Nanoparticles for Asphalt Composite Obtained by Milling

GABRIEL VASILIEVICI [1], DORIN BOMBOS[1]*, MIHAELA BOMBOS[2] TRAIAN JUGANARU[1]

[1]Petroleum - Gas University of Ploiesti, 39 Calea Bucuresti,100520, Ploiesti, Romania

[2]National Institute for Research Development for Chemistry and Petrochemistry- ICECHIM-Bucuresti, 202 Splaiul Independetei, 060021, Bucharest, Romania

Multitude of methods for the synthesis of nanoparticles express interest and importance of this field. In the present study, are addressed the aspects of obtaining nanoparticles by milling, in order to be used at nanostructured coatings in asphalt. Materials used for obtaining nanoparticles were powdery volcanic tuff with 0.4-1.6 mm grain and calcium carbonate. Stearic acid, sodium lauryl ether sulphate, a mixture of fatty acid imidazoline, a block copolymer polyethylene oxide- polipropilenoxid -polyethylene oxide and polyethylene glycol were used as additives. For grinding of inorganic particles was used a laboratory planetary mill Fritsch Pulverisette 6, equipped with a stainless steel grinding bowl. Grinding dry powdered volcanic tuff in the presence of additives influences particle size distribution. Wet milling of powdered volcanic tuff in the presence of ROT favours getting a bimodal distribution and decreasing the average particle size.

Keywords: nanoparticles, grinding, additives, particle size distribution

Nanoscience is one of the major areas of current research. Use of material in the form of nanoparticles have advantages due to their size, leading to a series of remarkable physical properties. Multitude of methods for the synthesis of nanoparticles express interest and importance of this field. In the literature there are two options for preparation nanoparticle by grinding: dry grinding or wet grinding [1-5]. Using suitable additives can be reduced energy consumption needed for particle dispersion by this method.

Effectiveness of grinding is limited by dispersion-reagglomeration balance. Optimal grinding specific energy value is obtained experimentally by assessing variation of granulometry depending of ground material specific energy [6]. To obtain fine and ultrafine powders are used ball mills, centrifugal mills and jet mills. In terms of fine grinding, chemical reactions can take place in the solid phase (mechano-chemical reaction) [7,8]. By using suitable additives energy required to disperse shrinks. Thus particle strength is reduced due to adsorption of additives inside cracks and in all material surface submitted to grinding. This is probably the mechanism by which particles are obtained at smaller dimensions by wet milling than by dry grinding in the absence of additives [8]. Nanoparticles with different sizes can be obtained in ball mills in the presence of surfactants in organic solvents, varying grinding conditions [10].

Use mineral nanoparticles in the asphalt is justified by improving properties of bitumen, especially hardness and wear resistance [11]. For example, bentonite was used for reinforcement and modification of melt bitumen under shear forces generated by an ultrasonic field. In bitumen modified with bentonite intercalated nanocomposites are formed, while for most of modified bentonite result exfoliated structures. Nanocomposites with bitumen shows higher softening point, improved rheological properties, increased resistance to cold cracking and improved aging behavior. For nanocomposites with modified bentonite is observed better properties than for nanocomposites with bentonite unmodified because of better compatibility and better dispersion structure of exfoliated clay platelets [12].

Mixture of bitumen modified with SBS / modified montmorillonite composite was prepared by mixing in the melt, resulting intercalated nanocomposites. Nano clay improves the storage stability of polymer modified bitumen significantly without adverse effects on its other properties, confirmed by morphological analysis [13].

Modification of radial copolymer SBS (styrene-butadiene-styrene) with montmorillonite and adding it into bitumen favours obtaining of intercalated and/or exfoliated structures being improved mechanical properties and storage stability of modified bitumen[14].

By adding clays in bitumen modified with copolymers of ethylene-vinyl acetate type is obtained a better dispersion of the polymer. Adding clay results in faster polymer compatibilising with bitumen, influencing the rheological properties of the systems studied [15].

In the present study, are addressed aspects of obtaining nanoparticles by milling, in order to be used at nanostructured coatings in asphalt.

Experimental part

Materials and equipment

Materials used for obtaining nanoparticles were powdery volcanic tuff with 0.4-1.6 mm grain (from Mirsid) and calcium carbonate (Merck). Stearic acid (Merck), sodium lauryl ether sulphate (from Kao Corp. EMAL.), a mixture of fatty acid imidazoline with an amino nitrogen content of 7.8% (ROT from ATICA CHEMICALS), three block copolymer polyethylene oxide- polipropilenoxid - polyethylene oxide (Pluronic P123 from BASF Corp.) and polyethylene glycol (PEG 4000 from Merck) were used as additives.

For grinding of inorganic particles was used a laboratory planetary mill Fritsch Pulverisette 6, equipped with a stainless steel grinding bowl with a capacity of 500 mL and 10 stainless steel balls (ball diameter ∅20 mm, density 7.8 g . cm^{-3} and weight 30 g / ball).

Operating conditions to grinding powders were:

-Material mass ratio grinding / ball 1/2 - 1/3,

-Grinding time 60 min.,

-Speed 500 rpm.

* *Tel.: 0740001304*

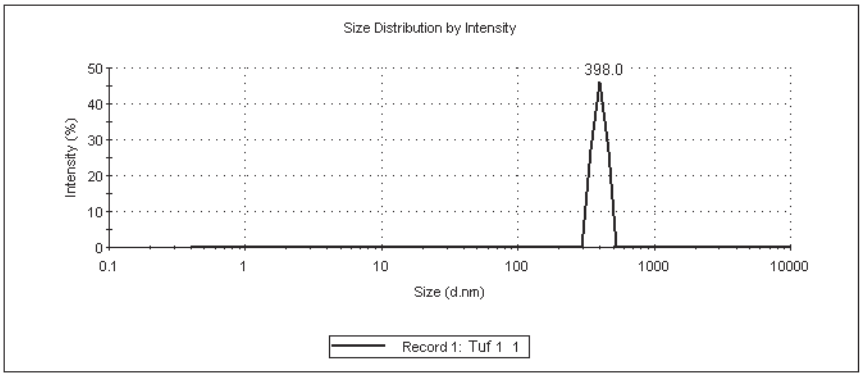

Fig. 1. Particle size distribution of crushed tuff at a mass ratio material grinding / ball of 1/2

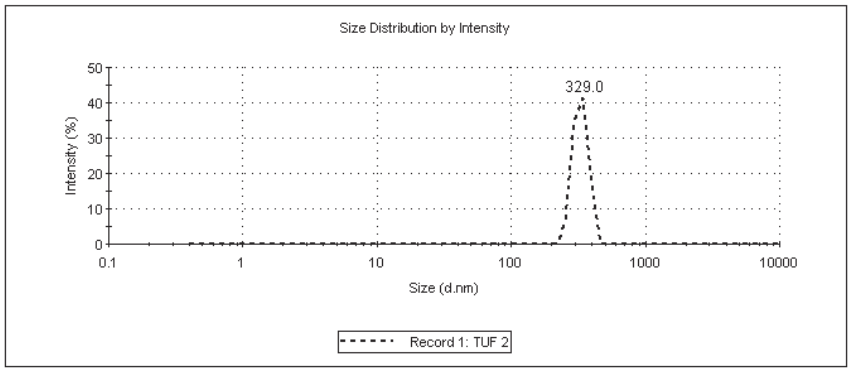

Fig. 2. Particle size distribution of crushed tuff at a mass ratio material grinding / ball of 1/3

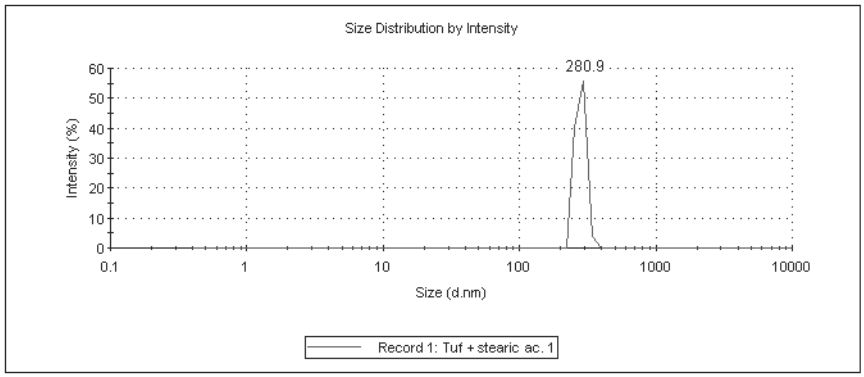

Fig. 3. Particle size distribution of crushed tuff in the presence of stearic acid at a mass ratio material grinding / ball of 1/3

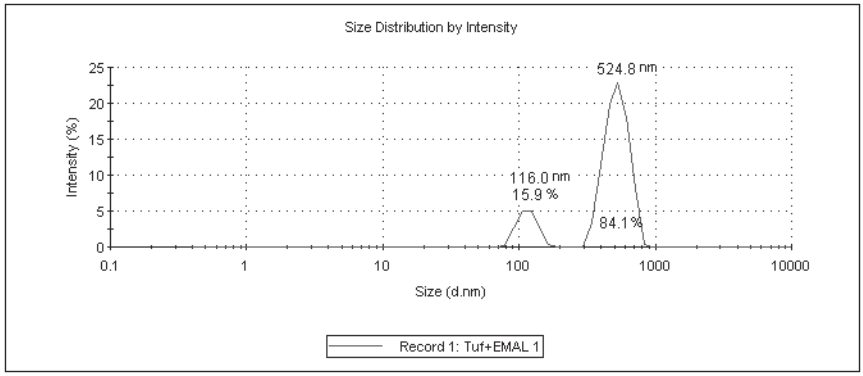

Fig. 4. Particle size distribution of crushed tuff in the presence of EMAL at a mass ratio material grinding / ball of 1/3

Measurements

The particle sizes were determined with a Malvern Zetasizer instrument NanoZS (Red Badge), performing measurements using a process called DLS - Dynamic Light Scattering (also known as PCS - Photon Correlation Spectroscopy). DLS measures brownian motion and correlated with particle size. For size analysis was prepared dispersion of powders in distilled water at concentration 0.1 mg / mL.

Results and discussions

Grinding dry volcanic tuff

The process was conducted at a mass ratio material grinding / ball 1/2 and 1/3. From figure 1 it is observed that for a mass ratio material grinding / ball of 1/2 is obtained a narrow distribution, average particle size of 398 nm and the minimum size of 300 nm (Tuff 1).

Figure 2 shows that for mass ratio material grinding / ball of 1/3 (Tuff 2) it is obtained a broader size distribution than for a mass ratio of 1/2, and particle size diminishes: the size average 329 nm, minimum value 205 nm (Tuff 2). Therefore for further experiments was used crushed material / balls mass ratio of 1/3.

DLS analysis of the powder resulting from the dry grinding of powdered volcanic tuff in the presence of stearic acid at a concentration of 5% and a mass ratio material grinding / ball 1/3, shows an average particle size of 280.9 nm, the minimum size of 210 nm and a narrow size distribution (fig. 3).

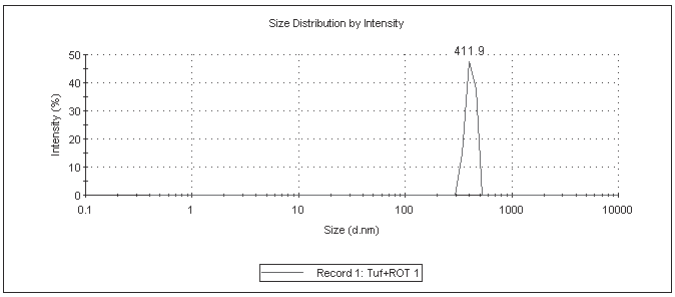

Fig. 5. Particle size distribution of crushed tuff in the presence of ROT at a mass ratio material grinding / ball of 1/3

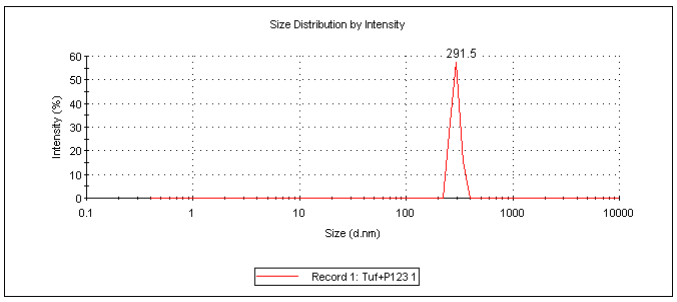

Fig. 6. Particle size distribution of crushed tuff in the presence of Pluronic-P123 to a mass ratio ground material / balls of 1/3

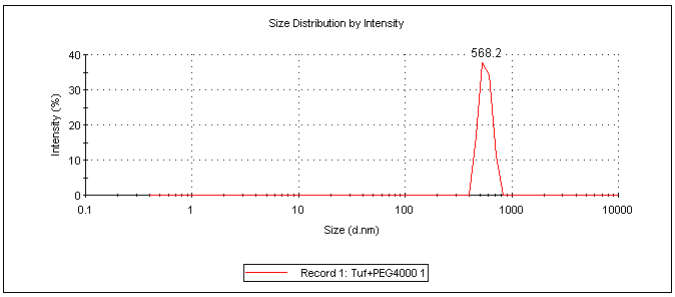

Fig. 7. Particle size distribution of crushed tuff in the presence of PEG 4000 to a mass ratio ground material / balls of 1/3

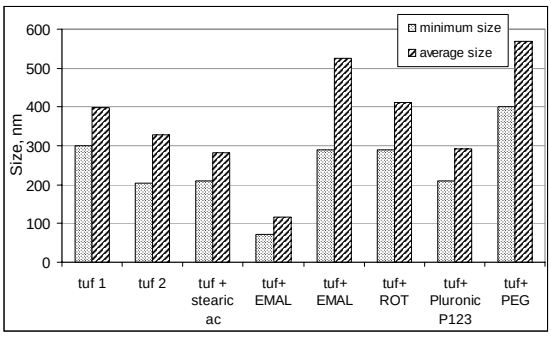

Fig. 8. Comparative distribution of particle size obtained by grinding dry tuff

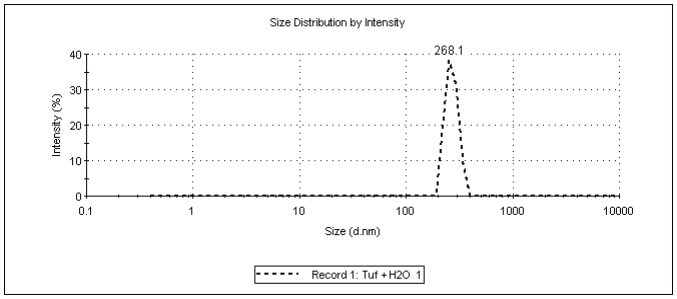

Fig. 9. Particle size distribution of tuff ground in the presence of water

DLS analysis of the powder resulting from grinding dry powdered volcanic tuff in the presence of lauryl ether sodium sulphate at a concentration of 5% and a mass ratio material grinding / ball of 1/3, shows a bimodal size distribution (fig. 4):

- 15.9% with an average size of 116 nm and the minimum size of 76 nm;
- 84.1% with an average size of 524.8 nm and the minimum size of 290 nm.

DLS analysis of the powder resulting from grinding dry powdered volcanic tuff in the presence of fatty acids imidazolines at a concentration of 5% and a mass ratio material grinding / ball 1/3, shows a mean particle size of 411.9 nm, a minimum size of 290 nm and a narrow size distribution (fig. 5).

DLS analysis of the powder resulting from grinding dry powdered volcanic tuff with copolymer polyethyleneoxide - polypropylenoxide - polyethyleneoxide at a concentration of 5% and a mass ratio material grinding / ball of 1/3, shows a mean particle size of 291.5 nm, a minimum size of 210 nm and a narrow size distribution (fig. 6).

DLS analysis of the powder resulting from grinding dry powdered volcanic tuff in the presence of polyethylene glycol at a concentration of 5% and a mass ratio material grinding / ball of 1/3, shows a mean particle size of 568.2 nm, a minimum size of 400 nm and a broad size distribution (fig. 7).

The analysis results from grinding dry powdered tuff shows that the smallest particles are obtained in the presence of sodium lauryl ether sulfate, while the use of high molecular weight compounds (PEG 4000), resulting in a particle size greater than by grinding in the absence of additives (fig. 8).

Wet grinding of volcanic tuff

Wet milling of a volcanic tuff was performed at tuff mass / water ratio of 2/1 and a mass ratio grinding material / balls 1/3. DLS analysis of the dimensions of crushed tuff in the presence of distilled water show that the resulting average particle size was 268.1nm, minimum size of 180nm and the size distribution was relatively wide (fig. 9).

Wet milling of volcanic tuff in the presence of fatty acids imidazoline was achieved at a mass ratio tuff / water: 2/1

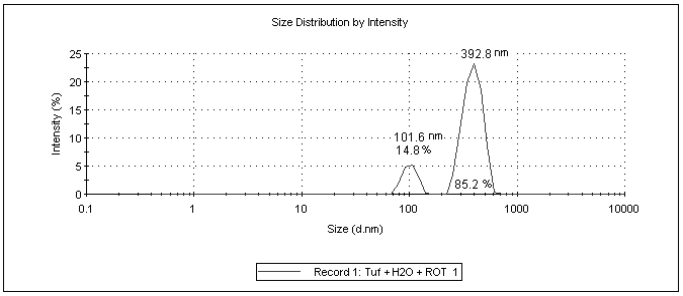

Fig. 10. Particle size distribution of tuff ground by wet grinding in the presence of ROT

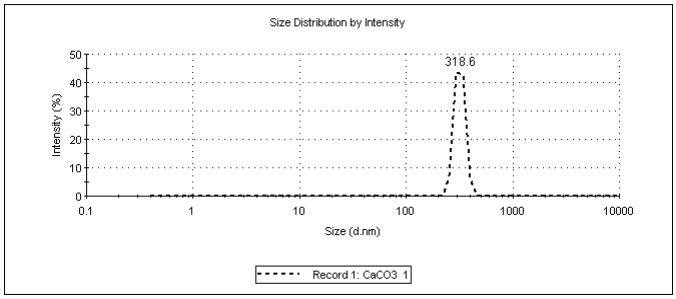

Fig. 11. Particle size distribution of dry ground calcium carbonate

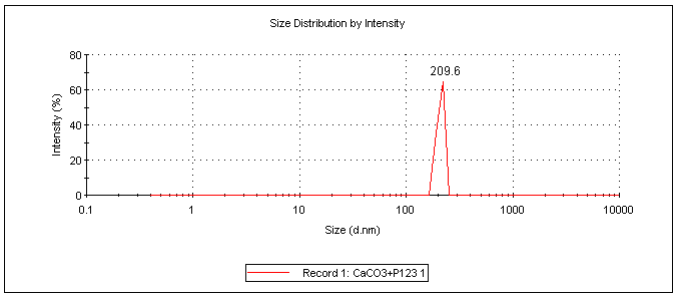

Fig. 12. Particle size distribution of calcium carbonate dry ground in the presence of Pluronic P123

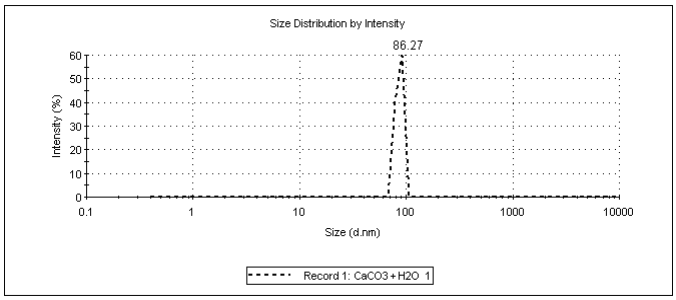

Fig. 13. Particle size distribution of calcium carbonate dry ground in the presence of water

and a mass concentration of the additive of 5% ROT from tuff. From DLS analysis shows a bimodal distribution of particle sizes (fig. 10):

- 14.8% with an average size of 101.6 nm and minimum size of 67 nm;
- 85.1% with an average size of 392.8 nm and minimum size of 210 nm.

Note that the wet milling of tuff in the presence of fatty acid imidazoline decreases the minimum size of nanoparticles to 67 nm and the average size at 101.6nm.

Grinding dry of powdered calcium carbonate

Dry grinding of calcium carbonate was performed at a mass ratio of calcium carbonate / balls: 1/3. DLS analysis of the dimensions shown an average particle size of 318.6 nm, the minimum size of 200 nm and a relatively narrow size distribution (fig. 11).

Grinding dry of powdered calcium carbonate at a mass ratio $CaCO_3$/ball of 1/3 was performed in the presence of additive type Pluronic P123 at a mass concentration of 5% on calcium carbonate. The DLS analysis shows an average particle size of 209.6 nm, minimum size of 150nm and a narrow size distribution (fig. 12).

Wet grinding of calcium carbonate

Wet grinding of powdery calcium carbonate at a mass ratio $CaCO_3$/ball of 1/3 was made with distilled water at a mass ratio $CaCO_3/H_2O$ of 1/1. The DLS analysis shows that average particle size is 86.27 nm, minimum size is 65 nm and the size distribution is narrow (fig. 13).

Wet grinding of powdery calcium at a mass ratio carbonate $CaCO3_3$/ball of 1/3, was performed in the presence of additive type EMAL to a mass concentration of 5% on calcium carbonate. On disposal of mill product was observed foaming powerful of mass processed. The DLS analysis shows that average particle size is 127.8 nm, minimum size is 103 nm and size distribution is narrow (fig. 14).

Wet grinding of powdery calcium carbonate at a mass ratio $CaCO_3$/ball of 1/3, was performed in the presence of additive type Pluronic P123 at a mass concentration of 5% compared to calcium carbonate. On disposal of product from mill distinguish strong foaming. The DLS analysis shows that average particle size is 340.7nm, minimum size is 240nm and size distribution is relatively narrow (fig. 15).

The analysis results in dry and wet grinding of powdered calcium carbonate shows that the best results are obtained

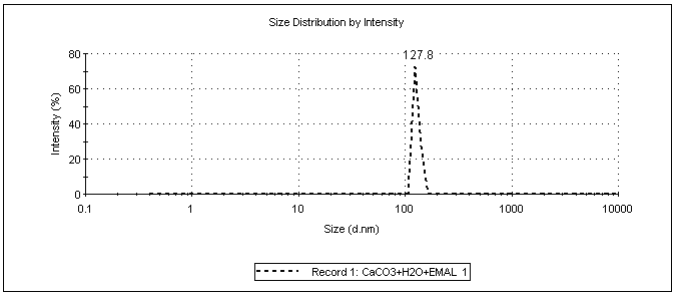

Fig. 14. Particle size distribution of calcium carbonate wet ground in the presence of EMAL

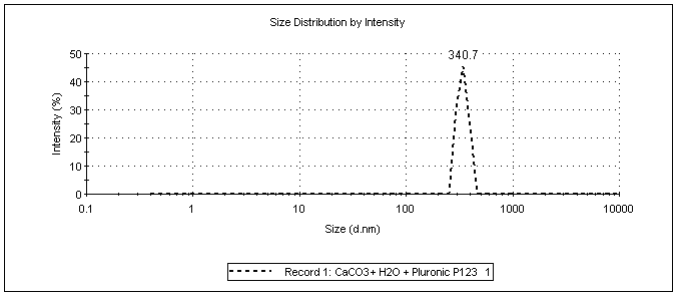

Fig. 15. Particle size distribution of calcium carbonate wet ground in the presence of Pluronic P123

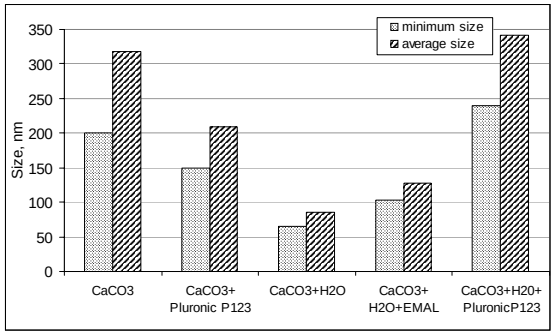

Fig. 16. Comparative particle size distribution of calcium carbonate obtained by grinding

from wet grinding without additives, when nanoparticles with average size of 86.27 nm and narrow size distribution are obtained (fig. 16).

Conclusions

Grinding dry powdered volcanic tuff in the presence of additives influences particle size distribution. Such stearic acid and Pluronic P123 decreases average particle size, EMAL favours getting a bimodal particle size distribution, ROT narrows particle size distribution and increases their average size and PEG 4000 affects negatively both the average particle size and their size distribution.

Wet milling of powdered volcanic tuff in the presence of ROT favors getting a bimodal distribution and decreasing the average particle size.

The dry grinding of calcium carbonate in the presence of additive type Pluronic P123 decreases average particle size and wet milling favours obtaining of a average size less than 90 nm; EMAL present during wet grinding favours narrow particle size distribution but also cause increasing their average size.

Acknoledgements: Authors recognise financial support from the European Social Fund through POSDRU/89/1.5/S/54785 project: "Postdoctoral Program for Advanced Research in the field of nanomaterials

References

1. DERLE, D., PATEL, J., YEOLE, D., PATEL, A., PINGLE, A., Int. J. Curr. Pharm. Res., **2**, no. 1, 2010, p.10
2. PATEL, J. K., PATEL, D. J., PANDYA, V. M., Int. J. Pharm. Sci. Nanotechnol.
Int. J. Pharm. Sci. Nanotechnol., **1**, no. 3, 2008, p.215
3. BHAKAY, A., MERWADE,M., BILGILI, E., DAVE, R.N., Drug. Dev. Ind. Pharm., **37**, no. 8, p.963
4. HOU, T. H., SU, C. H. AND LIU, W. L., Powder Technol., **173**, no.3, 2007, p.153
5. CHARKHI, A., KAZEMIAN, H., KAZEMEINI, M., Powder Technol., **203**, no. 2, 2010, p.389
6.KAWATRA, S.K., Advances in comminution, Society for Mining, Metallurgy and Exploration Inc.(SME), ISBN 978-0-87335-246-8, 2006
7. PINEDA, A., BALU, A. M., CAMPELO, J. M., ROMERO, A. A., CARMONA, D., BALAS, F., SANTAMARIA, J. LUQUE, R., ChemSusChem, **4**, no. 11, 2011, p.1561
8. YADAV, T.P., YADAV, R.M., SINGH, D.P., J. Nanosci. Nanotechnol., **2**, no.3, p.22
9. FUERSTENAU, M.C., Principles of mineral processing, Society for Mining, Metallurgy and Exploration Inc.(SME), ISBN 0-87335-167-3, 2003
10. POUDYAL, N., RONG, C.-B., LIU, J. P., J. Appl. Phys., **109**, 2011, p.07B526-1
11. JAHROMI, S.G., KHODAI, A., Const. Build. Mat., **23**, no.8, 2009, p.2894
12. ZARE-SHAHABADI, A., SHOKUHFAR, A., EBRAHIMI-NEJAD, S., Const. Build. Mat., **24**, no.7, 2010, p.1239
13. Galooyak, S.S., Dabir, B., Nazarbeygi, A.E., Moeini, A., Const. Build. Mat., **24**, no. 3, 2010, p.300
14. POLACCO, G., KRIZ, P., FILIPPI, S., STASTNA, J., BIONDI, D., ZANZOTTO, L., Eur. Polym. J., **44**, 2008, p.3512
15. SURESHKUMAR, M.S., FILIPPI, S., POLACCO, G., KAZATCHKOV, I., STASTNA, J., ZANZOTTO, L., Eur. Polym. J., **46**, 2010, p.621

Manuscript received: 29.03.2012