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The influence of milling conditions on the Ni_3Fe alloyed powder

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Abstract: The mechanosynthesis of Ni_3Fe intermetallic compound was carried out in a planetary mill. The effects of milling parameters such as balls diameters and ball milling speed defined by the vials rotation speed (ω) and the disk rotation speed (Ω), on morphology, microstructure and particle sizes of Ni_3Fe powder were studied. It was found that the impact frequency represented by the number of balls from vials is an important parameter a milling process. The smaller grain sizes and particle was obtained when milling process was performed in high frequency rate of impacts together with high balls velocity, meaning high energy conditions.

Introduction

Mechanical alloying is one of the mechanosynthesis methods involving the synthesis of materials by high-energy ball milling of powders. This technique is used to obtained structures far from equilibrium including here amorphous alloys, superalloys, metastable crystalline phases and nanosized materials, extended solubility and intermetallic compounds [1-3]. The base mechanisms of mechanical alloying process are successive cold welding and fracturing phenomena of the powder particles thus leading to the formation of new alloys or composite materials. The efficiency of the process is dictated by milling parameter [4]. These parameters such as milling time, milling speed, ball-to-powder weight ratio and process control agent effects the final structure and morphology of the powders [1]. A major influence on mechanical alloying products structures have the milling energy described by the ball velocity, kinetic energy transferred to powder during milling process and the frequency of impacts. The influence of all of these parameters on final structure of powder processed by mechanical alloying was investigated in several researches [5, 6]. It has been clearly established that all these parameters are addicted by the Ω/ω ratio, where Ω is the angular speed of the plate of the mill and ω – is the angular speed of vials, the balls dimension and their weight [7-9]. The newly formed alloys by mechanical alloying has a in general nanocrystalline or amorphous structure. Fe – Ni alloys around Permalloy composition are well known for their performance as soft magnetic materials. The alloy from Fe-Ni system obtained by mechanical alloying process has been the subject of several researches during the years [10, 11]. Among the alloys from Ni-Fe system, the synthesis of Ni_3Fe intermetallic compound by mechanical alloying from elemental powders is one of interest [12, 13]. Their applications and capability are limited at high frequency by eddy current loss [14]. The magnetic properties of Ni_3Fe powder are influenced by their particle sizes not only by their internal structures. In this work we were focused on influence of milling parameters such as milling energy (Ω/ω ratio, number of balls and their dimension) on structure and particle size of Ni_3Fe intermetallic compound powder obtained by mechanical alloying technique.

Experimental

123- carbonyl nickel powder with particle sizes less than 10 µm and NC100.24 iron powder with particle sizes up to 40 µm were used. The powders powder mixture was homogenized 15 minutes in a Turbula type blender. The mechanical alloying was performed in a high energy planetary ball mill (Fritch Pulverisette 4) up to 16 hours. In order to prevent the powder oxidation the milling process was carried out in argon atmosphere. The ball to powder mass ratio – BPR was 10:1. Two milling conditions and two types of balls were used. First condition involve Ω/ω = 800/400 and the second Ω/ω = 350/350 (Ω - is the rotation speed of the disc on which the vial holders; ω - rotation speed of vials) were fixed. In both conditions was used ball with 11 mm and 14 mm diameter. The filling degree of the vials was 50 % (volume percent). Ball to powder weight ratio was 10:1. The systems of milling condition are noted as follow: I (balls diameter with 11 mm and ω/Ω = -400/800), II (balls diameter with 14 mm and ω/Ω = -400/800), III (balls diameter with 11 mm and ω/Ω = -350/350) and IV (balls diameter with 14 mm and ω/Ω = -350/350) and are given more explicitly in table 1. Value for v_c - balls velocity and E_c – kinetic energy of balls was calculated with following relations [8, 9]:

$$\overrightarrow{\| v \|}^2 = (R\Omega)^2 + (r - r_b)^2 \omega^2 (1 + \frac{2\omega}{\Omega}) \quad (1)$$

where: v – ball velocity, R – distance between disc center and vial center, r – vial radius, r_b – ball radius, ω – rotation speed of vials and Ω - rotation speed of disk.

$$E_k = m \frac{\overrightarrow{\| v \|}^2}{2} \quad (2)$$

where: E_k – kinetic energy of ball, m – ball mass.

Table 1. Kinetic energy and balls velocity for various milling conditions.

Milling condition types	**D_{ball} (mm)**	**ω/Ω (rpm)**	**V_c (m/s)**	**E_c (J)**
I	11	-400/800	1.22	0.89
II	14	-400/800	0.98	1.19
III	11	-350/350	0.67	0.13
IV	14	-350/350	0.66	0.26

Several milling times, ranging from 1 to 16 hours has been chosen for collecting samples. The powder was investigated by characterized by X-ray diffraction using a Dron 3 diffractometer with Co K_α radiation ($\lambda_{CoK\alpha}$ = 0.179026 nm) were recorded in angular range 2θ = 85 ÷120°. The crystallite size and lattice strain was calculated using Williamson-Hall formula [15]. A JEOL-JSM 5600 LV scanning electron microscope (SEM) was used for investigation of particles morphology. The particles sizes distribution was analyzed by standard sieving method.

Results and discussion

In fig. 1 are presented the X-ray diffraction patterns of the starting sample (noted as ss) and the samples milled for 1, 2, 4, 8 and 16 hours. The milling type condition I was used and these are described in table 1. In the XRD patterns of the starting sample are noticed the Bragg reflections characteristic for the elemental Ni and Fe.

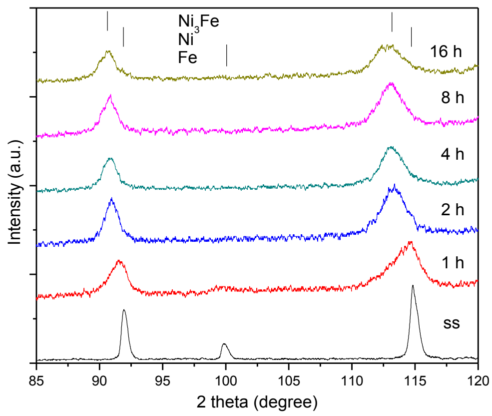


Fig. 1. X-ray diffraction patterns of the starting sample and the samples milled in type I condition for 1, 2, 4, 8 and 16 hour.

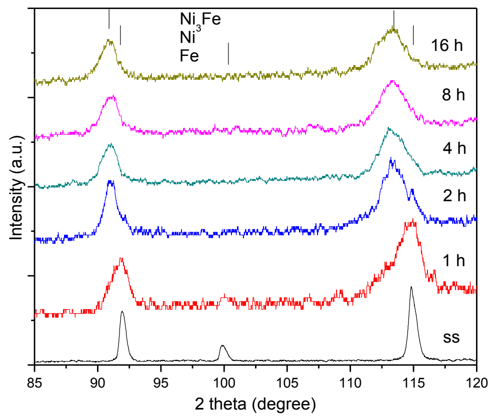


Fig. 2. X-ray diffraction patterns of the starting sample and the samples milled in type II condition for 1, 2, 4, 8 and 16 hour.

After 1 h of milling it can be shown the same diffraction maxima as for the ss (starting sample). The peaks are broadened indicating the crystallites size reduction and also the existence of the second-order internal stresses [16]. Also, it is observed that the Fe diffraction peaks are reduced in intensity and almost disappear from diffraction pattern. The Ni peaks are left shifted to Ni_3Fe position and the peaks profiles are asymmetric. This indicated that the new phase Ni_3Fe intermetallic compound begin to form. At 2 h of milling in diffraction pattern the peaks correspond to Fe does not exist. The new position of lines corresponds to Ni_3Fe intermetallic compound. Further, with increasing the milling time just the refinement of Ni_3Fe structure occurs. In fig. 2 are shown the X-ray diffraction patterns of powders milled up to 16 h in type II conditions. In the first hour of milling it can see that the Fe diffraction peak is not appearing. In diffraction pattern only Ni peaks are present. This indicated the dissolution of Fe atoms in Ni fcc matrix. The formation of a new phase (Ni based solid solution) is confirmed by Ni peaks shifted positions. With increasing the milling time, the peaks profile becomes asymmetric due to form the Ni_3Fe intermetallic compound. The asymmetry of peaks profiles is observed up to 8 h of milling. After 8 h of milling the peaks characteristic for intermetallic compound are almost at the same intensity as the one of Ni indicating that an important amount of Ni_3Fe was formed. One can notice is that using these specific milling conditions seems that the powder contamination with iron has been avoided or is at a lower level that XRD can detect.

Between 8 and 16 h of milling only the broadening of profile correspond of Ni_3Fe powder are produces. This is characteristics to crystallite sizes reduced and to internal stress induced in Ni_3Fe network. The evolution of the powder subjected to milling process in type III conditions is shown in fig. 3. In first 4 h of milling time only a broadening of characteristic line of Ni and Fe are produced. The broadening occurs due to the defects induced in powder by mechanical milling. After 8 h of milling it can be observed that the Fe peaks cannot be found in diffraction pattern. The Ni phase corresponding peaks are shifted to left, to 2θ position characteristics of Ni_3Fe intermetallic compound. Formation of the Ni_3Fe compound continues to increasing of milling time. At 16 h of milling the material according to X-ray investigation consists in a single phase: Ni_3Fe intermetallic compound. In the diffraction pattern can be noticed the symmetrical shape of Bragg peaks and the positions of those peaks are characteristic for the Ni_3Fe. Changing condition of milling (type IV) the corresponding X-ray diffraction patterns are shown in fig. 4.

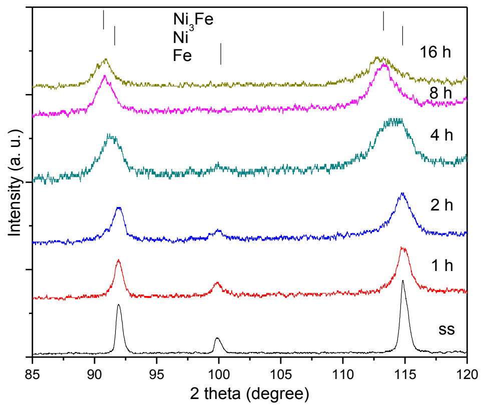


Fig. 3. X-ray diffraction patterns of the starting sample and the samples milled in type III condition for 1, 2, 4, 8 and 16 hour.

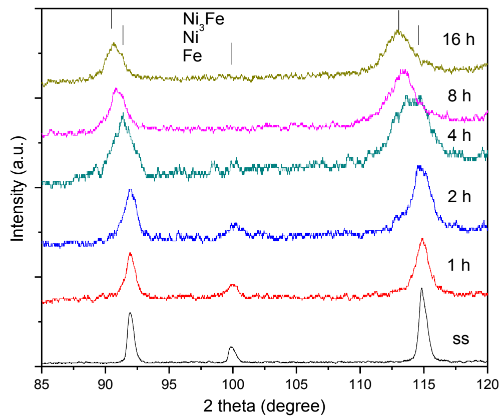
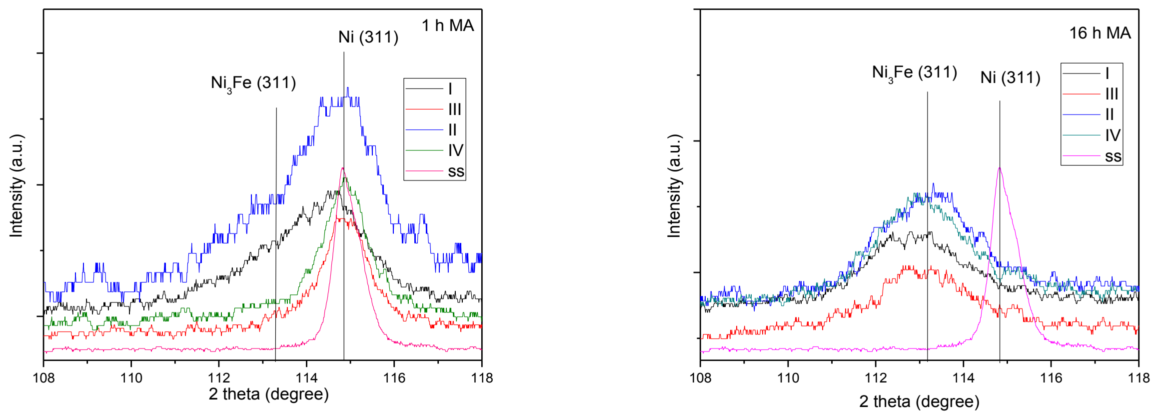


Fig. 4. X-ray diffraction patterns of the starting sample and the samples milled in type IV condition for 1, 2, 4, 8 and 16 hour.

It can been observed that the diffraction patterns are the same as it is correspond to starting sample up to 4 h of milling. The Fe and Ni characteristics lines are present in diffraction pattern after 1, 2 and 4 h of milling. Just a broadening of Bragg profiles are produce due to internal stress induced in powder by mechanical milling. At 4 h of milling the Ni peaks profile shows pronounced asymmetry. That is due to the Ni_3Fe intermetallic compound start to form. After 8 h of milling in this type of milling condition in diffraction pattern the Fe peaks are not be found. The positions of peaks correspond to 2θ angle characteristic for Ni_3Fe intermetallic compound after 16 h of milling.



a b

Fig. 5. Evolution of Ni_3Fe phase formation in different types of milling condition at 1 h – a, and 2 h of milling – b.

To study the influence of milling conditions on the formation and evolution of Ni_3Fe intermetallic compound obtained by mechanical alloying was represented the (311) Bragg peaks at 1 hour and after 16 hours of milling. This evolution is presented in fig. 5. After 1 h of milling of the start samples in all types of milling condition (fig. 5 a) it can be observed these:

- Position of the peak (311) in diffraction pattern correspond to 2θ angle value characteristic of Ni (311) for all types of milling condition;
- it can be observed that the peaks profile are more broadened for the sample milled in I and II types of milling condition than profiles correspond to III and IV milling condition;

- the asymmetry of the Bragg profiles are more clearly when the milling process was carried out in I and II types of milling condition due to start formed a new phase;

After 16 h of milling in all types of milling condition (fig. 5 b) it can see the following:

- in all the cases the position of characteristic lines from diffraction pattern correspond to 2θ value for (311) maxima of Ni_3Fe intermetallic compound, excepted the position of peak for powder milled in II condition;
- all the profiles of peaks become symmetrical after 16 h of milling;
- the intensity of peaks are smaller if mechanically milling process was performed in I and II types of milling condition than intensity correspond to III and IV conditions;
- profile of (311) peaks correspond to I and II types of milling condition become larger then peaks of Ni_3Fe obtained in III and IV condition after 16 h of milling.

Average grain sizes evolutions with milling time in al type of milling condition are shown in fig. 6. In first 2 hours of milling in type I and II milling conditions it can be observed a significant degrease of powder grains up to 35 nm and 27 nm, respectively. In the range of 2 and 8 h of milling in these conditions, the grain sizes are not affected if mechanical milling occurs in II type of milling conditions. The value for mean crystallite is about 35 nm. In the last stage of milling (between 8 and 16 h) the grain sizes is reduced from 35 nm to 22 nm. If the mechanical milling perform in type I milling conditions, in the range of 2 - 4 h there is continuous reduction crystallite sizes. The mean crystallite size is around 18 nm after 4 h of milling. From 4 to 16 h of milling the mean crystallite size remain constant approximately, at 16 h the value of average crystallite sizes are 16 nm.

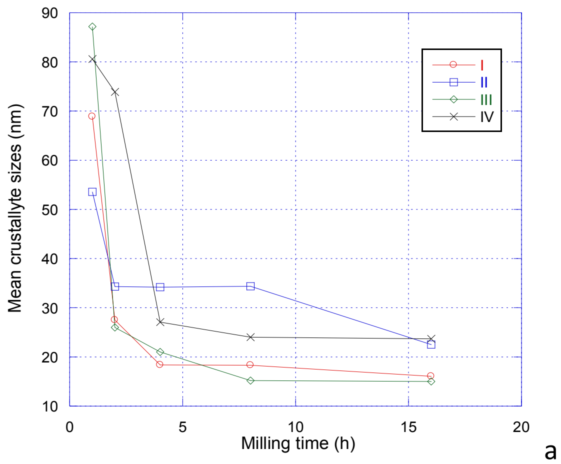


Fig. 6. Crystallite sizes vs. milling time for powder milled.

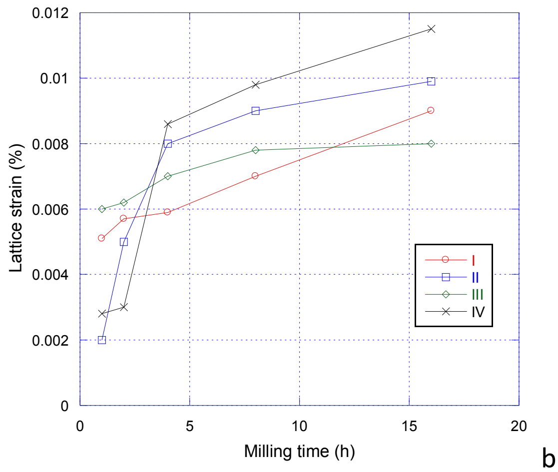


Fig. 7. Lattice strain evolution with milling time.

The mean crystallite sizes are strongly degreasing in first stage of milling (up to 4 h) when milling occurs both in III and IV type of milling conditions. The value for crystallite sizes are 21 nm and 28 nm respectively. In the range of 4 – 16 h of milling the crystallite sizes reduction are insignificant. After 16 h of milling the mean crystallite sizes is about 15 nm if powder was milled in III type milling conditions and 24 nm if Ni_3Fe compound was obtained in IV type of milling conditions. Although, the kinetic energy is higher if milling process occurs in I type conditions than II and in III than IV, the process efficiency is reverse. It can be explained by number of balls which give the number of impacts and their velocity (higher in I than II and about the same in III and IV type of milling conditions).

Also, using Williamson-Hall method was calculated the lattice strains induced in powder by mechanical milling process. The lattice strain variation with increasing of milling time is shown in fig. 7. It can be observed that the lattice strain increases moderately with increasing of milling time in I and III type of milling conditions up to 8 h of milling. In the range 8-16 h of mechanically alloyed powder the lattice strain shows a linear and continues increase if mechanical milling in type I milling conditions occurs. The value of lattice strain at 16 h of milling is about 0.009 %. For powder milled in III type of milling conditions between 8 and 16 the value of lattice strain remain approximately constant at 0.008 %. Initially, the lattice strains of Ni_3Fe powder milled in both II and IV milling conditions increases significantly with increasing of milling time in first 4 hours. The values of lattice strain after 4 h of milling are almost as well as the values for Ni_3Fe powder 16 h milled in type I and III milling conditions. In the range 8 – 16 h of milling the lattice strain continues to grow moderately. At the end of the milling process the values of lattice strain attained 0.01 % if milling process in type II occurs and 0.011 % for II type milling conditions were used.

The powder mixture was analyzed in terms of particle sizes evolution with milling time in all of types of milling conditions. In fig. 8 are shown the particle sizes distribution of Ni_3Fe powder after 16 h of milling.

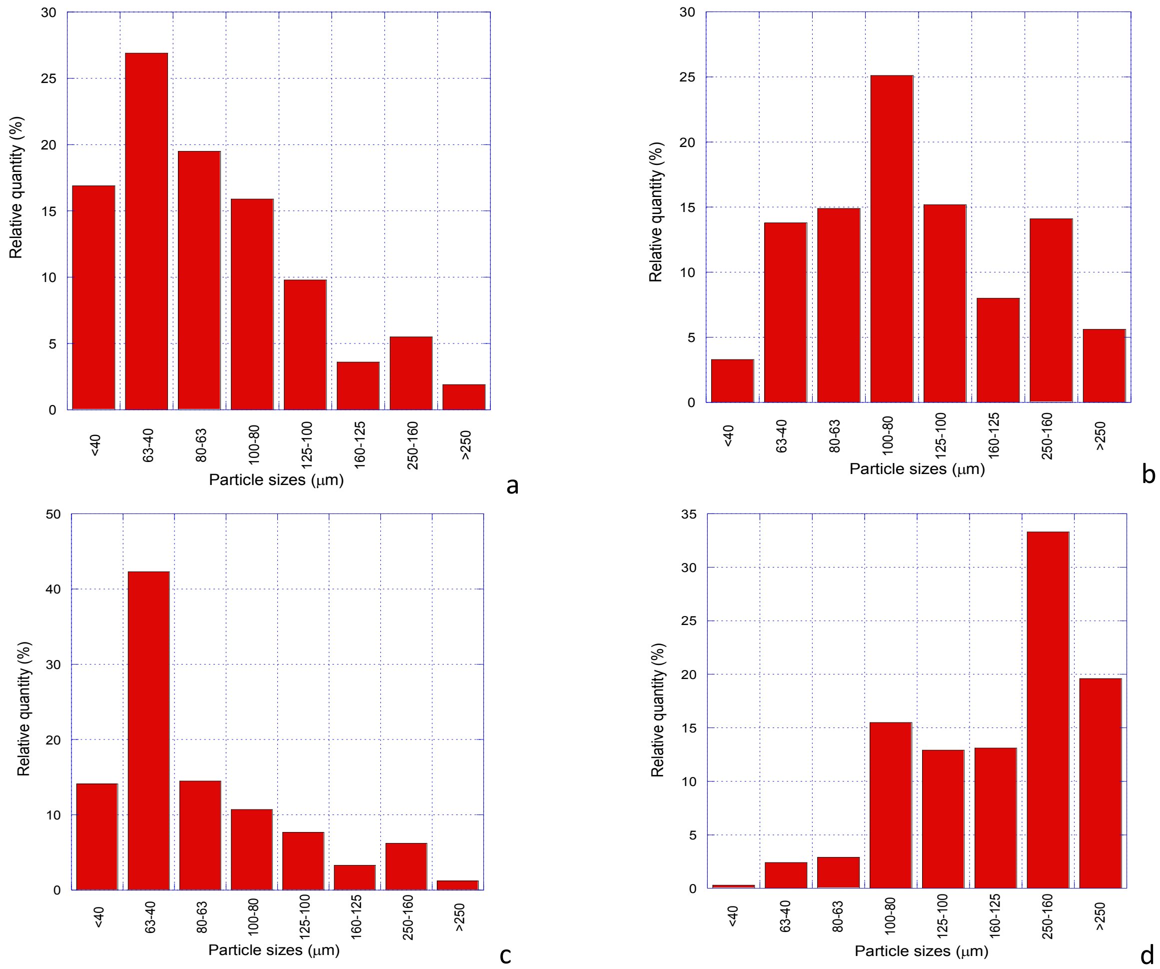
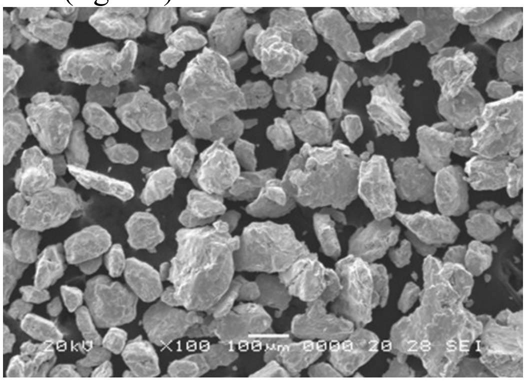


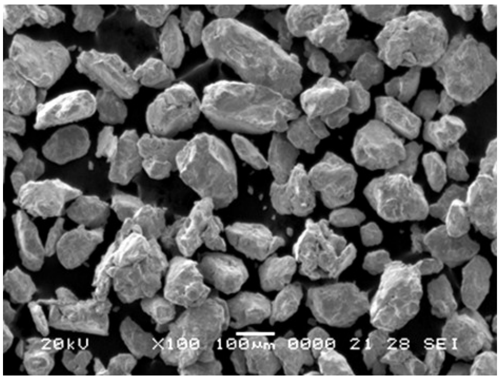
Fig. 8. Particle sizes distribution of Ni_3Fe powder milled 16 h in – conditions I -a), II –b), III – c) and IV – d)

From fig. 8 a, it can be noted that the most particle sizes of Ni_3Fe milled 16 h in I type milling conditions are less than 100 μm (more than 75 %) with a maximum ranging between 40 and 63 μm. If mechanical milling process was perform in II type of milling condition (fig. 8 b), it is observed an approximately uniform distribution of Ni_3Fe particle sizes 16 h milled than in I type milling condition used. The size distribution spectrum is shifted to higher values of particle sizes. Maximum volume of powder is in range 80 – 100 μm (about 25 %). Changing the milling parameters (type III milling condition – fig. 8 c), the Ni_3Fe powder particle sizes are smaller than type I and II milling condition used. After 16 h of milling more than 80 % of powder particles volumes are smaller than 100 μm. Also is noted that about 50 % from volume of powder particle sizes is less than 63 μm.

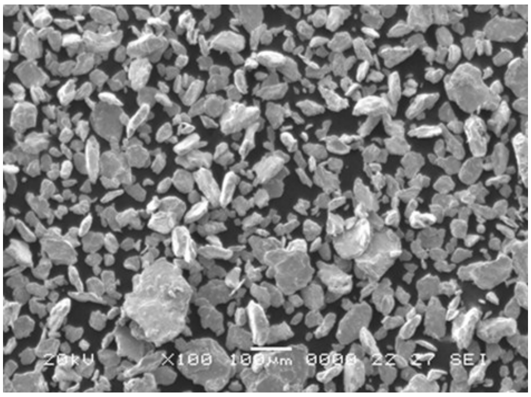
After 16 h of milling in type IV milling conditions the Ni_3Fe powder distribution is shifted to higher value of particle sizes (fig. 8 d). More than 90 % of mass powder has particle sizes higher than 80 μm with a maximum (33 %) in range 160 – 250 μm. The powder sizes analysis of Ni_3Fe powder 16 h milled shows that using type I and III milling condition results smaller particles than type II and IV milling conditions even if the balls energy are higher in last two milling conditions. This can be explained by higher number of ball – ball and ball – inner wall of vials impacts in case to use type I and III milling conditions (higher number of balls with 11 mm diameter). Cold-worker is grater in these types of milling conditions (I and III) due the more plastic deformation process. Because of this, fragmentations are prevalent cold welding. The SEM images of Ni_3Fe powder 16 h milled in all type of milling conditions are shows in fig. 9. After 16 h of milling in I type milling condition the Ni_3Fe powder particles have an irregular shapes with cutting edges who delimited flat surfaces (fig. 9 a).



a



b



c

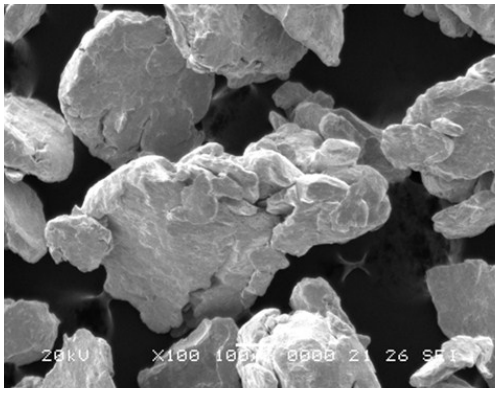

d

Fig. 9. SEM image of Ni_3Fe powder milled 16 h in condition I – a), II- b), III-c) and IV – d)

It can be observed the “sandwich” type structure of Ni_3Fe powder 16 h milled. This is a result of cold welding between flat powder particles (plastically deformed). The mean particle sizes is about 100 μm or less (confirmed by sieving analyze). For Ni_3Fe powder milled 16 h in II type milling conditions the SEM image is shown in fig. 9 b. The powder shows a high degree of homogeneity then in first case. The particle size is around 100 μm. The Ni_3Fe powder 16 h milled in III type of milling conditions have the same irregular shapes and their dimension much smaller than powder milled in type I and II conditions (fig. 9 c). At 16 h of milling in type IV conditions, the Ni_3Fe powder has the larger sizes of all types of milling conditions used (fig. 9 d). This is confirmed by sieved analyzed. It can be observed powder with particle sizes even 200 – 300 μm. In the center of image it is shown a particle with fragmentation crack due mechanical milling processing.

Conclusions

The Ni_3Fe powder was obtained in all four type of milling conditions. In type I milling conditions the Ni_3Fe powder was formed after 2 hours of milling. If type II conditions using, the Ni_3Fe intermetallic compound was formed after 8 hours and after 16 hours of milling when type III and IV milling condition was used. The crystallite sizes are smaller for Ni_3Fe milled in I and III milling conditions (15 nm) than using type II and IV conditions to milling, about 23 nm. SEM and particle sizes analysis investigations showed that the particles sizes are smaller when Ni_3Fe powder was processing in III type milling conditions and largest in case to milling process in type IV milling conditions occurs. From all analyzes made (XRD, SEM and particle sizes distribution) that one of the most important parameter of mechanical alloying process is frequency of impacts, more important than ball velocity and their kinetic energy.

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