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Optimization of Nanocrystalline Aluminium Production Using an Objective Function

Joan Solà-Saracibar, Jordi Llumà, Jordi Jorba

Dept. Materials Science and Metallurgical Engineering, EUETIB, UPC, Barcelona, Spain, jordi.lluma@upc.edu

Abstract
In recent years there has been an increasing interest and research effort focused on the development of nanocrystalline materials due to important improvement of their chemical, electrical, magnetic, optic and mechanical properties. Those materials can be produced by a wide variety of processes, for example: ball milling (BM), rapid solidification, equal channel angular extrusion (ECAE) or thermal spray processing. Several of these techniques (like BM) produce powdered materials which require a subsequent consolidation for any structural application. However these techniques obtain smaller grain size and can produce special intermetallic materials. For this reason, extensive studies of the dynamics of the powder processes and their influence on microstructural changes of materials have emerged. Nevertheless, many of these efforts are focused on the understanding of the process and on the improvement of the materials properties and they are not on the process output.
In the present paper, a methodology to improve the output of powder by the BM process is proposed. This methodology has been tested with pure aluminium powder and proposes an objective function and its optimization by a simplex downhill method. After 9 iterations, the output of nanocrystalline aluminium is increased in a 35%, the powder particle size is reduced to the half and the hardness is slightly increased.
Keywords: Ball milling, nanocrystalline materials, aluminium, output optimization.

1. Introduction
The remarkable improvement in the chemical, electrical, magnetic, optical and mechanical properties obtained in nanocrystalline materials has favoured an important increase in their study during the last decade [1],[2]. The processes which produce these nanocrystalline materials are many and diverse, but those ones which make possible smaller crystalline grain, often produce powder material which requires a subsequent sintering process to produce useful components for structural applications. In particular, several papers report the influence of milling parameters on the material microstructure and final properties as a consequence of mechanical milling [3][4][5], and there has been reported that this process allows reaching nanocrystalline grain sizes in several metals [6] and alloys [7].
However, the main effort has been focused on ferrous materials, less on aluminium alloys [8] and very little on pure aluminium. Due to that, there are few studies about pure aluminium that has not prevented detailed description of the evolution of the aluminium microstructure when cold work is applied [9] which could be broadly extrapolated to mechanical milling effect, and specific studies about mechanical milling applied to pure aluminium [10].
Probably, because of all research are not in industrial state, the interest in processes efficiency is poor and most studies focus on the influence of only one parameter, usually milling time, on the mechanical properties of powder and neglects the effect of other milling variables, such as impact energy (related with mill speed) or number of impacts per second (connected to the number of balls in the container). Nevertheless, the effect of these variables on the mechanical properties and on the powder particle size seems to be similar, but differs significantly on the output process [11]. This evidence opens the possibility to optimize the efficiency of the process keeping constant, or even improving, the powder quality. This is the aim of the present work.

2. Optimization method
Any optimization method needs an optimization algorithm and a quality parameter which, if it is possible, should be defined as a combination of quantitative properties related with the process and the outputs. This combination is the objective function to be optimized and the numerical values of the quantitative properties are determined experimentally. In the present work, material hardness, powder particle size and process efficiency have been considered in the objective function, and milling time (t), ball to powder ratio (BPR), milling speed (rpm), work-rest interval (WI) where interval of working time and resting time are the same, and control agent percentage (EBS) have been used as process parameters to be modified.
The simplest objective function that considers hardness and efficiency as a parameter to be increased is:

$$F_0 = H \cdot \frac{r}{t} \tag{1}$$

where H is the hardness of the material and the efficiency is expressed as the mass recovery (r) per time unit (t). Unfortunately Eq. (1) has a trivial maximum when time tends to zero which will make to fail any optimization algorithm.

In order to avoid this unsuccessful result, some cut-offs should be introduced to guarantee certain value properties in the product, for example a minimum in hardness of the product to ensure a nanometric grain size and a maximum particle size to ensure a proper filling of the die during the compression process before sintering.

Although it is possible to use a step function as a cut-off function, the optimization algorithms work better with a smoothed step functions, such as the Boltzmann one. Then the objective function could be written as:

$$F = H \cdot \frac{r}{t} \cdot \left[\frac{1}{1 + e^{-\frac{H - H_0}{\Delta H}}} \right] \cdot \left[\frac{1}{1 + e^{\frac{D - D_0}{\Delta D}}} \right] \tag{2}$$

where H_0 and D_0 are the half-step values of hardness and particle size, and ΔH and ΔD are the step width.

Despite of Eq. 2 could be maximized by any optimization algorithm; the presence of experimental errors in the experimental measurement of the numerical values strongly recommends optimizing that function with a robust algorithm instead of an efficient one. Moreover, the optimization algorithm must work in a discrete space because some parameters of process are not continuous as a consequence of the balls mass is discrete or constrictions in the milling parameters. Due to that, the downhill simplex algorithm [12] was chosen but one modification was made. The multiple contractions was discarded during each iteration because the time required for each sample preparation and experimental measurements is huge compared with the computing time required in each single contraction.

3. Materials and experimental procedure

The original material used in this study is a pure aluminium powder supplied by the company ECKA Granules ® as ECKA Aluminium AS 51. It has been screened at the laboratory and only the fraction between 72-100 µm is used. Figure 1 shows the distribution of particle size of the sieved fraction determined by image analysis of the projected surface of the particles [13]. Figure 2a shows a SEM image of the morphology of the particles and Figure 2b an optical picture of the section of a particle. The hardness of the original powder is 30,0 ± 2.7 HV 0.025 determined using the method described later.

Approximately 7.5 g of aluminium powder has been milled in Fritsch's Pulverisette 5 planetary ball mill using cylindrical containers of 250 ml made with X 5 Cr Ni 18 10 steel and 100 Cr 6 steel balls of 10 mm of diameter. The milling has been made according different sets of the process parameters time (t), ball to powder mass ratio (BPR), mill speed (rpm), work-rest interval (WI) and control agent Clarient's Licowax C amide wax (EBS).

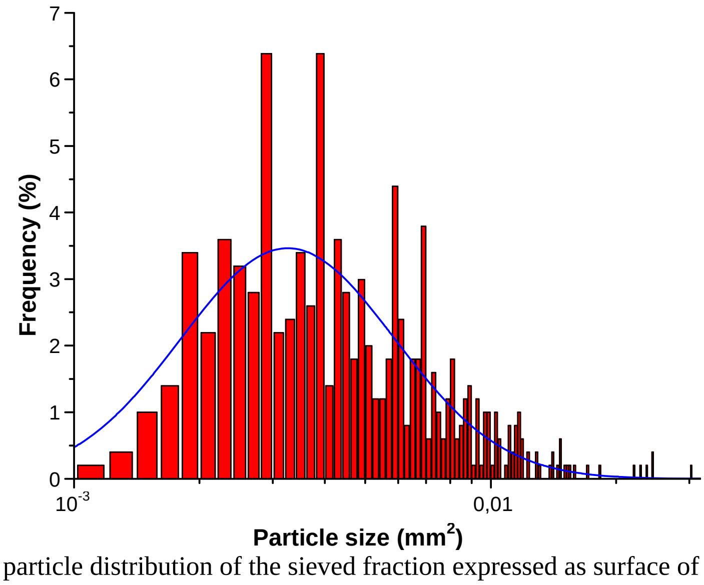

Figure 1: Size particle distribution of the sieved fraction expressed as surface of projected area.

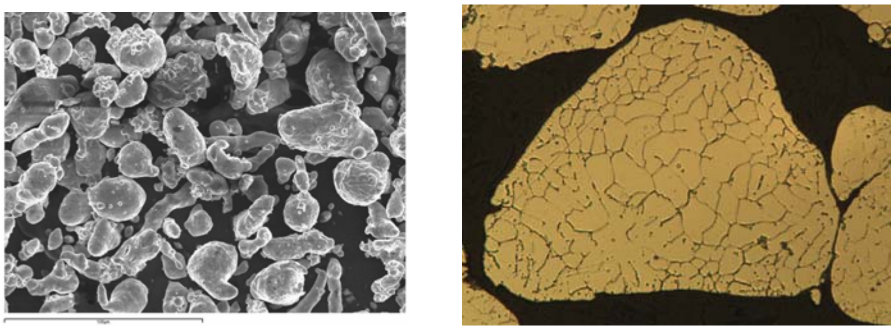

Figure 2: SEM image of the sieved powder morphology (left) and optical image of a particle's interior (right).

In each mill the recovery was determined as the percentage ratio between the mass of the recovered aluminium powder and the initial one. The particle size was determined as the mode of a lognormal distribution fitted to the histogram of the projected area determined by UTHSCSA Image Tool 3.0 of at least 400 particles. After that, and in order to determine the hardness of the material, the sample was embedded in hot cured (150 ºC) high strength epoxy resin (Buehler's Epomet) and polished following standard metallographic procedure to approximately the half of powder diameter. The hardness was determined in Vickers scale (HV 0.025) in a Buehler's Micromet 5114 micro indentation hardness tester. The Cauchy criterion was used to reject wrong experimental values and the hardness value reported is the average value of at least 15 valid measures.

The experimental values were introduced in Eq. 2 to obtain the score of the sample, fixing the cut-offs at H_0 = 95 HV 0.025 and D_0 = 0.5 mm^2 and the width of the steps at ΔH = 2 HV 0.025 and ΔD = 0.1 mm^2.

The starting point of the simplex algorithm was selected between the best scores calculated with Eq. 2 using the results obtained in 50 samples of a previous work [11] that were milled in processes with linearly independent parameters. The values of these parameters are shown in Table 1.

Table 1: Starting conditions of the simplex.

Process parameters					Experimental values			**Score**
EBS (%)	*BPR*	*WI* (min)	*t* (h)	*rpm* (rpm)	r (%)	H (HV0,25)	D (mm^2)	
0.4	*15*	*30*	*20*	*160*	98.6	108.0	0.22326	**500.12**
0.8	*20*	*30*	*20*	*160*	97.2	103.1	0.01135	**488.91**
0.4	*20*	*12*	*20*	*160*	97.8	98.9	0.16727	**408.47**
0.4	*20*	*15*	*20*	*160*	90.7	100.0	0.32434	**357.98**
0.0	*20*	*30*	*15*	*160*	80.9	98.3	0.46333	**262.67**
0.0	*20*	*30*	*20*	*120*	82.2	99.3	0.55899	**130.37**

Then a simplex iteration was run to obtain a new proposal of process parameters. Because of process parameters are discontinuous, proposal should be approached to the closest possible value; so any BPR is accepted and the

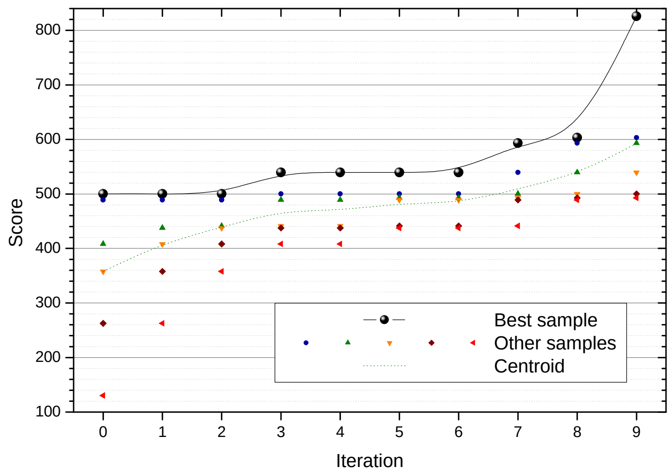

Figure 3: Evolution of samples score with Simplex iterations.

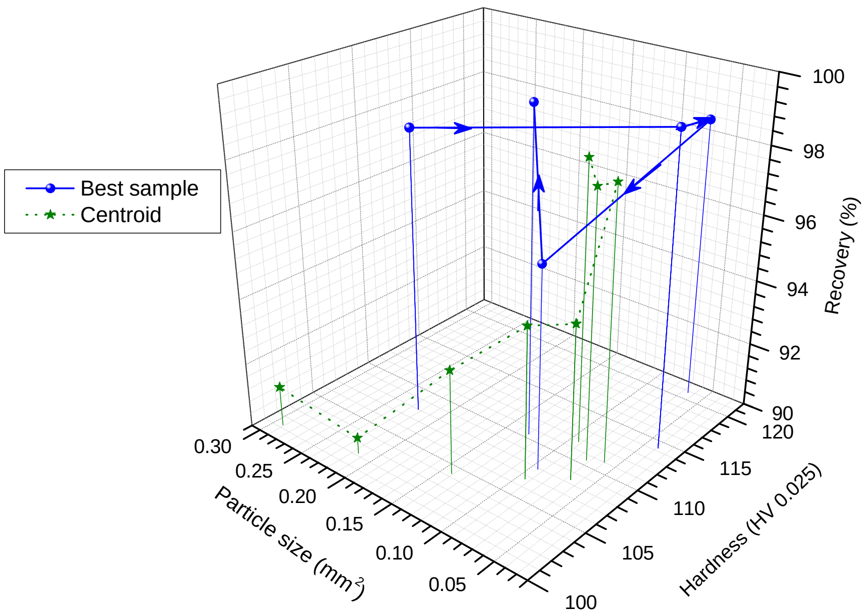

Figure 4: Trajectory of the experimental values of the best sample and the simplex centroid.

number of balls and powder mass are chosen to fit powder weight as much closer to 7.5 g as could be possible keeping integer the number of balls, WI is rounded to a minute precision, t to an integer number of WI and rpm to a multiple of 10. This set of parameters was used to mill a new sample that was measured and rated before introduce the values in a new simplex iteration.

Results

The different values of simplex iterations with process parameters, experimental values, simplex actions and score obtained using Eq. 2 are shown in Table 2.

4.1 Process parameters.

Due to simplex is a multidimensional optimization algorithm, all the process parameters varies with each new proposal calculated with simplex. The analysis of EBS percentage was initiated at high EBS values but gradually tends to decrease until the initial value of 0.4 % as a consequence of the reduction induced in the mechanical properties. On the other hand, BPR oscillates between 14 and 18 indicating that the initial value 20 BPR is slightly high. The milling speed (rpm) started at high values because high rpm helps to improve the powder hardness, but the energy concentration induced by the balls kinetics motion also increases the adhesion on the container surface unless the concentration of EBS is increased. As EBS was reduced, the speed must be also reduced. As there has been shown in a previous work [13], the WI parameter has little influence on the mechanical properties, for this the reason, is less stable and oscillates between 12 to 60 minutes.

Table 2: Values of simplex iterations

Simplex suggestion	*Process parameters*					*Experimental values*			*Add*	*Simplex Action*	**Score**
	EBS (%)	*BPR*	*WI* (min)	*t* (h)	*rpm* (rpm)	r (%)	H (HV0,25)	D (mm^2)			
1	0,80	18,04	17	18,13	200	77,7	104,8	0,10245	Yes	Reflection	**437,53**
2	1,12	17,26	12	24,20	180	97,8	111,0	0,02000	Yes	Reflection	**444,78**
3	1,01	16,17	25	20,83	180	99,3	114,3	0,03897	Yes	Reflection and expansion	**539,40**
4	1,32	14,25	30	21,00	190	98,8	96,0	0,00305	No	Reflection	**279,20**
5	1,26	14,64	34	21,53	190	98,3	108,8	0,00292	Yes	Reflection	**492,76**
6	1,04	15,29	35	24,50	150	98,0	45,8	0,00199	No	Contraction	**0,00**
7	0,86	17,32	22	19,80	190	98,5	120,8	0,06495	Yes	Reflection	**593,29**
8	0,61	16,00	44	16,87	170	96,1	107,9	0,09406	Yes	Reflection and expansion	**603,27**
9	0,36	15,35	60	13,00	160	99,7	110,4	0,12972	Yes	Reflection	**825,94**
10	0,04	17,30	38	14,57	150	Not studied					

4.2 Score
Figure 3 shows the evolution of the score of the simplex centroid and all the samples involved in a simplex iteration. The rating of the centroid is always improving, except when the simplex explores a wrong area (with powder no enough hard) and must reflect, then the value maintains. As usual, the trends of the best and worse results are more abrupt and one extreme point must remain constant when the other improves.

4.3 Experimental values
The trajectory of the experimental values as a function of simplex iteration is shown in Fig. 4. As it is expected the hardness and recovery of the samples almost always are increasing because they are present in the numerator of the objective function (Eq.2), but the reduction in particle size is more surprising because the effect of this value on the objective function is very low when this value is far from the cut-off ($D < 0.2\ mm^2$). The properties of the best samples at original stage and after 9 iterations are shown in Table 3.

Table 3: Initial and final conditions of the simplex.

Iteration	*Process parameters*					Experimental values			**Score**
	EBS (%)	*BPR*	*WI* (min)	*t* (h)	*rpm* (rpm)	r (%)	H (HV0,25)	D (mm^2)	
0	*0.4*	*15*	*30*	*20*	*160*	98.6	108.0	0.22326	**500.12**
9	*0.36*	*15*	*60*	*13*	*160*	99.7	110.4	0.12972	**825.94**

Although the simplex still is far from the convergence (as it is shown in Fig. 3), there are a slight improvement of the recovery (1%) and hardness (2%) after 9 simplex iterations, but an impressive reduction of the particle size (42 %) and the production time (35 %). At this point the milling containers were successfully redesigned to keep the gas leaks bellow 10^{-3} Pa·l/s that to allow us a correct control of the milling atmosphere. At this point, the simplex algorithm was stopped and restarted including this new process parameter. The new results will be published elsewhere.

4. Conclusions

The proposed methodology could satisfactorily optimize an industrial process, as the optimization of the production of nanocrystalline aluminium powder shows.
In fact, only 9 iterations of simplex using the proposed objective function have produced a 35% increase in productivity and a 42% reduction of particle size compared with the best result previously obtained in a standard set of experiments to scans the influence of the process variables in the powder properties.

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