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CHARACTERIZATION OF THE CHANGES OF THE PARTICLE SIZE DISTRIBUTION OF DIFFERENT AEROSOLS DURING AN ACOUSTIC AGGLOMERATION PROCESS

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ABSTRACT

In this work the acoustic agglomeration process of well characterized SiO_2 aerosols was examined. The aerosols used were composed by different particle sizes: SiO_2 spheres of 0.3 µm, 1 µm and 2.5 µm diameters with different concentrations. An ultrasonic standing wave field with an average sound pressure level of about 155 dB at 21 kHz was generated inside a parallepipedic chamber. The particle size distributions were measured with an aerodynamic particle sizer at the outlet of the chamber in the range 0.5 to 20 µm and the agglomeration effect was characterized. The aerodynamic mass median diameter was increased up to 30% and the particle concentration was reduced up to 90%.

RESUMEN

En este trabajo se examina el proceso de Aglomeración Acústica de Aerosoles de SiO_2 bien caracterizados. Los aerosoles se componían de partículas esféricas de SiO_2 de distintos tamaños: 0.3 µm, 1 µm y 2.5 µm y presentes en diferentes concentraciones. Un campo acústico estacionario con un nivel de presión acústica promedio de 155 dB a 21 kHz se generó en el interior de una cámara paralelepipédica. La distribución de tamaños de partículas fue medida con un Aerodynamic Particle Sizer (APS) a la salida de la cámara en el rango de 0.5 µm a 20 µm caracterizando así la aglomeración. El diámetro mediano aerodinámico en masa se incrementó en un 30% y la concentración numérica se redujo en un 90%.

INTRODUCTION

Power ultrasounds have a very broad field of application that is thoroughly revised in [1]. In material processing power ultrasounds allows wielding of metals or plastics [2] and also 3D printing [3]. In the food industry power ultrasounds have application in drying processes at lower temperatures [4], foam control [5] or emulsification [6]. It has applications also in other fields such as fabrication of nanomaterials or nanostructures [7]. Another application is the Aerosol Acoustic Agglomeration (AAA) [8]. AAA is a process in which an acoustic field induces in the aerosol particles a relative motion that leads to collisions and so to agglomeration. The particle relative movement is caused by different mechanisms that are detailed described in several works [9-12]. These mechanisms can be classified according to its linearity of the wave-particle interaction. Among the linear effects are the entrainment of the particle with the sound field and the linear interaction of the particles with the obstacle scattered sound waves. Among the nonlinear effects are the momentum transference of the sound waves (primary and scattered ones) so-called radiation force and the hydrodynamic effect produced by the wake effect that consists in the flow caused by moving particles and the effect on other nearby particles. The main application of the AAA is the reduction of the very small particles, below 0.3 µm, because the size increase facilitates the aerosol deposition, filtering, or retention [8][13]. From the early works on the applications of the AAA [14] the improvement on the efficiency of the ultrasonic plate-transducers [15] has allowed its application to industrial processes in coal and diesel power plants [16]. This work, founded by the EU-PASSAM project (Grant agreement No. 323217 – Euratom 7FP)[17], extends the evaluation of AAA to the nuclear aerosols produced during severe nuclear accident venting [18][19].

EXPERIMENTAL DESIGN

The experimental design is aimed to replicate as close as possible the expected conditions during hypothetical an accident venting of a nuclear reactor [18]. In which the particles of interest have an aerodynamic diameter around 1 µm as the ones found in the Chernobyl and Fukushima accidents [20][21]. For this purpose an experimental facility has been designed and constructed, which is schematized in figure 1. This installation is composed of two parts, a system of aerosol generation and characterization (PECA) and a Mitigative System - Acoustic Agglomerator MSAA.

The MSAA system consists on a rectangular section chamber that contains the acoustic field and is circulated by the aerosol. Placed inside there are two power ultrasonic transducers, each one have a stepped circular radiator driven at its center by an ultrasonic piezoelectric vibrator and connected to a power electronics generator. The transducers generate a 21 kHz stationary acoustic field inside the chamber with an average sound pressure level of 155 dB. This system was placed inside the PECA. The PECA is a configurable installation that makes air circulate controlling the flow and monitoring the temperature, humidity and pressure.

PECA also generates the aerosol using a brush system PALAS RGB 100-ph able to achieve a maximum aerosol concentration inside the MSAA chamber of 200 mg/m^3. For the characterization of the aerosols several instrument have been used at the inlet and the outlet of the MSAA. At the inlet, just after the aerosol generator, is installed an Electronic Low Pressure Impactor (ELPI) of DEKATI and a probe to an absolute membrane filter. At the outlet an Aerodynamic Particle Size (APS) of TSI and the same kind of membrane filter as the inlet have been switched on the same outlet probe. Again the disposition can be seen in figure 1. The aerosols did consist on mixtures of SiO_2 particles of different sizes and in one experiment SiO_2 mixed with TiO_2. The particles were mono disperse SiO_2 particles 0.3 µm, 1 µm and 2.5 µm in

diameter and polydisperse 1 µm agglomerated TiO_2 particles with primary particles sized between 0.01 µm and 0.05 µm.

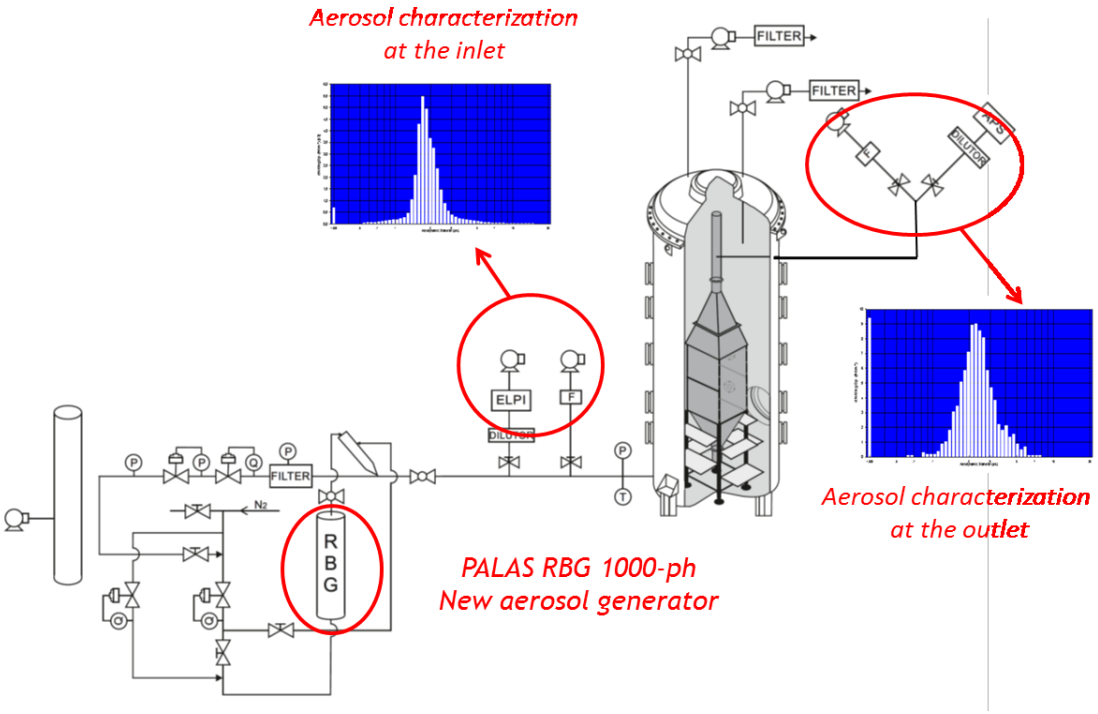

Figure 1. Scheme of the experimental system of PECA-MSAA

MEASUREMENTS

Experimental Matrix

The measurements were carried out during the months January to May of 2015 in the Madrid CIEMAT installations. During this time ten experiments were done with different experimental conditions summarized in Table 1. Each experiment had different aerosol carrier air flow and different composition of the aerosol. In the AAA1 and AAA2 experiments the aerosol was composed by monodisperse aerosols of 1 µm at different flows. Since aerosol generator released the particles at the same mass/time rate an increase in the flow meant a decrease in the aerosol mass concentration. Also higher flow meant less time of the aerosol in the acoustic field. The effect of the time of residence and concentration has been also studied for poly-disperse aerosols with particles of 1 µm and 0.3 µm. The AAA1, AAA3, AAA4 and AAA7 experiments aim to see the effect of the balance of SiO_2 particles in mass on aerosol under the same total concentration and time of residence conditions. The AAA8 and AAA9 experiments study the influence of bigger particles (2.5 µm) in the agglomeration and lastly the AAA10 experiment study the influence of very poly-disperse TiO_2 aerosol.

Measurement Process

The Table 2 shows the schematics of the process that has been followed in each experiment. In the first sub-phase the system is stabilized during approximately 10 minutes while the input aerosol particle size distribution is measured by the ELPI. In the next sub-phase the membrane filters are connected and recollect aerosol particles at the inlet and the outlet for 7 minutes. After

this sub-phase, the filters are removed and the APS start the measurements. The APS keeps measuring till the end of the whole phase. Finally, in the "flush" sub-phase the flow is suddenly increased to 200 kg/h and the particle generation is shutdown. The next phase follows the same steps but the ultrasound system is activated from the start till the start to the flush phase. These two phases were called F1 and F2. Finally the experiment is repeated with the same phases (F1' and F2') but in the APS measurement sub-phase the ultrasound generation is switched on and off in 2 minutes intervals.

Table 1. Experimental variables of the experimental matrix

Name	Flow (kg/h)	Residence time (s)	Input Total Mass Concentration (mg/m^3)	Mass proportion of SiO_2 0.3 µm (%)	Mass proportion of SiO_2 1 µm (%)	Mass proportion of SiO_2 2.5 µm (%)	Mass proportion of TiO_2 (%)
AAA2	100	10	25	0	100	0	0
AAA6	100	10	25	75	25	0	0
AAA5	50	20	50	75	25	0	0
AAA4	12.5	80	200	75	25	0	0
AAA1	12.5	80	200	0	100	0	0
AAA3	12.5	80	200	50	50	0	0
AAA7	12.5	80	200	90	10	0	0
AAA8	12.5	80	200	0	75	25	0
AAA9	12.5	80	200	50	30	20	0
AAA10	12.5	80	200	50	30	0	20

Table 2. Scheme of the measurement Process

Experiment		F1 (Reference Phase)				F2 (Ultrasound phase)			
		Stabilization Sub-phase	Membrane Filter Sub-phase	APS measurement sub-phase	Flush sub-phase	Stabilization Sub-phase	Membrane Filter Sub-phase	APS measurement sub-phase	Flush sub-phase
Duration (minutes)		10	7	5	2	10	7	5	2
Inlet	**Aerosol Generation**								
	ELPI measurement								
	Membrane Filter								
Ultrasounds									
Outlet	**Membrane Filter**								
	APS Measurement								

Instrumentation

One membrane filters have placed at the inlet and other at the in the outlet. These filters collect the aerosol particles that pass through them. With the weight changes in the filters and the PECA data of the flow that passes through the membrane during the 7 minutes the inlet and outlet aerosol concentration has been calculated. Comparing the inlet and outlet concentration we can quantify the retention of the system with and without ultrasounds. The ELPI have measured continuously during the each phase the inlet aerosol particle size distribution and it allows us to measure the aerosol generation and normalize the aerosol output distribution. The Figure 2 illustrates the ELPI measurement of the F2' of the experiment AAA10 with the times of the different sub-phases marked in blocs of different colors. Finally the APS measured the outlet

aerosol size distribution during the “APS measurement” sub-phase and “flush” sub-phase. The measurement of F2’ of experiment AAA10 can be seen in Figure 3 where the ultrasonic system has been switched off and on. The ultrasonic system started on and then was switched off three times producing a rise in the number of particles each time.

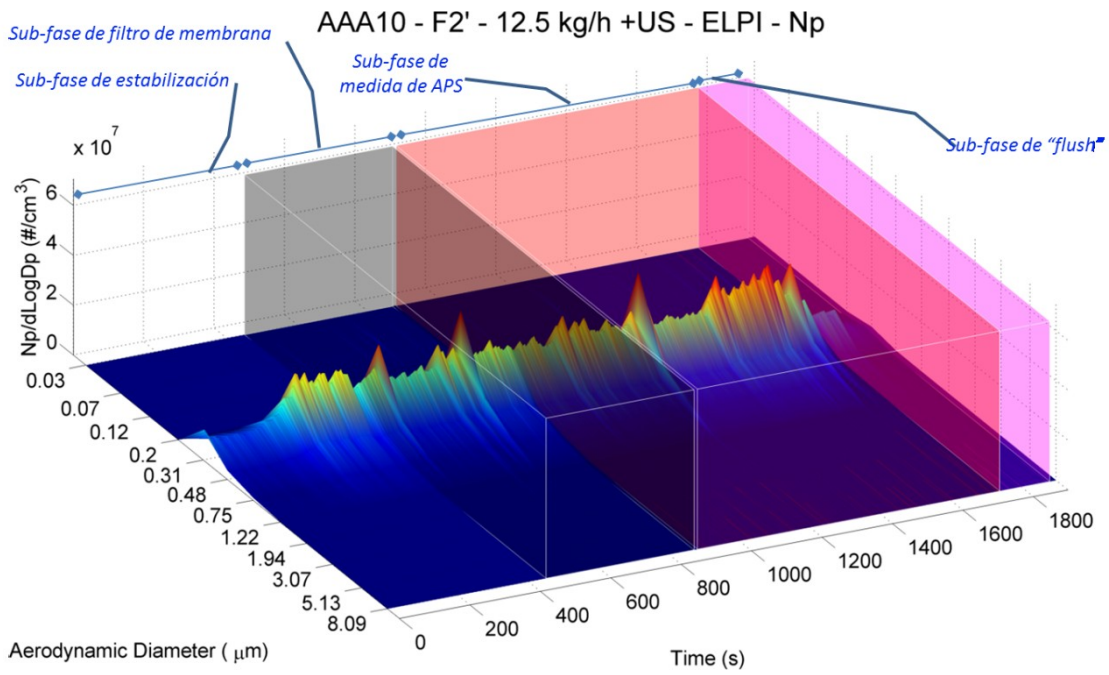

Figure 2. ELPI measurement of F2’ phase of experiment AAA10. The different color blocs correspond to the different sub-phases.

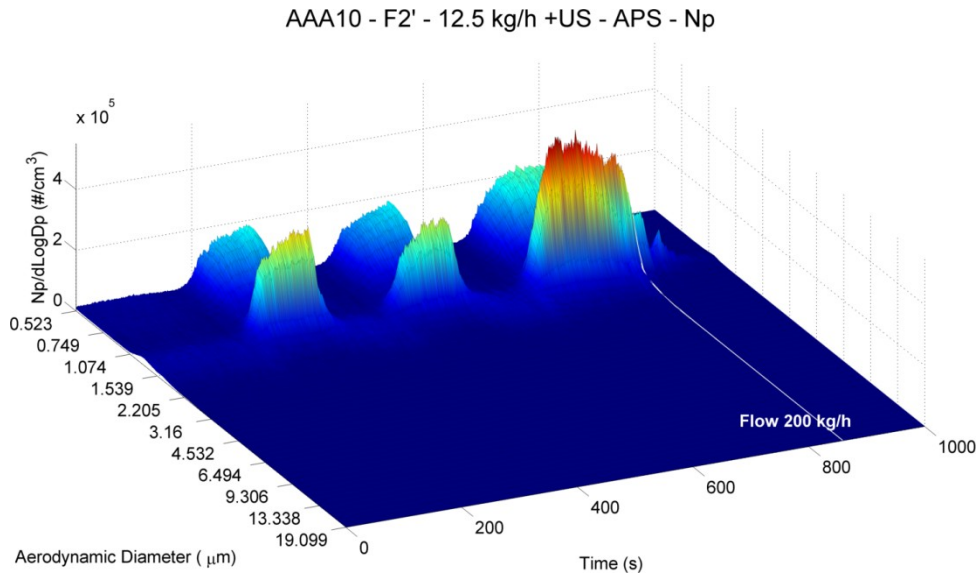

Figure 3. APS measurement of F2’ phase of experiment AAA10. The white line represents the time at in which the sub-phase “flush” starts.

DISCUSION

The reduction of number concentration or reduction of mass concentration has been calculated by several methods. The results of the reduction calculated by the membrane filters are shown

in Table 3. The membrane filters show that for high flows there is no significant increase of retention associated with the application of the ultrasound field and that the retention increases with the reduction of the flow. The effect also increases as the ratio of 1 µm particles increases respect of the 0.3 µm particles going from nearly a 0% of increase retention to a 77% when the proportion gets to 50%. The reduction has been calculated also from the APS measurement data. For this the concentration has been normalized by the ELPI concentration measurement, only the common size range between 0.523 µm and 9.86 µm has been used. The reduction coefficients have been calculated using the equations 1 and 2 where Np_b and M_b are the relative number concentration and mass concentration respectively, the subscripts ELPI and APS refers to the measurements made by those instruments, and RC_n y RC_m are the number and mass reduction coefficients calculated with the average of 3 minutes of the "APS measure" sub-phase.

$$Np_b = \frac{Np_{APS}}{Np_{ELPI}} \qquad M_b = \frac{M_{APS}}{M_{ELPI}} \tag{1}$$

$$CR_n = \frac{media\ (Np_{b\ con\ US})}{media\ \ (Np_{b\ sin\ US})} \qquad CR_m = \frac{media\ (M_{b\ con\ US})}{media\ (M_{b\ sin\ US})} \tag{2}$$

The results are also summarized in Table 3. The effects goes up to 89% for RC_M and up to 91% para RC_{Np} showing an increase on the retention with the increase of the total concentration and time of residence (due to the lower flow). Also there was an increase of the retention with the increase of the poly-dispersion when balancing the mass proportion of the particles of the aerosol in the experiments AAA1, AAA3, AAA4 and AAA7.

Table 3. Mass and number reduction coefficients, increase of retention measured by membrane filters, Median size and GSD changes measured by the APS

	Reduction coeficients		Membrane Filters	Size and GSD Changes					
Name	RC_M (%)	RC_{Np} (%)	Relative Increase of mass retention ratio (%)	Δ Median %	Δ GSD %	Δ Median %	Δ GSD %	Δ Median %	Δ GSD %
AAA1	*60*	*56*	*-67.16*			*-12.2*	*-9.5*		
AAA2	*12*	*15*	*~0*			*-1.8*	*-0.3*		
AAA3	*86*	*91*	*-76.60*	*1.5*	*5.8*	*28.0*	*16.4*		
AAA4	*84*	*87*	*-61.21*	*0.5*	*-0.3*	*34.6*	*12.7*		
AAA5	*18*	*25*	*-30.80*	*8.4*	*6.4*	*3.4*	*4.2*		
AAA6	*26*	*29*	*~0*	*-4.2*	*-3.0*	*-1.0*	*0.6*		
AAA7	*55*	*67*	*~0*	*18.6*	*3.7*	*27.9*	*14.2*		
AAA8	*55*	*45*	*-43.52*			*-4.2*	*-4.2*	*-0.4*	*-67.7*
AAA9	*87*	*83*	*-63.96*	*4.8*	*2.0*	*23.3*	*15.3*	*4.6*	*-87.3*
AAA10	*77*	*90*	*-73.02*	*-100.0*	*-100.0*	*37.0*	*15.3*		

To characterize the measured distributions at the outlet we adjust by minimum squares a sum of lognormal distributions to the 3 minute average APS measured distributions. The Figure 4 shows such adjustments for the experiment AAA4, the crosses are the averaged distribution, the dotted lines each lognormal and the thick line the sum of the lognormal distributions. The median diameter and the geometric standard deviation (GSD) of each particle type were calculated by the lognormal distribution associated to that particle size. The increases of the median diameter and the GSD are calculated by equation 3 in which U^+ and U^- the phases are with and without ultrasounds and X is the magnitude of the increment. The increase of the two couple of phases (F1-F2 and F1'-F2') is averaged presented in Table 3.

$$\Delta X = \frac{X_{U^+} - X_{U^-}}{X_{U^-}} \tag{3}$$

The median diameter and the GSD of the 0.3 µm particles do not show change in size because the agglomeration mainly happens among different size particles and not among particles with the same size but the seen effect is the lowering of the particle concentration. The particles of 1 µm do show an increase up to 24% in median size that represent the agglomeration that is higher when the flow is lower.

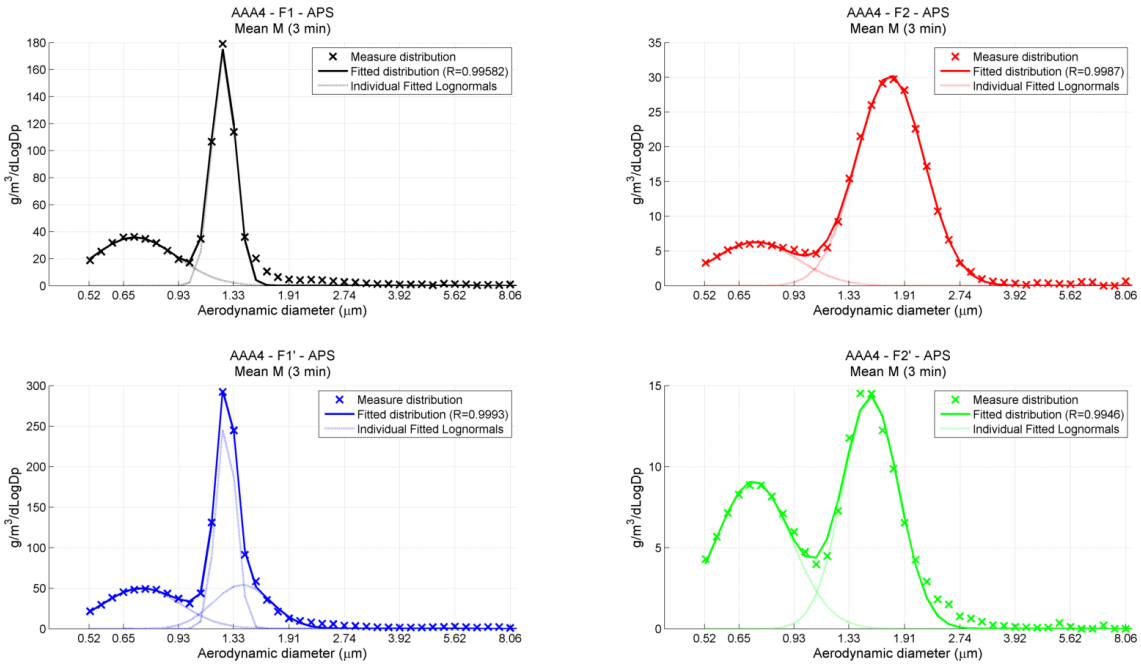

Figure 4. Setting of the distributions for AAA 4 experiments

CONCLUSIÓN

The effects that a standing ultrasonic field at 21 kHz and 155 dB has over several aerosols of around 100 mg/m^3 have been studied. The effect measured by means of an APS is a mass retention increase up to 87% and an increase of the median diameter up to 37%. These effects increase with the polydispersion of the aerosol and with its concentration. In future works this effects will be compared with the effects predicted by the different AAA models.

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REFERENCES

[1] J. A Gallego-Juárez and K. F. Graff, *Power Ultrasonics, Applications of High-Intensity Ultrasound*, ISBN: 978-1-78242-028-6
[2] E. Kicukov, A. Gursel, *Ultrasonic welding of dissimilar materials: A review*, Periodicals of Engineering and Natural Sciences, Vol. 3 No. 1 (2015), pp 28-36, ISSN 2303-4521
[3] J. O. Obielodan, A. Ceylan, L.E. Murr, B. E. Stucker, *Multi-material bonding in ultrasonic consolidation*. Rapid Prototyping Journal, 16(3), 2010, 180-188.

[4] J.V. Santacatalina, M. Contreras, S. Simal, J.A. Cárcel, J.V. Garcia-Perez, *Impact of applied ultrasonic power on the low temperature drying of apple*, Ultrasonics Sonochemistry, Volume 28, January 2016, Pages 100-109, ISSN 1350-4177.
[5] G. Rodríguez, E. Riera, J. A. Gallego-Juárez, V. M. Acosta, A. Pinto, I. Martínez, A. Blanco, *Experimental study of defoaming by air-borne power ultrasonic technology*, Physics Procedia, Volume 3, Issue 1, 1 January 2010, Pages 135-139
[6] M. T. Hamed Mosavian, A. Hassani, *Making Oil-in-Water Emulsions by Ultrasound and Stability Evaluation Using Taguchi Method*, Journal of Dispersion Science and Technology Volume 31, Issue 3, 2010, Pages 293-298
[7] D. G. Shchukin, D. Radziuk, and H. Möhwald, *Ultrasonic Fabrication of Metallic Nanomaterials and Nanoalloys*, Annual Review of Materials Research, Vol. 40: 345-362
[8] D. Zhou, Z. Luo, M. Fang, H. Xu, J. Jiang, Y. Ning, Z. Shi, *Preliminary Experimental Study of Acoustic Agglomeration of Coal-fired Fine Particles*, Procedia Engineering, Volume 102, 2015, Pages 1261-1270
[9] E.P. Mednikov, *Acoustic coagulation and precipitation of aerosols*, New York, Consultants Bureau, 1965
[10] N.L. Shirokova, *Aerosol coagulation*, in Rozenberg LD, Volume 2, Physical Principles of Ultrasonic Technology, New York, Plenum Press, 1973, 475-539
[11] S. Temkin, *Gas dynamic agglomeration of aerosols. I. Acoustic waves*, Phys Fluids, 6, 1994 2294-2303
[12] D.T. Shaw, *Acoustic agglomeration of aerosols, Recent developments in aerosol science*, New York, John Wiley and Sons, 1978, 279-319
[13] E. Riera, J.A. Gallego-Juárez, L. Herranz, G. Rodríguez, V. M. Acosta, A. Blanco, A. Pinto, I. Martínez, R. R. Andrés, *Aplicación de los transductores ultrasónicos de placa en procesos de aglomeración de partículas en el marco del proyecto europeo PASSAM* Conferencia invitada. Tecniacústica, 29-31 Oct. 2014, Murcia. Publicación oficial del Congreso, 1396-1405
[14] H. W. Denser, E. Neumann, *Industrial sonic agglomeration and collection systems*. Industrial & Engineering Chemistry, 41(11), 1949, 2439-2442.
[15] J.A. Gallego-Juarez, G. Rodriguez-Corral, L. Gaete-Garreton, *An ultrasonic transducer for high power applications in gases*, Ultrasonics, Volume 16, Issue 6, 1978, Pages 267-271, ISSN 0041-624X
[16] J. A. Gallego-Juárez, E. Riera-Franco De Sarabia, G. RodrÍguez-Corral, T. L. Hoffmann, J. C. Gálvez-Moraleda, J. J. Rodríguez-Maroto, F. J. Gómez-Moreno, A. Bahillo-Ruiz, M. MartÍn-Espigares, M. Acha, *Application of Acoustic Agglomeration to Reduce Fine Particle Emissions from Coal Combustion Plants* Environmental Science & Technology 1999 33 (21), 3843-3849
[17] T. Albiol, L. Herranz, E. Riera, S. Guieu, T. Lind, G. Manzini,A. Auvinen, N. Losch, New studies on passive and active systems towards enhanced severe accident source term mitigation - The PASSAM Project, Eurosafe, Toward Convergence of Technical Nuclear safety Particles in Europe.
http://193.174.114.174/userfiles/file/Eurosafe2012/Seminar%201/Abstracts/01_10-New%20studies%20PASSAM.pdf
[18] H.-J. Allelein, A. Auvinen, J. Ball, S. Güntay, L. Enrique Herranz, A. Hidaka, A. V. Jones, M. Kissane, D. Powers, G. Weber, *State-Of-The-Art Report On Nuclear Aerosols*, Nuclear Energy Agency Committee On The Safety Of Nuclear Installations, NEA/CSNI/R(2009)5
[19] D. T. Shaw, N. Rajendran, Application of Acoustic Agglomerators for Emergency Use in Liquid-Metal Fast Breeder Reactor Plants, Nuclear Science and Engineering / Volume 70 / Number 2 / May 1979 / Pages 127-134
[20] E. I. Kauppinen, R. E. Hillamo , S. H. Aaltonen and K. S. Sinkko, *Radioactivity Size Distributions of Ambient Aerosols in Helsinki, Finland, during May 1986 after the Chernobyl Accident: Preliminary Report*, Environ.cl. Technol. 1986, 20, 1257-1259
[21] H. Malá, P. Rulík, V. Bečková, J. Mihalík, M. Slezáková, *Particle size distribution of radioactive aerosols after the Fukushima and the Chernobyl accidents*, Journal of Environmental Radioactivity, Volume 126, December 2013, Pages 92-98