BALL MILLING AMORPHIZATION IN A VIBRATING FRAME GRINDER

Y. Chen, R. Le Hazif, G. Martin

COLLOQUE DE PHYSIQUE
Colloque C4, supplément au n°14, Tome 51, 15 juillet 1990

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Y. CHEN, R. LE HAZIF and G. MARTIN

Centre d'Etudes Nucléaires de Saclay, CEREM-DTM-SRMP, F-91191 Gif sur Yvette Cedex, France

Résumé : On propose un modèle simple du transfert d'énergie par chocs au matériau en cours de broyage dans un dispositif à plateau vibrant : l'énergie par choc varie comme la masse de la bille et le carré de l'amplitude et de la fréquence de vibration du plateau. Le tout est réduit d'un facteur d'efficacité du transfert qui dépend, entre autres, de la rigidité relative de la bille par rapport au matériau traité. La fréquence des collisions entre la bille et le matériau varie comme l'amplitude et la fréquence de vibration du plateau et comme l'inverse de la gravité terrestre. Pour tester l'influence de ces paramètres sur la stabilisation par broyage de phases métastables, un dispositif original a été construit, qui permet entre autres, de procèder à des broyages prolongés sous bon vide. Les résultats préliminaires obtenus sur $Ni_{10}Zr_7$ montrent l'importance du facteur d'efficacité pour atteindre une amorphisation complète du matériau. Sous bon vide, il est possible d'obtenir une feuille continue de matériau amorphe.

Abstract : We propose a simple model of energy transfer in the course of ball milling in a vibrating frame device : the energy transferred per impact goes as the mass of the ball and the square of the amplitude and frequency of vibration of the frame. A reduction efficiency factor must be introduced, which depends on the ball rigidity with respect to that of the material being processed. The frequency of impact between the ball and the material goes as the amplitude and frequency of vibration of the frame and as the inverse of the terrestrial gravitation constant. For assessing the role of the above parameters in stabilizing metastable phases by ball milling, a new device has been built which allows to keep good vacuum during long time milling treatments. Preliminary results on $Ni_{10}Zr_7$ point to the crucial role of the efficiency factor in reaching full amorphization. Under good vacuum, continuous foils of amorphous material can be obtained.

1 - INTRODUCTION

As discussed in a companion paper /1/ , the formation of a non equilibrium phase under ball milling can be viewed as an example of a dynamical phase transition in a forced system. The stability field of such phases must be looked for in the appropriate control parameters space. We suggested that besides the temperature and alloy composition, such parameters as the energy per impact and the impact frequency should play a crucial role in stabilizing non equilibrium phases. The latter two quantities are easy to adjust in a vibrating frame device. The purpose of this note is to give a very simple mechanistic model of milling in such a device, and to present preliminary results obtained on a modified mill we developed at Saclay.

2 - MECHANISTIC MODEL OF MILLING WITH A VIBRATING FRAME

For the sake of simplicity, we consider a vibrating frame on which a vial is attached. The vial contains powder and a single ball of mass m (specific mass ρ) and radius R. The frame vibrates sinusoïdally with a pulsation ω and amplitude A ; g is the terrestrial acceleration constant. We assume, for the time being, that every time the ball hits the powder, the kinetic energy of the ball with respect to the frame is fully absorbed by the powder : under such circumstances, the movement of the ball is periodic in time.

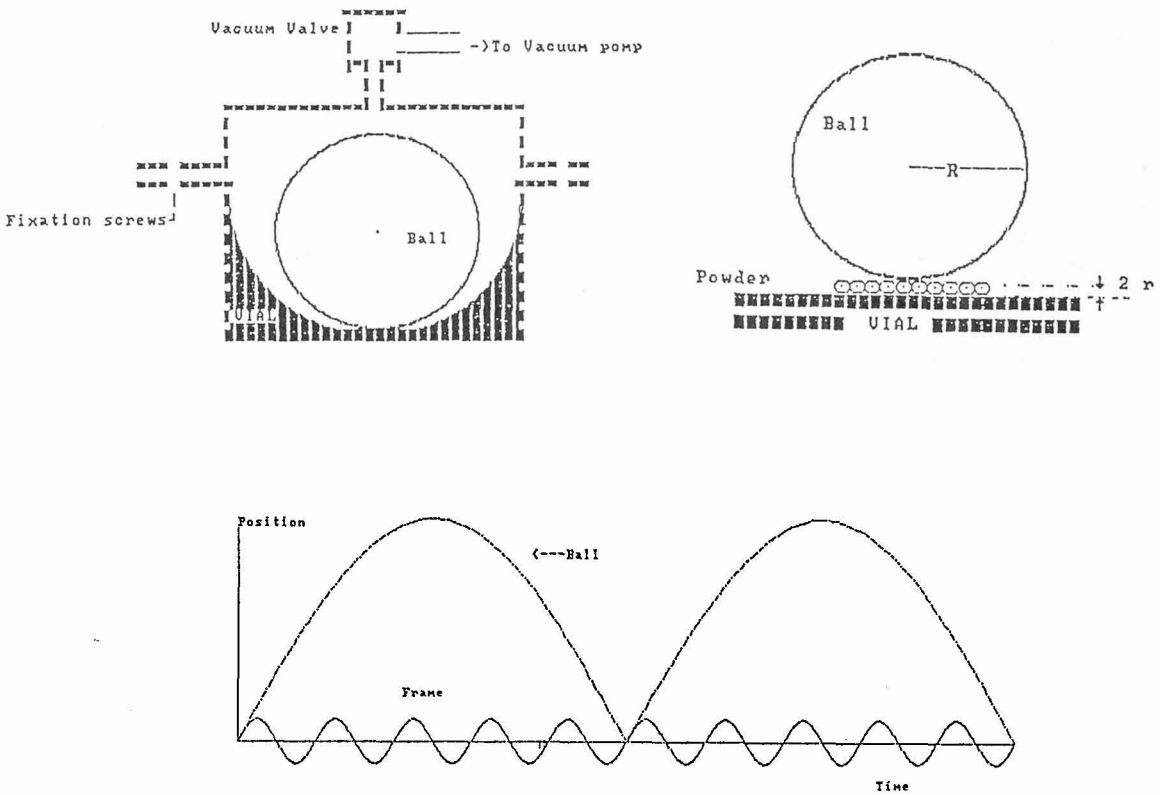

Fig. 1 : Schematical representation of the ball in the vial, the model for the impact between the ball and the powder, and positions vs time of the ball and the frame.

It is easily found that:

- the velocity of the ball, as it leaves the frame, is :

$$v_{bi} = \omega A \sqrt{1-g^2/A^2\omega^4} \approx \omega A \qquad (1)$$

- the latter fixes the height h reached by the ball in the laboratory ($h = v_{bi}2 / 2g$), the time τ needed for the ball to fall back on the frame :

$$\tau \approx 2 v_{bi}/g \qquad (2)$$

- the average kinetic energy of the ball relative to the frame at the collision is (assuming a random dephasing of the ball movement with respect to that of the frame) :

$$\langle E \rangle \approx 1/2 \; m \; (\langle v_b 2 \rangle + \langle v_f 2 \rangle) \qquad (3a)$$

where v_f stands for the velocity of the frame, and <> holds for the average over long times. Simple algebra yields:

$$\langle E \rangle \approx 3/4 \; m \; A^2 \omega^2 \qquad (3b)$$

for the energy per impact ;

- Eqs. 2 and 3 yield the mechanical power transfered to the material undergoing the collision:

$$P \approx 3/8 \; m \; A \omega \; g \qquad (4)$$

- the amout of material being strained at a collision is :

$$\upsilon \approx 4 \pi \; R \; r^{\,2} \qquad (5)$$

where r is the radius of the grains of powder (see fig. 1);

- as a result, the specific power injected into the material is :

$$p \approx 1/16 \rho \; (R^2/r^2) \; g \; A \omega \alpha \qquad (6)$$

The parameter α is a reduction factor, which accounts for two effects :

- the amount of material, υ, (eq. 5)which is strained at a collision, is only a fraction of the total amount of material being processed; assuming perfect mixing of the powder between two impacts results in the following expression :

$$p \approx 1/5 \; (m/m') \; \rho' g \; A \; \omega \alpha' \qquad (7)$$

with m' and ρ' for the mass and density of the powder in the jarr;

- at the collision, the impact energy is not totally transferred to the powder; part of it is absorbed by plastic and elastic deformation of the ball; the fraction of the impact energy transferred to the powder is α', a number which is material dependent.

Typical values are as follows:

A = 1mm, ω = 314 rad/sec., m = 1 Kg, R = 2.5 cm, r = 1 μm : p ≈ some eV/ atom/ second times α. The latter reduction factor is unknown. Most of the above power is dissipated into heat, a small fraction of it (some %)will result in point-defects or antisite defects creation /2/.

Such figures are typical of power injections into the lattice during high energy high intensity irradiations with charged particles (e.g. in a high voltage electron microscope) /3/. The latters are known to induce phase

changes, such as precipitate formation or dissolution, order-disorder transitions, amorphization or crystallization.

For assessing whether the type of phase to be stabilized by ball milling can be rationnalized in terms of injected power and of impact frequency, we equiped a vibrating frame and started studying the conditions for ball milling amorphization of $Ni_{10}Zr_7$, much in the same way as in ref. /1/; comparing the milling conditions for amorphization under both types of milling (planetary and vibrating frame) will help choosing the proper control parameters of the dynamical state of the alloy under milling. We now give preliminary results.

3 FIRST RESULTS

3a- Experimental procedure:

For the sake of comparison with the results obtained in ref.(1), we started with a $Ni_{10}Zr_7$ compound produced by levitation melting in the laboratory, starting from 99.9 % purity material. Typically, pieces of ingot weighting approximately 5 grams were introduced into a modified vial of a "Pulverisette 0" vibrating frame manufactured by FRITSCH.

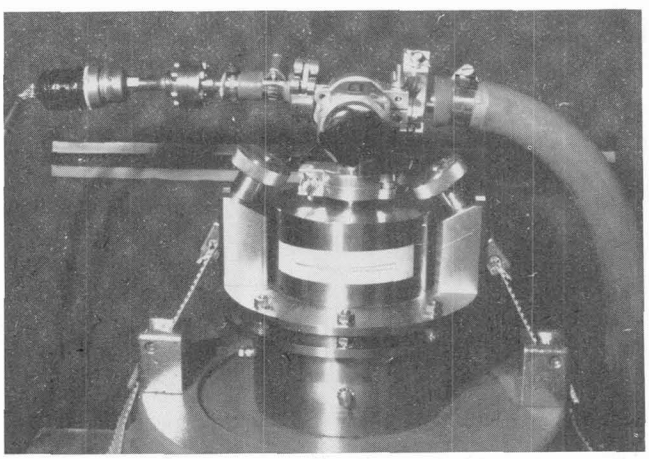

Fig. 2 : Modified Vial attached to the frame.

For grinding *in vacuo* or under a controlled atmosphere, the vial was modified as can be seen on figs.1,2. Static vacuum better than some 10^{-2} Torrs could be kept for at least 130 hours milling time. The vials and balls are either made of tungsten carbide (WC) or of quenched steel (QS). Typical weight of the ball is 1 Kg, which corresponds to a ball diameter of approximately 50 mm and 63 mm for WC and QS respectively.

The vibration amplitude (2A in eqs. 3-7) ranged from .5 to 2.5 mm, most experiments being done with 1 mm. The vibration frequency was 50 Hz.

Milled material was analysed by X ray diffraction and, in some cases, observed by electron microprobe, and scanning or transmission electron microscopy.

3b- Experimental results

The results obtained depend both on the type of atmosphere and on the nature of the ball (WC or QS).

At early times, the following is observed, irrespective of the procedure : after a few minutes, the ingot is broken into coarse pieces ≈ 0.1mm in size, and then into a finer powder (10 to 50 µm in diameter after about 5 hours). The evolution at later stages depends in a crucial way on the nature of the ball used and of the atmosphere.

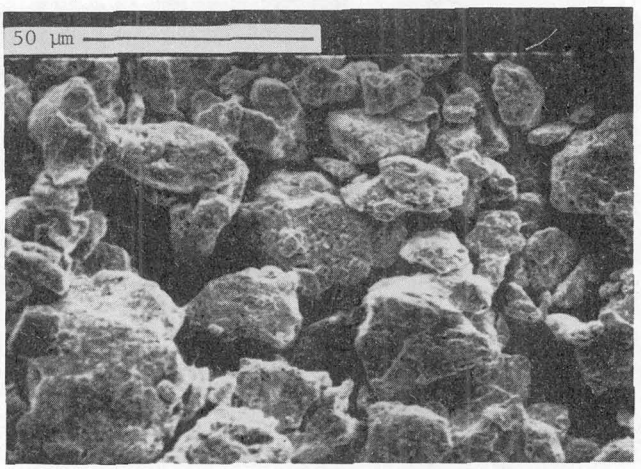

Fig. 3 : Amorphous powder after 30 hours milling with WC ball (residual O_2 atmosphere).

After about 30 hours milling, a fully (respectively partial) amorphous state is observed when milling proceeds with the WC (resp. QS) ball. If milling is performed in the presence of some Oxygen partial pressure, the material is obtained in the form of a powder, with a grain size of about 20 µm (fig. 3); such morphologies have often been described in the literature. On the contrary, if milling is performed in vacuo , the metallic powder sticks to the vial and gets compacted in the form of a continuous meniscus which covers the bottom of the vial (fig. 4). In this case, only the thinner periphery of the foil is fully amorphous. The center part, 2 mm thick is amorphous in the upper part only. As usual, some tungsten carbide contamination is observed when using the WC ball.

At still later stages (>100 hours), the evolution of the compound depends dramatically on the atmosphere. Under some oxygen partial pressure, the alloys decomposes into three distinct phases : Zirconia, pure Nickel and

an unidentified intermetallic compound. That the oxidation of amorphous ZrX alloys (X= Cr, Cu) leads to the rejection of X is a well established fact /4/. More unusual is the observation that, *in vacuo* , the structure obtained at earlier stages remains stable : the metallic foil still covers the bottom of the vial. Whether or not the thickness of the amorphized layer increases with milling time is not yet clear.

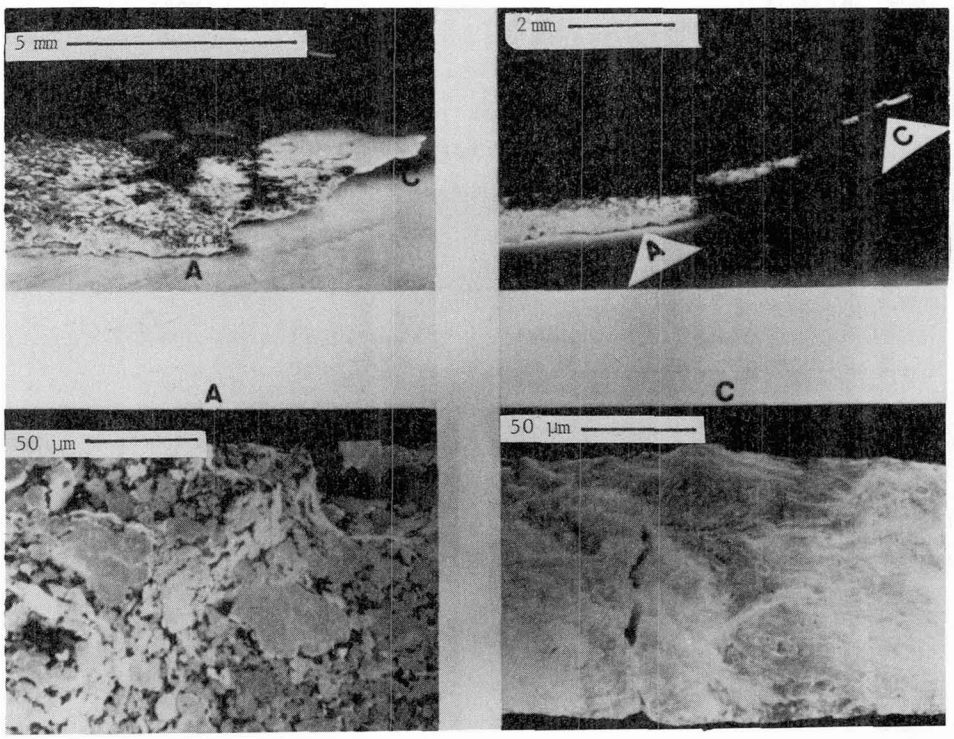

Fig. 4 : Meniscus obtained after 130 hours milling with WC ball under good static vacuum.
C) amorphized zone
A) mixture of amorphized and crystallised zones

4 - DISCUSSION AND CONCLUSION

We have proposed a very simple analysis of energy transfers occurring in ball milling on a vibrating frame, as well as preliminary results on solid state amorphization of $Ni_{10}Zr_7$ in a modified device allowing for good vacuum to be maintained in the course of milling.

The mechanistic analysis shows that the energy transferred at each impact, by the ball to the material being processed, scales with $mA^2\omega^2\alpha$, and that the impact frequency goes as $\omega A/g$.

Although very preliminary, the present results point to the strong influence of the efficiency factor α. A QS ball is softer than a WC one, by about a factor of 4 : full amorphization is more difficult with the latter ball. Wether this decrease of efficiency can be compensated using a higher vibration amplitude and ball mass as predicted by eq.7 remains to be assessed.

Also, the good vacuum we could maintain in the present device, allows for the direct obtention of metallic amorphous foils in the vial.

Acknowledgements : The assistance of Drs. Boulanger, Maurice and Sapin in performing the TEM, SEM and X ray diffraction analysis, is gratefully acknowledged, as well as useful discussions with Pr. Bakker and Dr. Gaffet.

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