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A R C H I V E S O F M E T A L L U R G Y A N D M A T E R I A L S

Volume 57 2012 Issue 1

DOI: 10.2478/v10172-011-0156-z

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A MODEL FOR ABNORMAL GRAIN GROWTH IN NANO-CRYSTALLINE MATERIALS BASED ON ZENER DRAG FORCE

MODEL NIEPRAWIDŁOWEGO WZROSTU ZIARNA W MATERIAŁACH NANOKRYSTALICZNYCH OPARTY O SIŁĘ PĘDNĄ ZENERA

Abnormal grain growth of a matrix in which normal grain growth has stagnated due to the presence of fine incoherent ceramic particles is studied. A balance between driving and retarding forces is used as the criteria for estimating the steady state. Random and non-random approaches are applied for coarse and nano-grained structure respectively.

Keywords: Grain size, abnormal grain growth, nano-crystalline

Badano nieprawidłowy wzrost ziaren w materiale, w którym prawidłowy wzrost ziaren został zahamowany z powodu obecności drobnych cząsteczek ceramicznych. Równowaga pomiędzy siłami pędną i opóźniającą zostały przyjęte jako kryterium oszacowania stanu równowagi. Zastosowano przypadkowe i nieprzypadkowe podejście odpowiednio do struktury grubo i drobnoziarnistej.

1. Introduction

Nanocrystalline materials are thermodynamically unstable due to the presence of a large fraction of interface boundaries. Accordingly, abnormal grain growth is observed in the case of various nanostructured materials [1-10]. Stabilization of the fine grained structures is of critical importance for retaining their unique properties [11-13]. There are several mechanisms that are responsible for the enhanced thermal stability against grain growth [14, 15]: (a) solute drag, (b) grain boundary segregation, (c) Zener pinning by fine ceramic particles and (d) chemical ordering. In the case of Zener pining, however, abnormal grain growth can be expected when the normal grain growth is prevented by the secondary phase particles. This could be due to: 1) a decrease in the Zener back stress because of a reduction in the number of particles (either by dissolution or Ostwald ripening) [16], 2) anisotropy in grain boundary energy [17] and 3) a broad distribution size of the grains in which normal growth stagnated because of the presence of a low volume fraction of the ceramic particles [18].

The present model concerns the latter case, assuming that grains are spherical and growing in the presence of incoherent stable ceramic particles. The only driving force considered is the reduction of total grain boundary energy. Random and non-random approaches are used for the interaction of particles with boundaries in the case of coarse and nano-grained matrix respectively.

2. Coarse grained materials-Random Approach

First one should define retarding and driving forces applied on a unit surface of grain boundary in a matrix with a uniform distribution of second phase particles. The driving force is arisen from the presence of grain boundaries with surface energy of γ. The number of grains in a unit volume of a material of an average grain size of D_i is obtained by

$$n = \frac{1}{\frac{\pi D_i^3}{6}} \tag{1}$$

Considering the surface of each grain is shared between two grains, the total grain boundary area per unit volume would be

$$S_v \approx 0.5n \times \pi D_i^2 = \frac{3}{D_i} \tag{2}$$

Thus the driving pressure for abnormal growth is

$$P_d = \frac{3\gamma}{D_i} \tag{3}$$

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This is opposed by two forces; drag from the distribution of particles P_p, and effect of the finite size of the abnormally growing grain(s) P_a.

For a volume fraction f_V of randomly distributed spherical particles of radius r, the number of particle per unit volume (N_v) is given by

$$N_V = \frac{3f_V}{4\pi r^3} \tag{4}$$

The number of particles intersecting an unit area of boundary is then

$$N_s = 2rN_V = \frac{3f_V}{2\pi r^2} \tag{5}$$

The drag pressure caused by the randomly distributed particles on unit area of the boundary is given by

$$P_{p(r)} = F \cdot N_S \tag{6}$$

where F is the maximum restraining force of a particle intersecting a grain boundary of specific energy γ [19].

$$F = \pi r\gamma \tag{7}$$

and hence

$$P_{P(r)} = \frac{3f_V\gamma}{2r} \tag{8}$$

The retarding force due to the radius of curvature of the abnormal grain(s) P_a is given by

$$P_a = \frac{2c\gamma}{D_a} \tag{9}$$

where D_a is the radius of curvature of the abnormal grain and c is a constant taken as 2 [20].

The condition under which a very large grain will grow is

$$P_d \geqslant P_a + P_{p(r)} => \frac{3\gamma}{D_i} \geqslant \frac{2c\gamma}{D_a} + \frac{3f_v\gamma}{2r} \tag{10}$$

The above analysis can be modified to take into account **n** abnormally growing grains which each one reduces the total driving force for growth, thus

$$\frac{3\left(1 - \frac{n\pi D_a^3}{6}\right)}{D_i} - \frac{3f_V}{2r} - \frac{2C}{D_a} \geqslant 0 \tag{11}$$

In the case of a matrix in which the normal grain growth stagnates due to the particle pinning, the limited grain size is obtained via an equilibrium between P_p and driving force for normal growth, thus

$$\frac{3f_v\gamma}{2r} = \frac{2a\gamma}{D_i} => D_i = \frac{4ar}{3f_v} \tag{12}$$

where $\alpha = 0.375$ is a geometrical constant [20].

Inserting D_i in Eq. 11 one finds

$$0.5n\pi D_a^4 + (2\alpha - 3)\, D_a + \frac{16\alpha r}{3f_v} \geqslant 0 \tag{13}$$

This equation shows that in a given r and f_v, abnormal growth would take place only if the value of D_a results in positive values of Eq. 13. There are four answers to Eq. 13, two imaginary and two real answers among those the latter can be considered as the limits of the start and stagnation of the abnormal grain growth(s).

Using a defect model, Hillert found that initiation of abnormal grain growth vastly depends on the distribution of grain size in a matrix, in a way that abnormal growth seems unlikely in a matrix consisted of grains of the same size [16]. Indeed, the presence of one or more grains larger than a critical size (D_{ac}) is the essential criteria for abnormal growth. The first answer of Eq. 13 yields the critical grain size under which abnormal growth of a grain is impossible.

Eq. 13 was solved by MATLAB software for $10<r<90$nm and $0.001<f_v<0.22$. $X = D_{ac}/D_i$ can be used as a criteria for estimating the condition under which abnormal growth is initiated. Since the presence of a grain much larger than the average grain size is unlikely, the larger X, the less the probability of abnormal growth is expected. The results of solution of Eq. 13 show that X=1.78 and it is independent of r and f_v values. Moreover at the initial stages, since the number of abnormally growing grains has no effect on both the retarding and driving forces, the value of X is independent of n.

To the best of our knowledge, this is an unsatisfactory result. The higher volume fraction of particles should reduce the probability of abnormal growth [16, 18, 21, 22]. This unfair result may caused from the assumption that considers that interaction of particles with boundaries is purely random. Several studies, experimentally [23, 24] or theoretically [25-29], have shown that the number of particles correlating with boundaries is much higher than the value estimated using a random approach. The assumption of a random distribution of precipitates at grain boundaries does not seem to be representative of the real materials.

Accordingly the random approach should completely be revised. The non-random approach is exactly the case when nanostructured matrixes are studied.

3. Nano grained materials-Non-Random Approach

From a statistical point of view, interaction of grain boundary-particles in a nanostructured matrix is different from that of a coarse grain one (Fig. 1). Even for a distribution of nano-particles in a nano-grained matrix,

one must consider that approximately all particles are in contact with boundaries (Fig. 1-b). It is reasonable to assume that all particles lie not only on boundaries, but at vertices in the grain structure, because in these positions the particles, by removing the maximum boundary area, minimize the energy of system.

This represents a situation in which all particles are on boundary corners but not all the boundary corners are occupied by particles, and thus the growth will actually continue until all grain corners are pinned (Fig. 1-c).

When studying grain growth in a dispersion of Fe_3C particles in Fe matrix, Helman and Hillert found that most of the particles where situated in the grain corners at the inhibited grain growth status [24]. Using the estimation of Hillert [30], there are 24 grain corners in each grain but each corner is shared between 4 grains. There would thus be enough particles to fill all the grain corners in a material if there are on the average 6 particles per grain volume, thus

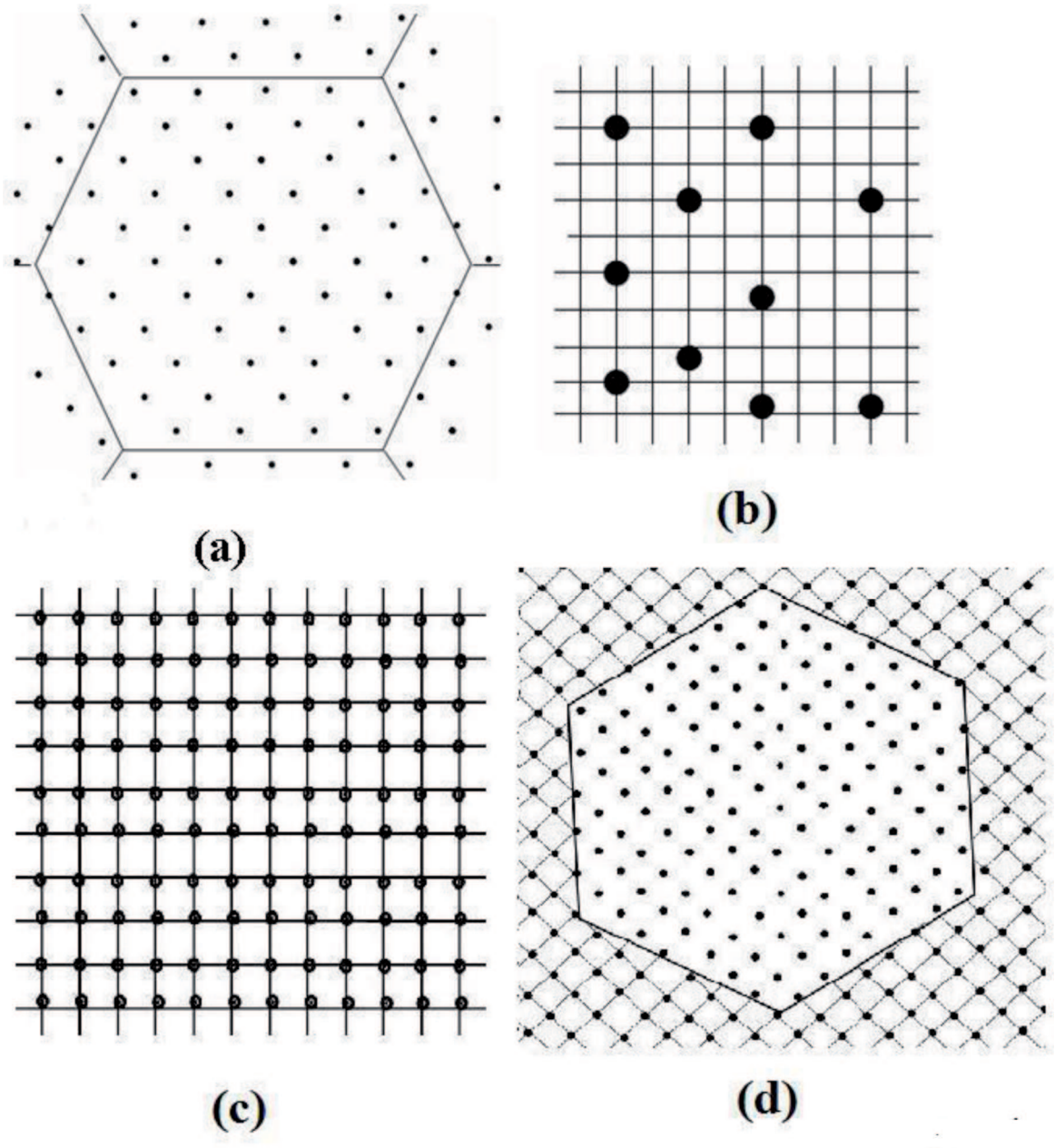

Fig. 1. Correlation of boundary-particles in a) random approach for coarse grained materials, b) non-random approach for nano-crystalline materials, c) stagnant of non-random approach predicted by Hillert [28] and d) abnormal growth in a nano-grained matrix

$$\frac{N_V}{N_D} = 6 \Rightarrow \frac{D}{r} = \frac{\beta}{f_V^{1/3}} \tag{14}$$

where ß = 3.6.

Computer simulation of Hazzledine [26] and Anderson [31] resulted in relationships similar to Eq. 14 only with a slightly different ß value. Considering the interaction mode of particle-boundary in the nanocrystalline materials, it seems reasonable to use Eq. 14 for estimation of limiting grain size in a normal grain growth state.

The retarding force in a non-random distribution of particles is different from that of a random one. Considering that all the particles are in contact with grain boundaries, the number of particles per unit area of surface (n_s) would be obtained by

$$n_s = \frac{N_v}{S_v} = \frac{f_v D}{4\pi r^2} \tag{15}$$

For a nanostructured matrix in which stagnant occurs due to the presence of particles, D is obtained by Eq. 14, thus

$$n_s = \frac{3.6 f_v^{\frac{2}{3}}}{4\pi r^2} \tag{16}$$

The drag pressure caused by the presence of non-randomly distributed particles in a unit area is then

$$P_{p(nr)} = F \times n_s = \frac{0.9 f_v^{\frac{2}{3}}}{r}\gamma \tag{17}$$

Accordingly one can rewrite Eq. 11 in the case of non-random distribution approach.

$$\frac{3\left(1 - \frac{n\pi D_a^3}{6}\right)}{D} - \frac{0.9 f_v^{\frac{2}{3}}}{r} - \frac{2c}{D_a} \geqslant 0 \tag{18}$$

Inserting D in Eq. 18 one finds

$$\left(\frac{n\pi f_v^{\frac{1}{3}}}{7.2r}\right) D_a^4 + \left(\frac{0.9 f_v^{\frac{2}{3}}}{r} - \frac{f_v^{\frac{1}{3}}}{1.2r}\right) D_a + 4 \geqslant 0 \tag{19}$$

Hunderi calculated that interaction of particles with various position of a boundary induces different pressures. While obtaining Eq. 19, however, the drag pressure caused by the presence of particles at grain boundaries, triple lines and quadruples are assumed to be identical.

Results of solution of Eq. 19 give us a critical grain size under which the growth of an abnormal grain is impossible. As mentioned above, X = D_{ac}/D_i can be used as a criteria for estimating the condition in which abnormal growth is initiated. Fig. 2 shows that r has no effect on X, but increase in f_v results in larger X values. This means that as f_v increases the probability of abnormal growth decreases, in a way that for an uniform distribution of grain size with f_v larger than 0.03, occurrence of abnormal growth seems impossible irrespective to the particle size. The effect of f_v on the initiation of abnormal grain growth is an unanimous opinion, but it needs a more precise study in the case of the effect of r.

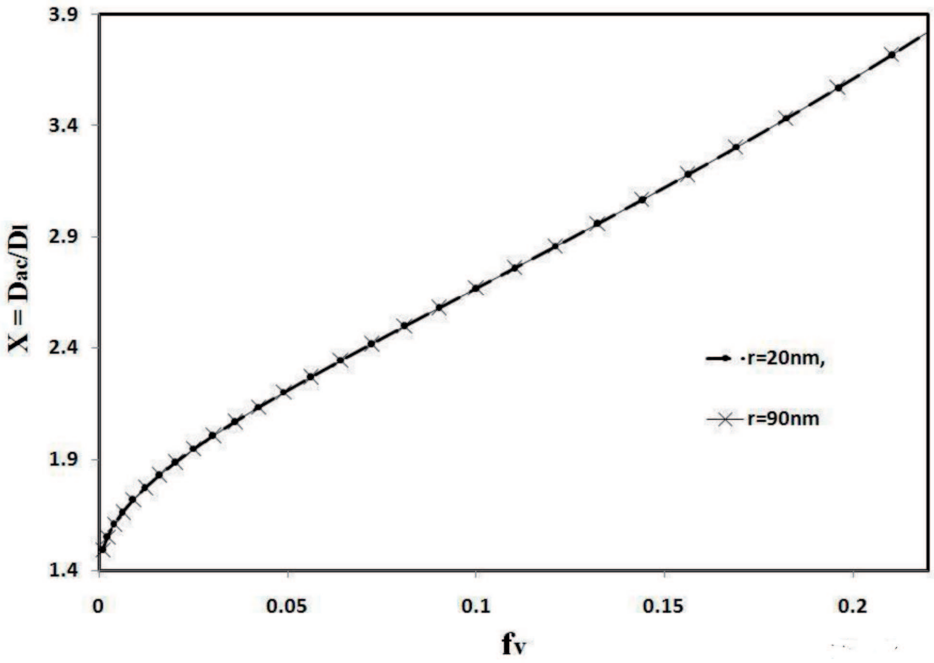

Fig. 2. Effect of volume fraction of nano-particles on X (D_{ac}/D_i) in a non-random approach

Considering special grain boundary locations such as quadruple points, Y. Brechet et al also found that r has no effect on the initiation of abnormal grain growth [32]. However the study of Al_2O_3/SiC nanocomposites from the point of view of abnormal grain growth occurrence shows that a maximum particle size limit exists [33].

After the initiation of abnormal growth, driving force becomes smaller as larger amounts of the volume of matrix are consumed by the abnormally growing grains. It would stagnate when the driving force arisen from the presence of nano-grained matrix equals the retarding force caused by particles in addition to the effect of curvature.

For an extremely large abnormal grain that is growing within a nanostructured matrix, the assumption that all the particles are in contact with boundaries seems unsatisfactory (Fig1-d). Accordingly in Eq. 18, the term that represents the drag pressure due to a non-random correlation of boundary-particles ($\frac{0.9 f_v^{2/3}}{r}$) must be replaced by a random one ($\frac{3 f_v \gamma}{2r}$). Nevertheless the driving force resulted from the presence of grain boundaries is obtained by the term $\left(\frac{3\left(1-\frac{n\pi D_a^3}{6}\right)}{D}\right)$, in which $D = \frac{\beta \times r}{f_v^{\frac{1}{3}}}$. This changes Eq. 19 to

$$\left(\frac{n\pi f_v^{\frac{1}{3}}}{7.2r}\right) D_a^4 + \left(\frac{3f_v}{2r} - \frac{f_v^{\frac{1}{3}}}{1.2r}\right) D_a + 4 \geqslant 0 \qquad (20)$$

Fig. 3 shows that final size of abnormal grains reduces as f_v or n increases but r has no effect on it. As n increases, the driving force would be divided into a larger number of abnormal grains. Meanwhile the retarding forces remain constant, thus the larger n, the smaller the final abnormal grains would be obtained.

Using a defect model, Hillert also found that increase of f_v decreases the probability of abnormal growth. Our model shows that the final size does depend on n and the volume fraction of particles.

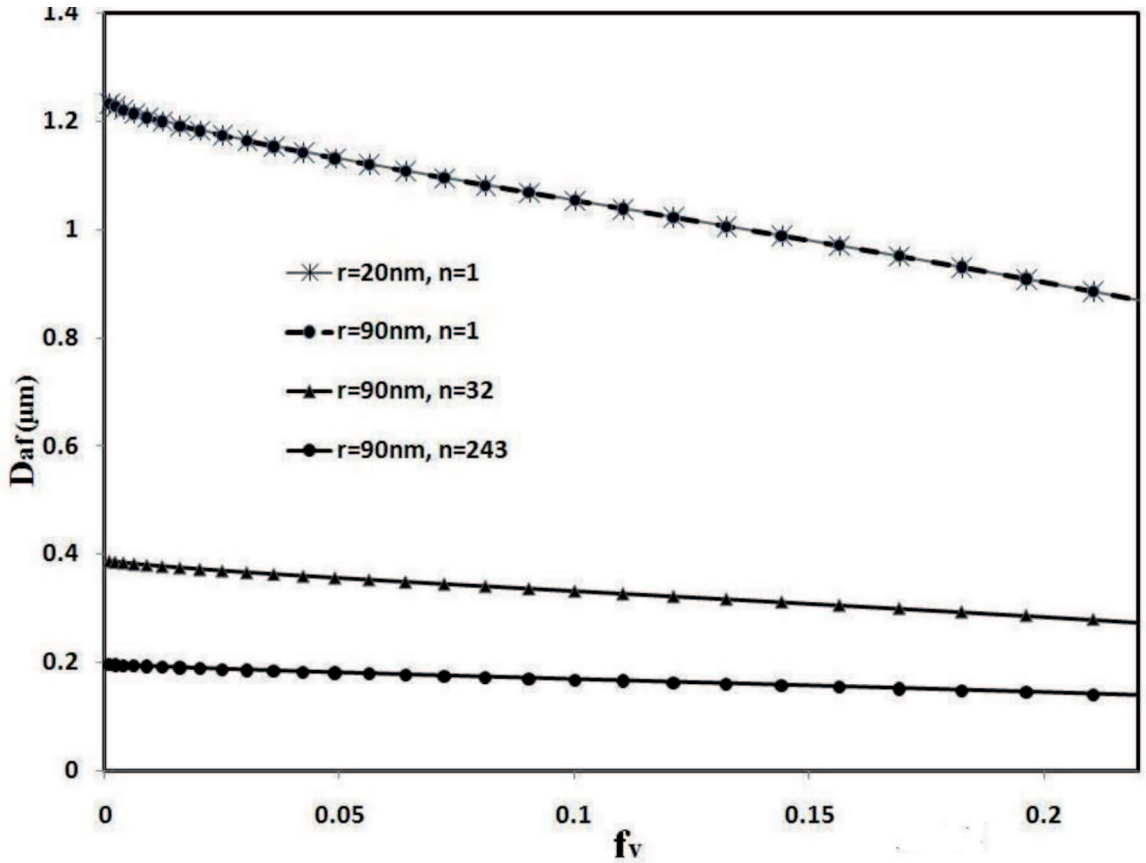

Fig. 3. Effect of volume fraction of nano-particles on the final size of abnormally growing grains

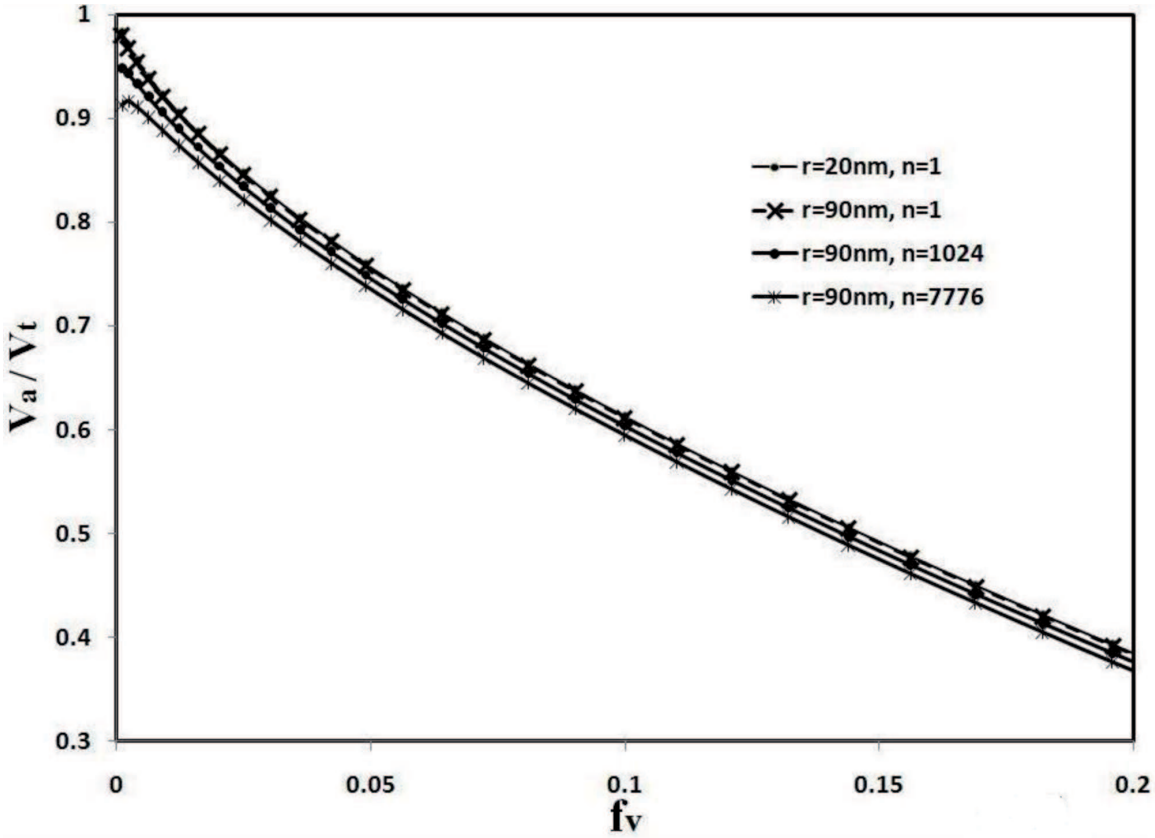

Fig. 4. Effect of volume fraction of nano-particles on the ratio of abnormal grains to the total volume (V_a/V_T)

Moreover the volume percentage of the matrix which is consumed by the abnormal grains rarely reaches 100%. Fig. 4 shows that as f_v increases, the fraction of volume which is absorbed by the abnormal grains reduces. It can be confirmed by the study of A. Simchi et al which shows that as a function of annealing time in a given temperature, volume fraction of abnormal grains in Cu–2.7 vol.% Al_2O_3 reaches a steady state below 100% [2]. G.D. Hibbard et al in a similar way found that in the case of Ni-Co alloys [34], "for a given set of annealing conditions the volume fraction of nanocrystalline matrix consumed by the abnormally growing grains decreases with increasing Co concentration". In this case quantity of impurity drag can be considered the same as the amount of particle drag. They also found that some embedding nano-grains remain in an abnormally grown Ni-Fe structure [5].

4. Conclusion

A model based on non-random distribution of particles was used to investigate the initiation and stagnation of abnormal grain growth within a nanocrystalline matrix. Results show that for a uniform distribution of grain size, abnormal growth can only be initiated at a very low f_v. The final size of abnormal grains and their volume fraction decrease with f_v.

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Received: 20 February 2011.