systems. Complete vacancy tubes [13,14,26] are not generated until later stages of the simulation when the dislocation density becomes very large and secondary slip systems are activated (see discussion in the next section). A large number of such defects are created and appear as straight, thick lines in the plot of the potential energy of atoms.
In the simulation, we find that dislocations on secondary slip systems are generated by cross slip and Frank-Read mechanisms. This is an unexpected observation because cross slip is only possible, according to the classical dislocation mechanics, along at least locally constricted screw segments (Friedel-Escaig's mechanism) [1]. Due to the low stacking fault energy of the model material used here, only partial dislocations exist in our simulation. Each occurring cross-slip event leaves behind a straight, sessile stair-rod dislocation to conserve the Burgers vector. These sessile segments can clearly be observed in Figs. 4(a)–4(d). The main result is that by this novel mechanism it is possible to observe cross-slip in a situation when only partial dislocations, together with very high stresses are present. Even in systems with only partial dislocations, nature finds a way to relieve elastic energy into secondary slip systems!

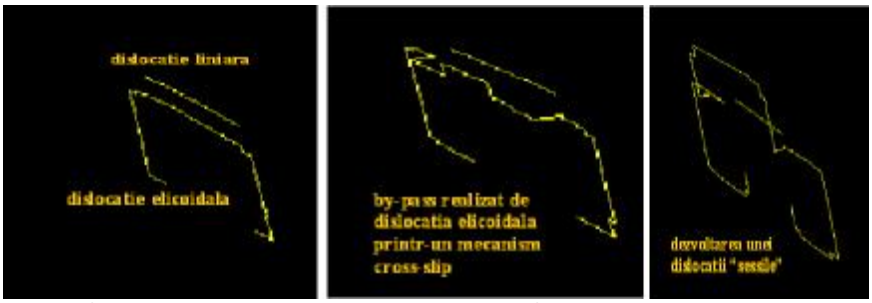
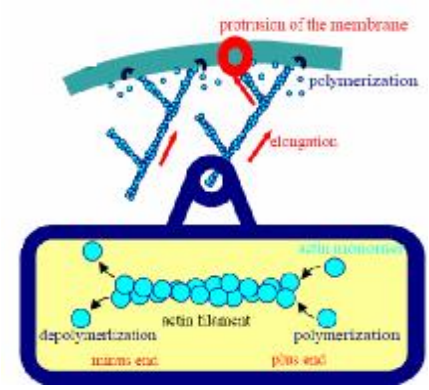
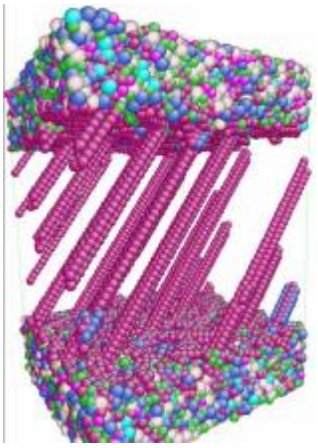


Fig. 5 – Formation of the sessile dislocation by making a by-pass of one dislocation at the intersection of the other dislocation – activation of the secondary slip system

Remark no. 4 - Discovering of the new deformation mechanims that act only below 8 nm that apprear to have a self-organized structure (property that is characterized to the nanometals). The shape of the tube vacancies of the dislocations looks like molecule chains and that are connected by small distances such bridges. The lines are straight until the concentration of the vacancies increases at the very high deformation pressure. At this time, the tube vacancies of the defects become wiggly bluish.
Remark no. 5 - A new kind of crystal defect as trail of point with a dipole structure that generate a dragging force and make possible the movement of the vacancy tube. It is a obvious similarity in vacancy tube movement and the movement of the protein chains in the cell. The protein, particulary actine chain polymerise by the forces between the two dipoles of the actine segment of the protein chain.





a)
Movement of the cell by formation of the citoskeletonului [E.Yamaoka,T,Adachi, RIKEN Symposium 2007]

b)
Sessile dislocation representation in the nanocrystal [Y.Wang, J. Li, A. V. Hamza, T. W. Barbee, Apl. Ph. Sci.2007]

Fig. 6 – Representation of the results of the molecular dynamics simulation of a) the actin filaments inside a cell and of b) sessile dislocations inside the nanocrystal at the interface between two grains

5. The hypothesis proposed by the research team to explain the deformation mecanism at nano scale

From the critical and in the same time constructive analysis of the state-of-the-art briefly described within point 1, 2,3,4 and 5, and based on the remarks no. 1, 2, 3,4 and 5 and reinforced by the conclusion of the recently published report by Dr. Buehler "Large-scale hierarchical molecular modeling of nanostructured biological materials" that presents the absolutely newest results concerning the comparison of the deformation mechanisms of biological materials with crystalline materials such as metals or ceramics, with emphasize on the importance of entropic contributions to plasticity, and the interplay of chemical bonding of different strengths, at different length- and time scales, the team of the present proposal elaborate the following original hypothesis:
The deformation mechanisms that govern nanostructuring of the crystal materials in Region IV (under 20 nm grain size) could be modeled using the continuum mechanism modelling of the protein chains evolution from the live cell. Tube vacancies shape that are the trails of point defects in the nanocrystal evolve by mimics like the protein folding and unfolding generating the increased capacity of plastic deformation of the material at this scale. This is superplasticity that was noticed in the severe plastic nanostructured materials and presently not explained. This property is accompanied by the magnetism phenomena probably caused by the perturbation of the elastic field arround the vacancy tube (like the magnetic field generated by the electricity in conductors).

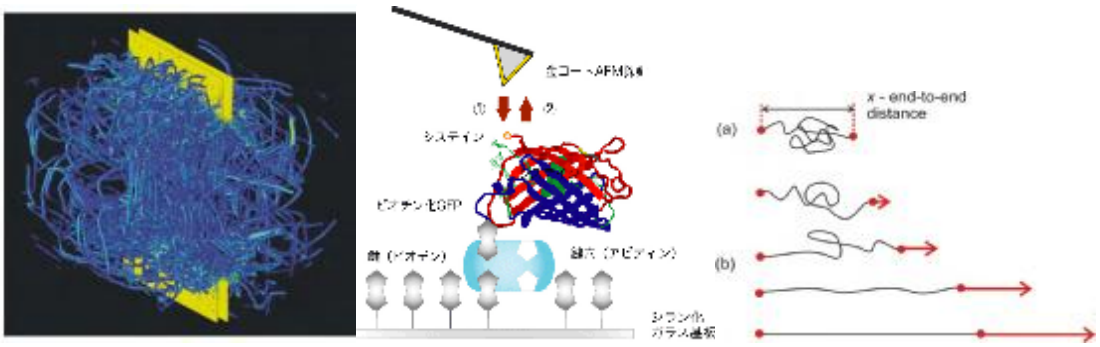


Fig. 6 Comparison of optical representation of (a) dislocations in a ductile copper [Buehler 4,5], (b) protein molecule representation [RIKEN,14] and (c) unfolding path of a protein molecule [Buehler, 4,5,11] that lead to the elaboration of the hypothesis.

Importance of the thematic proposed:

As it results from the literature review, a model form the deformation mechanism at the nanocrystalline level (under 10 nm) is not fully determined. Recent researches noticed some components of the deformation mechanisms that concure in deforming at this scale, but (1) there are not complete descriptions, (2) there are not mathematical modeled in order to facilitate the prediction of the mechanical behaviour at the nanoscale that could be bridged to multiscale analysis either by [Teodosiu, 2007]:

i) information-passing approach – the information gained at a lower scale is used to reduce the number of error at a larger scale. Typically applied to define internal state variables and their evolution equations (ex. work-hardening laws for dislocation-dislocation interactions and formation and effect of dislocation structure);

ii) concurrent approach – the analysis is conducted simultaneously at a smaller and a larger scale, providing each other boaundary conditions and homogenized properties (ex. analysis of the stress concentrations around dislocations and other defects, the self-consistent method and the numerical mesoscope method.

Importance of the project is obvious in the above-mentioned context where is a lack of explanation concerning the deformation mechanism that represent the major objective of the project. Finding the model of the deformation mechanism by similarity with the protein chains evolution in the live cell will open the new gates:

- to develop new dislocation based hardening laws for crystalline structures underlined to severe plastic deformation until nanostructuring that will give a better identification of the internal state of the material;
- to control and predict the mechanical properties of the nanocrystalline materials at the macroscale by implementation in the existing constituive laws of supplementary intrenal variables that characterize the material at nanoscale;
- to design new top-down* metal forming technologies that are compatible with the bottom-up** nanotechnologies. The nanotechnologies are expensive and presently there are not produced the bulk parts or workmaterials with nanostructure isotropic properties;
-to transform ordinary materials by metal forming materials into high quality materials with improved properties such superplasticity, magnetism and resistance to wear that could open new applications of the bulk formed parts.

*top-down (realizarea pieselor prin prelucrarea materialului si obtinerea de proprietati si structuri dorite);
**bottom-up (construirea materialului atom cu atom)

**Scientific impacts:* The control of the deformation mechanisms in nanometals will open the gate to design new metal forming processes by settings the process and conditions of the process that pass beyond the ultrafine-grained materials to nanocrystalline materials and to apply the principles of self-organization in device manufacturing that will provide opportunities to go far beyond currently existing technologies.

**Technological impacts:* Identification of atomic behaviour of the nanocrystalline materials will able to create materials models necessary to an accurate modeling of the molecular dynamics and to predict the materials behaviour by similarities with the protein behaviour from proteins data bank.

Relevance of the thematic proposed:

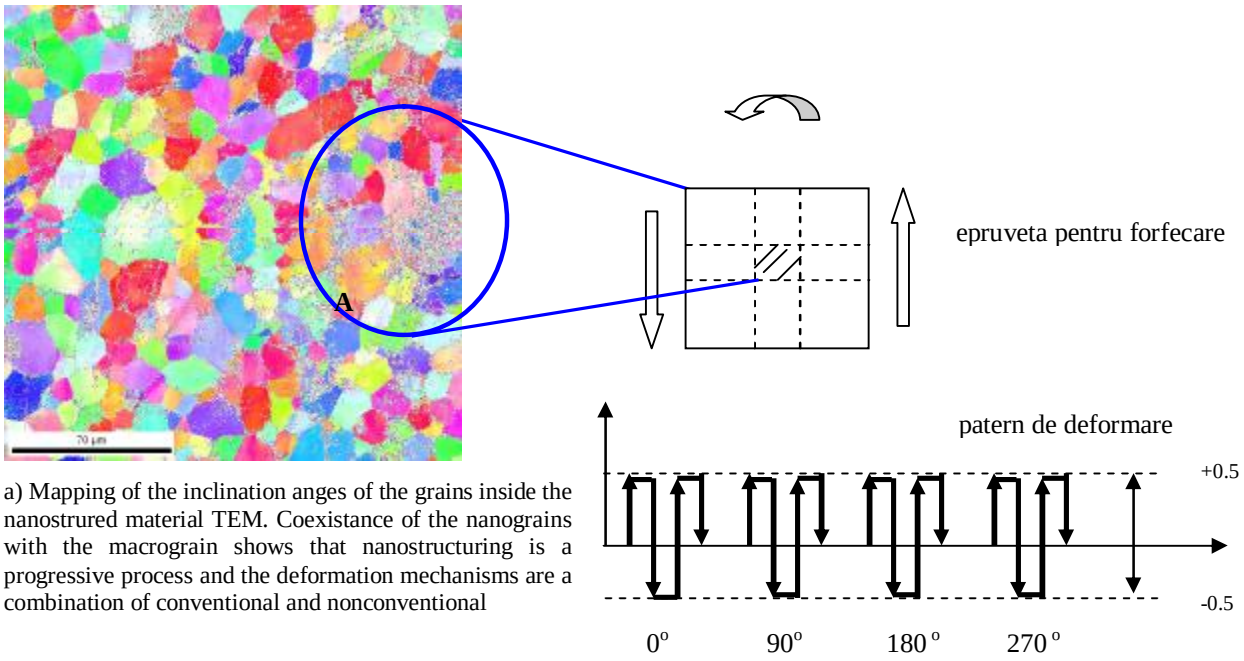
The project refferes to the scientific essence of developing a new way of modeling nanostructuring deformation mechanisms in the bulk cristalline materials. The research activities proposed within the project are placed inside the problem that represents the fundamental description of the atomic scale arrangement of the nanostructured materials.

The thematic of the project is relevant because it proposes: 1) to solve one technological complex problem by using knowledge from live cell modelling that stay to the basis of creation, 2) to bring into attention the metal forming processes in the same place as nanotechnologies are in this century; 3) to create the bridge for the next generation of the products with high level of mechanical properties; 4) to develop a new field – nanometal forming - that presently is not included in the categories of the nanotechnologies (chemical vapor deposition, powder sinterising, lithography) because of its limitation of transforming the materials in nanostructured materials with a self-organized structure and 5) to contribute to the next generation researchers formation by dealing with top-level subjects of the research and creation of material resources according to the research excellence standards.

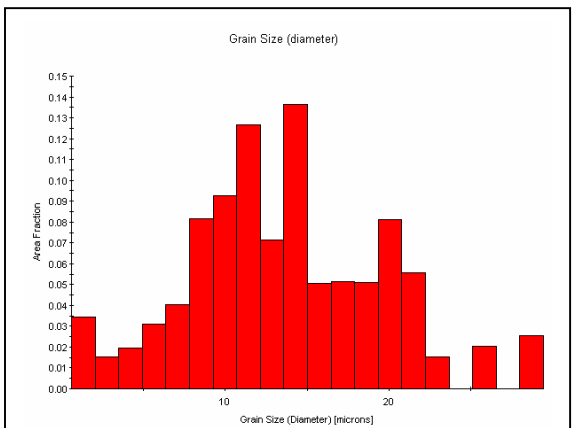
Results obtained by the team project preliminary to this proposal

¨Description of the deformation mechanism for severe plastic deformation of the aluminium alloy by multidirectional controlled shearing process

In october 2007, at LPMTM France, some mechaniscal tests in order to study the effect of nanostructuring on the aluminium alloy microstructure were carried out. Some Bauschinger sheares until 20% strain were done by progressive rotation of the specimen. The specimen was a aluminium sheet of 1 mm thickness with the dimensions indicated in figure 7b. The path of the deformation is presented in figure 7c, and the obtained SEM/TEM image of the nanostructured probe is presented in figure 7a..



a) Mapping of the inclination anges of the grains inside the nanostrured material TEM. Coexistance of the nanograins with the macrograin shows that nanostructuring is a progressive process and the deformation mechanisms are a combination of conventional and nonconventional



b) Shape of the specimen 15 mm x 15 mm x 1. mm The central zone is cumulating the higher degree of deformation..

c) Hystogram of the grain size distribution inside the microstructure. The smallest grain size is 2-3 100 micrometers and the coarse grain size was 100 micrometers.

Fig. 4 – MEB (Microscope Electronic a Baleiaj) analysis of the nanostructured aluminium alloy

The researches underlined that the severe plastic deformation determine the evolution of dislocations to an equilibrium state, passing through the states where the orientation of the grains is totally different and their size is in a very large range. Changing of the path of the deformation determine that finally the microstructure to be diseoved and nanostructured material to be achieved.

The international image of the project team researches in connected domains with the proposed theme:

1. Collaboration with Laboratoire des proprietes Mecanique et Thermodinamique – CNRS, Universite Paris 13, France – european laboratory specialized in analysis of the deforming mechanisms and advanced microscopy. TCM Department has a research bilateral agreement which makes reference to the utilization of the common research platform which consist in SEM microscope – Scanning Electrone Microscope, TEM microscope – Transmission Electronic Microscope, HRTEM microscope – High Resolution Electronic Microscop. The team members has common performances in the domain of the constitutive laws identifying for advanced materials as steel with high limit of elasticity, Al-Mg alloys, TRIP steel, dual-phase [H. Haddadi, S. Bouvier, M. Banu, C. Maier, C. Teodosiu, Int. J. of Plast., 2006, 21 citations in ISI journals; M. Banu, S. Bouvier, C. Maier, C. Teodosiu, Digital Die Design 3DS Report, 2001, 8 citations in ISI journals].

2. Collaboration with V-CAD Integrated Research project, Materials Fabrication Laboratory, The Institute of Physical and Chemical Research – RIKEN Japan – the biggest research institute in Japan. The goal of this project is to connect the numerical modelling methods of the intelligent manufacturing processes with the numerical modelling of the living cell evolution. The scop of this interdisciplinary project is to approach the numerical modelling as an artificial manufacturing instrument and microstructural analisys of some products from mechanical engineering (jet moulding, forging, deep drawing), and also as an investigations instrument of phenomena which are the base of human cell evolution (the modelling of proteins chain, the modelling of living tissues behaviour under mechanical actions). In this VCAD Programme, the finit element analisys of threedimensional internal structure of a crystaline material is coupled with the modelling with molecular dinamics of interactions between mechanical and biochemical factors of proteins chains evolution. The mathematical models of this interactions is based on the mechanics of continuous medium, geometrie modelling and statistics.
The project director is associated researcher of VCAD Project (Volume CAD) and has common performances in the numerical modelling domain of the deformation processes, the anticipation of material properties using multiscale biomechanical analisys. [M. Banu, T. Hama, C. Teodosiu, A. Makinouchi, Int. J. of Mat. Proc. Tech., 2006] – 5 citations in ISI journals, [M. Takamura, M. Banu, C. Teodosiu, ESAFORM 2006], [J.L. Alves, M. Banu, O. Naidim, E. Naghi, C. Teodosiu, Int. J. of. Mat. Proc. Tech., on publication].

3. Collaboration with Centre de Mise an Forme des Materiaux, CEMEF, Sophia Antipolis, France – CNRS Excellency Research Center, specialized in the numerical modelling of advanced materials, microforming and nanomaterials. The project team collaborates with CEMEF since 1993 by appointing some research stages in the numerical modelling domain with finit element. CEMEF is the coordinatorof FP6 Project – Virtual Intelligent Manufacturing VIF_CA 507330, in the Nanomaterials field, nanotechnology and new treating processes (2004-2008). The goal of this project is to create an excellency network between 49 european partners (research centers, research laboratory, universities, companies) in the domain of coldflow and experimental and numerical modelling study of the relation between deformed materials properties with high deformation degree and microstructure. TCM Department is a member of this project and of this consortium by being in European Research Excellence Network, enjoying the consultation and the material base of every laboratory from this network, and offering also for the other members of the network its own material base.

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