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Impact Frequency and Energy Transfer in Milling Processes: An Experimental Approach

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Abstract. An experimental procedure is outlined for the in situ evaluation of the number of collisions per second and the averaged shock power in the course of ball milling and mechanical alloying processes. A piezoelectric device was used to measure the impact frequency while a calorimetric method was concurrently employed to determine the hit energy dissipated as heat. Tests were performed with a commercially available shaker-mill on nickel powders. Because of the progressive change in the elasticity of the hits, impact frequency decreases by increasing the amount of powder loaded in the vial; a limit value is approached which correlates closely with the inherent period of the vial. The energy flux was found to be constant for largely different charges of the processed powder. Values are reported for the hit frequency, the transferred energy per hit and the relative velocity of the ball at the impact time.

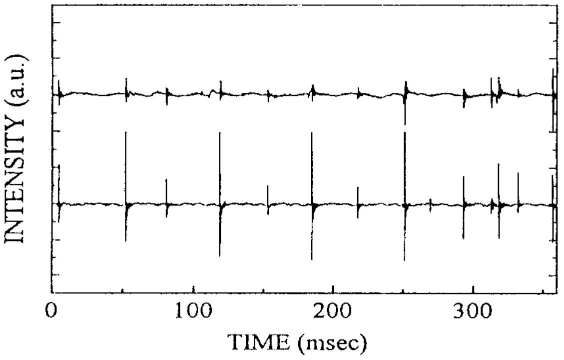
Introduction

Solid state transformations induced by intensive mechanical deformations have received a considerable scientific attention in view of their importance for technological applications. Mechanical Alloying and Milling (MA/MM) have been utilized to produce equilibrium compounds and a variety of metastable systems as amorphous alloys, nanostructured phases and nanocomposite particles, confirming the great potential and flexibility of these nonequilibrium processing methods [1-2]. Despite this growing interest, fundamental questions concerning mechanically sustained processes remain largely unanswered. There exists a growing body of evidence that transformation paths are not univocal, but depend on equipment and experimental conditions used. Gaseous impurities and contaminants from grinding media can play an uncertain role in determining the final products [3]. However, our concern here is to focus on the intrinsic parameters of the MA/MM processes. The milling time and the ball-mass to powder-mass ratio are very often the only parameters given to characterize an experimental set up. These data are totally inadequate to describe the inherent mechanism involved. The majority of experiments have been carried out in commercially available machines which differ in their mechanical action and milling efficiency so impeding cross-comparisons and useful correlations. As a consequence, a suitable organizational framework has not been developed to reconcile the sometimes conflicting results obtained in different laboratories with different devices. On the other hand, extensive modeling efforts give a realistic description of the basic physics and also provide some hints concerning the characteristics of the final product [4-9]. It has been argued that the kinetic energy transferred at the collision event and the impact frequency are two of the main parameters governing the various transformation routes by which a particular reaction proceeds. The measurement of these key parameters however is a worrisome problem and no systematic attempts have been undertaken along these lines. This suggested to us to establish a methodological approach and to develop the related experimental procedures for simultaneously evaluating impact frequency and shock energy in the course of MA/MM processes. The method was conceived for and applied to vibromachines and mechanochemical reactors set up in our laboratory. In this brief report however, we present results obtained using a commercial mixer/mill.

Outline of the experimental procedures

The Spex Mill mod. 8000 that we used in the following experiments, has been modified to meet the requirement of adjustable speed. Usually it can be equipped with two motors which operate at 1725 RPM (115 V, 60 Hz) or at 1425 RPM (230 V, 50 Hz) corresponding to 17.7 or 14.6 mill cycles per second (cycles/sec). Noticeable differences were observed depending on the two milling arrangements. Our reported results refer to data obtained at 14.6 cycles/sec, as continually monitored by an optical tachometer.

Our initial efforts were to set up a simple technique for the direct measurements of collision frequency. Numerous sensing methods have been devised to measure transient shock in seismic and vibrating systems and each application needs to be careful considered. A complicating factor in vibro mills and shakers is the distributed nature of the mechanical system; the frequency spectrum experienced in machine tools overlaps with the frequency at which the sensor naturally vibrates when excited by a pulse input. Furthermore, a distribution of largely different amplitudes characterizes the impact events. Eventually, we resorted to a piezoelectric transducer (RS Component) coupled with a signal amplifier and a two channel analyser (TP208 20MHz, Tie Pie Engineering). The resulting device is quite simple, reliable and inexpensive. Two sensors were fixed on the exterior wall and cover of the vial and a single ball was employed (12.7 mm in size for a mass of 8.3 g). Each impact appears as a rapidly oscillating signal fading out to the background level before the following collision event. Typical traces are presented in Figure 1.



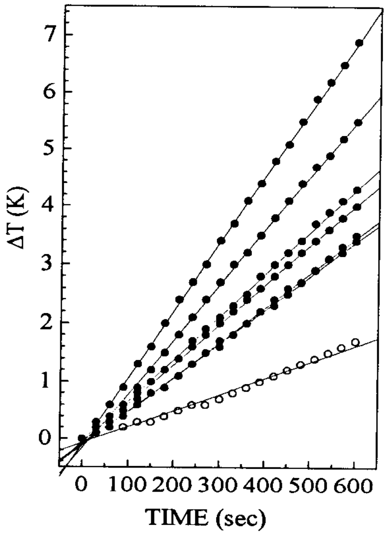


Figure 1. Snapshot of the colliding events registered in 360 msec by two piezoelectric sensors located at the lateral side (upper trace) and on the cover (lower trace) of the vial. A load of 14.2 g of Ni power was employed.

Figure 2. Increase of the vial temperature as registered at the beginning of the milling treatments for the empty vial (open circles) and for increasing Ni powder charges (darkened circles); from below: 3.6, 5.3, 7.1, 14.2, 20.0, 28.5 g.

Apart from the inherent power of the impact, signal intensities depend on the impact loci in respect to the transducer position. An on-line computer matches the two registered traces, marks corresponding signals according to their simultaneity and discriminates on the basis of the amplitude. A possible way to evaluate the intensity of the registered impacts is to produce a calibrated shock by allowing the ball to free-fall on to the inside of the vial. This method provides an absolute calibration, but in practice, there are uncertainties associated with the differing responses that the sensors give to isoenergetic impacts occurring at various sites in the vial. This compelled us to use calorimetric techniques to measure the average shock power.

It has been shown that the average kinetic energy of the colliding media can be approximated by the amount of heat released during the mechanical treatment [10]. It is reasonable to assume that the kinetic energy transferred to the powder is transformed in thermal energy through subsequent relaxation phenomena. The validity of this approach stems from the experimental evidence that only a few percent of the impact energy is stored in the processed material as lattice defects and plastic deformations. The absorbed energy diminishes progressively as the comminution and deformation proceed. At the saturation level, additional energy storage for permanent deformation is impeded.

Commercial thin lamellar-shaped Pt resistors (RS Components) were fastened by a thermal conductive adhesive on the vial under an insulating sheath. Quasi adiabatic conditions were realized. The heat capacity of the empty or charged vial, the ball included, here after referred to as C_v, was preliminary determined by heating the system with a carefully measured quantity of electrical energy; to this, a coil of a calibrated fine wire inside the vial was connected with its insulated leads crossing the vial cover.

The increase of the vial temperature as a function of the milling time was registered in parallel with the survey of the collision events. Thermal gradients, $\partial T/\partial t$, were obtained by a linear regression routine. Figure 2 shows the experimental trends for the empty vial and for different powder charges. The following equation has been employed to derive the energy flux intensity I (I in watt): $I = \partial T/\partial t \; C_v \; M_t$, where M_t stands for the total mass of the working system. From this relation, the energy dissipated per hit, E (E in joule) is then obtained by dividing I by N, the collision frequency (hits/sec). It should be noted that E is an averaged quantity.

Ni powders from Aldrich, 100 mesh 99.999%, were used in the trials.

Results and discussion

The number of the registered impacts and the operative conditions vary greatly as a function of the powder mass. This can be immediately perceived comparing the hystograms reported in Figure 3: the time intervals for 70 consecutive hits are plotted according to their sequence for the empty vial (left side) and for a load of 14.2 g of nickel. In the former case, in which 172 hits/sec were averaged over a time of ten seconds, the majority of the longer intervals are followed by a number of progressively shorter times and it can be inferred that a number of rebounds is generated at each collision event. The ball is not able to dissipate totally its kinetic energy and an uncontrolled trajectory originates apart from the throwing motion of the vial. Thus a definite periodicity is not discernible to characterize the ball motion inside the empty vial.

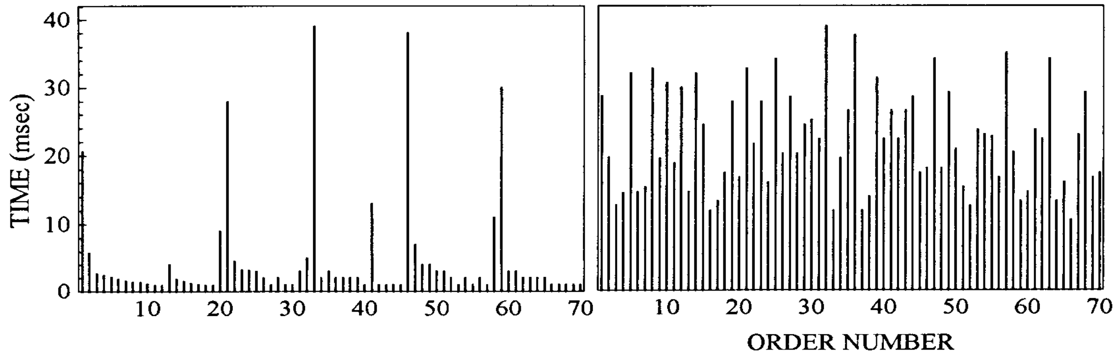
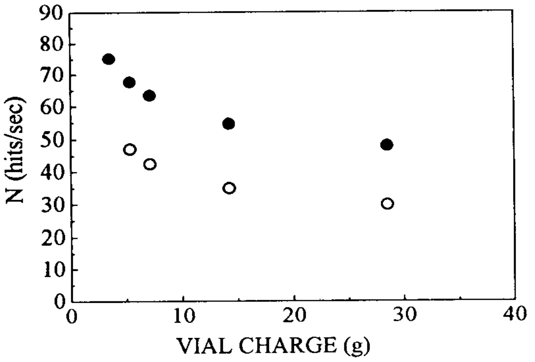


Figure 3. Time between consecutive collision events for the empty vial (left side) and for a nickel load of 14.2 g.

Different inferences can be drawn from the hystogram on the right hand side of Figure 3. One notices the absence of the shortest components. The times between collisions are now spread over a narrower band. We believe that the cushioning effects from the trapped and adhering powder create inelastic shock conditions and are to be considered responsible for this behaviour.

To pursue this correlation somewhat further, impact frequencies were monitored as a function of increasing loads. Data were registered for the first ten minutes of the mechanical treatment. As shown in Figure 4, the collisions per second drop to about 70 for an initial charge of 3.6 g and progressively tend to a limit value. It is interesting to note that an additional decrease is observed when Ni powders are mechanically pre-treated up to 24 hours (open circles in Figure 4). The relevant frequency limit shifts to around 29 hits/sec. This final result is complemented with and confirmed by the trend presented in Figure 5. Here the impacts per second relevant to a constant Ni charge of 28.4 g are plotted as a function of the milling time.



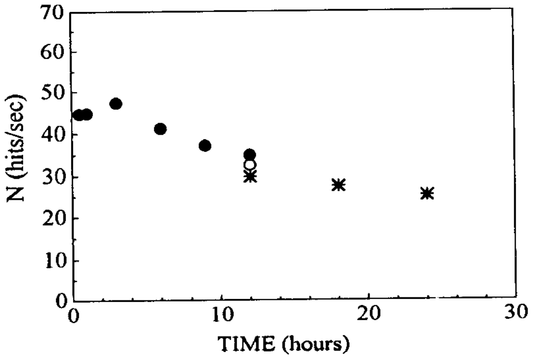


Figure 4. Hit frequency as a function of the Ni powder charges (darkened circles); the open symbols refers to Ni powder pre-milled up to 24 hrs before the measurements of the collision frequency.

Figure 5. Hit frequency registered *in situ* as a function of the milling time for a constant charge of 28.4 g Ni powder. Different marks refer to differing trials.

It is possible to draw some conclusions from this data.

In respect to the empty vial, impact frequency is drastically reduced under real milling conditions. A constant plateau is actually observed pointing out that a definite amount of material is necessary to minimize rebound phenomena and therefore to approach inelastic conditions. Keeping in mind that the kinetic energy is totally released when these ideal conditions are met, the vial charge appears as a critical parameter to fulfil this requirement as well as to maximize the efficiency of the milling action. Impact frequency also depends on the structural conditions experienced by the powders which evolve during a milling run. As a consequence, modeling data for collision geometry and ball kinetics in the absence of powder seem of little value when extrapolated to rationalize fracture and coalescence phenomena, deformation events and structural reconstruction paths.

The 29 hits/sec obtained as limit value require additional comment. There is an interesting coincidence with the natural frequency of the mill, 14.6 cycles/sec. This full correspondence suggests that the lateral displacements of the vial are of little effect and that its main component can be correctly approximated by an angular harmonic motion, as already pointed out [5]. In a simplified view, the ball accelerates along with the vial up to the maximum velocity, then detaches from the vial underside continuing its motion on a rectilinear free trajectory at a constant velocity. Under the hypothesis of absolutely inelastic collision, the ball impacts the vial base each half cycle, so reaching 29 hits/sec. Considering that the main swing occurs through an arc of approximately 40° at the end of a 8.9 cm arm, the essential motions of vial and ball can be represented by simple equations. Taking into account the actual dimensions, a graphical solution to these equations is easily obtained providing the angular velocity of the vial along its displacement and the time of the collision event. The ball velocity at the detachment, which corresponds to the maximum velocity reached by the vial, is 2.86 m/sec, giving a kinetic energy of 0.034 J. It must be pointed out however that the ball collides on the vial after this latter has passed its extreme displacement point and has already acquired speed in the reverse direction: its calculated velocity at the impact time is 1.75 m/sec. It can be therefore assumed that the relative velocity of the ball at the collision is 4.61 m/sec corresponding to a kinetic energy of 0.088 J for the ball of 8.3 g. The energy range so obtained is just a rough guideline, nevertheless it constitutes the natural frame of reference to which we can compare the collision energy intensity estimated by the calorimetric method.

In this regard preliminary tests were carried out concerning the effect of the ball collisions in the empty vial. From the registered thermal gradient, an I value of 1.02 watts was obtained. Using the previously determined 172 hits/sec, an energy dissipated per hit can be calculated as $5.9 \cdot 10^{-3}$ joules.

There are two important factors that can justify this value standing far below the above calculated energy range. The first being that under elastic conditions, as in the case of the empty vial, the ball does not totally dissipate its kinetic energy. The second factor being the decrease of the ball velocity following an elastic collision. A series of confounding rebounds prevent the ball from returning to its proper position before the subsequent detachment and therefore reducing the ability of the vial to accelerate the ball up to its maximum velocity . This series of side collisions can be seen in Figure 3. Both the reduced velocity and the inefficient transfer of energy contribute to the marked difference here observed.

As for the energy dissipated per hit during the powder processing, it must be initially noted that elastic conditions depend on the peculiarity of the adhering powders in creating dissimilar coats on the colliding surfaces. These coats can change for the same sample through the milling process, ranging from highly polished deposits to coarse uneven layers. As a consequence, the actual fraction of the kinetic energy transmitted at the impact event is effected by the tribological properties of the powders even if the kinetic energy of the colliding tools is a constant parameter determined only by the mill dynamics and the ball mass. Therefore is not possible, from a specific series of experiments, to derive general quantities encompassing a variety of distinct systems. The following Ni results must be considered within this emerged framework.

The $\partial T/\partial t$ data are collected in Figure 6 as full circles together with the value pertaining to the empty vial.

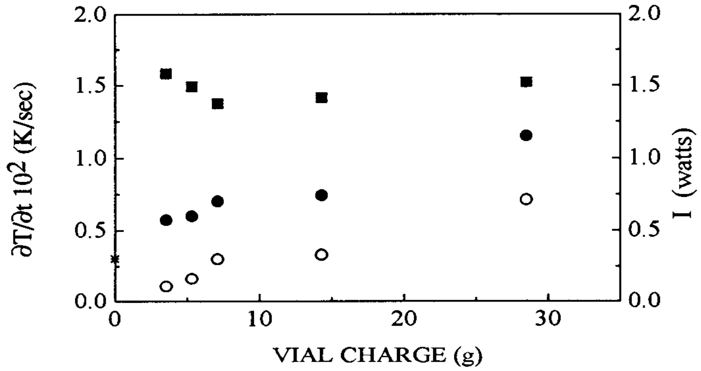


Figure 6. The darkened circles show the $\partial T/\partial t$ behaviour for the increasing charges of the Ni powder. The symbol at zero load marks the value registered for the empty vial. The contributions to the thermal gradient due to inherent effects of the powder shaken inside the vial without the ball are presented as open circles. The side by side differences of the reported values quantify the heat transfer due to the collision events alone and are plotted in watt (square): the pertinent scale is presented on the right hand side of the figure.

Two separate contributions must be taken into accounts to evaluate the whole behaviour that thermal gradients follow in function of the powder amounts. It has been initially surmised that the heat released at the ball collision was the primary event by which kinetic energy is transferred to the vial [3]. Actually a significant amount of the heat transfer arises directly from the microcollisions of the powder particles against the vial walls and from frictional and wearing powder effects. This quantity was measured by operating the vial loaded only with the powder charges. The relevant data are plotted in Figure 6 as open circles. This contribution is so important as to determine the overall $\partial T/\partial t$ trend. It must be noted that extremely large powder/ball mass ratios were used for these trials further exaggerating these effects. The powder mass contribution must therefore be subtracted from the total thermal increase to isolate the heat transfer due to ball collision events alone. Resulting data are plotted in Figure 6 directly in watts as I values. Within the experimental uncertainties, the power flow appears constant across the range of powder loads we explored. The average value stands as 1.47 watts. A further increase to 1.80 watts, which we believe is experimentally significant, pertains to the Ni powders premilled up to 24 hours. Following division by N, the energy per hit progressively increases, reaching its maximum of 0.062 joules at the frequency of 29 hits/sec. This corresponds to a relative impact velocity of 3.8 m/sec, within the range of the forecasted values.

Conclusions

We believe that we have produced a coherent description of the energy transfer processes in the Spex Mill, also providing a reliable in situ evaluation of the most important average and local milling parameters. In addition to the relatively good agreement between experimental and forecasted values, we would like to point out that the methodological approach briefly outlined here, gives a consistent view when the mill velocity was changed and different powders were processed. The capability of the method was also tested with a laboratory vibro-mill, for which the simpler motion allows for a more exact definition of the geometry and basic mechanics. A more general exploitation of this technique is therefore possible.

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References

[1] "Mechanically Alloyed and Nanocrystalline Materials", Proc. Int. Symp. on Metastable Mechanically Alloyed and Nanocrystalline Materials, A. R. Yavari Ed., Trans Tech Publication Ltd. Switzerland, 1995.

[2] Materials Transactions, JIM 36 (2), February 1995: a special issue on Mechanical Alloying.

[3] G. Cocco, G. Mulas, L. Schiffini, ref.2, pag. 150.

[4] R. B. Schwarz and C. C. Koch, Appl. Phys. Lett. 4, 146 (1986); R. M. Davies, B. T. McDermott and C. C. Koch, Metall. Trans. 19A, 2867 (1988); C. C. Koch, J. of Mechanochem.and Mech. Alloying 1, 56 (1994); ref.2, pag. 85.

[5] D. R. Maurice and T. H. Courtney, Metall. Trans. A21, 289 (1990); J. of Metals 44, 10 (1993); Metall. Mater. Trans. 25A, 147 (1994). T. H. Courtney, ref.2, pag.110.

[6] E. Gaffet and L. Yousfi, Mat. Sci. Forum 88-90, 51 (1988); E. Gaffet and M. Harmelin, Mat. Sci. Eng. A19, 185 (1989); M. Abdellaoui and E. Gaffet, J. All. Comp. 209, 351 (1994); ref.1, pag. 339; E. Gaffet, M. Abdellaoui and N. Malhouroux- Gaffet, ref.2, pag.198.

[7] N. Burgio, A. Iasonna, M. Magini, S. Martelli and F. Padella, Nuovo Cimento 13, D459 (1991); F. Padella, E. Paradiso, N. Burgio, M. Magini, S. Martelli, W. Guo and A. Iasonna, J. Less-Common Metals 175, 79 (1991); M. Magini and A. Iasonna, ref.2, pag.123.

[8] H. Hashimoto and R. Watanabe, Mater. Sci. Forum 88-90, 89 (1992).

[9] P. Le Brun, L. Frojen and L. Delaey, Mater. Sci. Eng. A161, 75 (1993).

[10] P. Yu. Butyagin and I. K. Pavlichev, React. of Solids 1, 361 (1986); A. N. Streletskii, Proc. Int. Conf. on Structural Applications of MA, J. J. de Barbadillo, F. H. Froes and R. B. Schwartz Ed.s, ASM International, Materials Park, Ohio, USA (1993), pag. 51.