sults presented here demonstrate that in certain applications, the performance of Dixon rings clearly exceeds that of any of the other common random column packings. This performance advantage makes Dixon rings the ideal choice of packing for a number of very important applications including scrubbing of carbon dioxide from the air in critical applications such as CO_2 recovery in submersibles, difficult distillations such as that of tritium removal from water, small scale research and development where precise highly accurate results are required and more generally in the improvement of existing processes (for example, retrofitting columns containing inadequate packing with more superior Dixon rings).

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WET PARTICLE CLASSIFICATION BELOW 1 µm – CHALLENGE FOR BASIC RESEARCH AND TECHNICAL DEVELOPMENT

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The conventional processes of wet and dry classification in the particle range of more than 1 µm are sieving and the different variants of stream classification. Approaching particle sizes of 1 µm and below the conventional methods reach their present physical and technical limits. The economic relevance of particle systems below 1 µm, and especially between 1 µm and 0.1 µm, is constantly growing and thus a high demand for improved and new classification processes exists. The grade efficiency, selectivity and cost effectiveness of the methods used for such applications are unsatisfactory or don't exist. In this paper the physical background of classification processes and the problems of fine particle classification are described and discussed. The different physical principles and phenomena are analysed with regard to their potential for technical classification systems in the sub-micron range. Examples are given for promising techniques together with ideas for new approaches.

INTRODUCTION

The splitting of particle distributions into fractions of different particle size is a standard process in various technical applications. These techniques are until now mainly related to particle sizes of more than 1 µm. They are based primarily on the principles of sieving and flow separation by counterflow or crossflow processes in the earth's or a centrifugal field as shown schematically in Figure 1.

Being influenced by competing forces, particles of different size are first transported to different locations in the process area and then collected and discharged

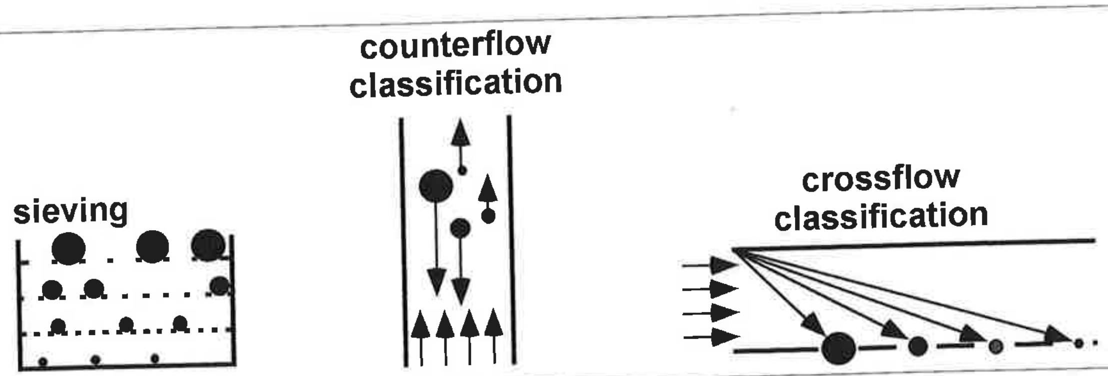


Figure 1: Sieving and flow separation for particle classification.

there. The particles are dispersed either in liquid or gas. Due to advancing technical progress, the application of dispersed solids in the micron or sub-micron and nanometre range is growing remarkably. This also places the requirement for the necessary classification technology before completely new challenges. Many serious problems can arise if particle size distributions are too broad. Examples of the typical risks of particles in a system which are too large include the decreased bio-availability of pharmaceutical active substances, the quality requirements of parenteral nutrition, the impairment of the surface quality of paint films and coatings, the increased probability of defects during the Chemical Mechanical Planarization (CMC) process in chip manufacturing, reduced switching speed of ink jet printed circuits or nozzle blockage while ink jet printing. Figure 2 shows an example of such a micro tool, that is a ceramic capillary made of Al_2O_3/ZrO_2 with high precision face geometry.

An example of unwanted fine particles would be the less favourable separation of dispersed solid catalysts due to high filtration resistances. The necessity of having a tight size fraction is required if special optical effects are to be realised, and the particles must have diameters within the wavelength spectrum for visible light of 400-800 nm. A further example may be the production of particle size standards in the sub-micron range for calibration purposes.

For the purpose of particle size analysis in the nanoscale range some methods of particle classification like impactors or diffusion batteries still exist for microscopically small amounts of material. Good overviews of the present possibilities are given by Kulrattanarak *et al.*[1] and Katasonova and Fedotev[2]. On the other hand, tasks of classifying within the sub-micron range for technical purposes are solved to date, usually unsatisfactorily, by sedimentation in special centrifuges[3] or one-way depth filters. Numerous tasks in classifying are considered as yet unsolved. The main cause for these difficulties are strongly decreasing mass forces with reducing particle size which contrasts with the substantially increasing surface forces. Hence, attractive interactions between the particles can lead to agglomeration and they can be separated only with difficulty, or not at all, depending on their size. The stabilization of such particle systems is more difficult in a gaseous atmosphere than in a liquid environment. In liquids, adhesive van der Waals forces are significantly reduced in comparison to those in a gas. In aqueous liquids, particles can be stabilized by adjusting the pH value or limitation of the ion concentration and strength. However, this is often impossible due to functional demands, for instance additives, and thwarted by mechanical influences like shear which are generated, for example, by agitators or pumps.

TERM DEFINITION AND SETTING OF TASKS

Particle classification includes sorting, separation and splitting, and is one classical subsection of the unit operation of separation in the field of mechanical process engineering.

Classification means the separation of a particle col-

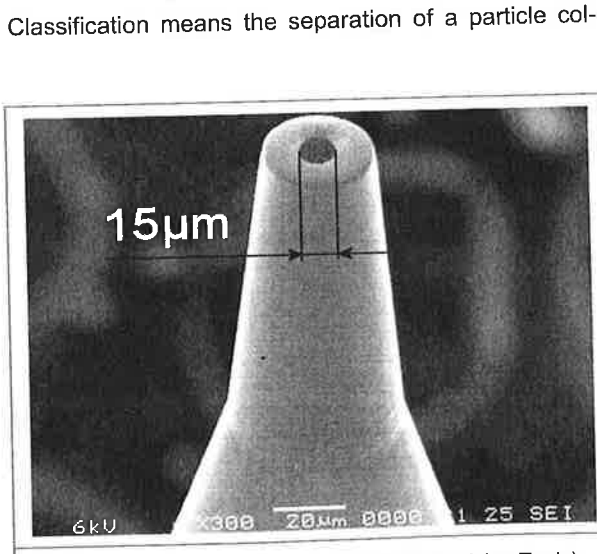


Figure 2: Ceramic capillary (SPT-Small Precision Tools).

lection according to the geometric dispersity attribute size, from a minimal particle size, x_{min}, to a maximal particle size, x_{max}, into a coarse and a fine fraction. The particle size at which the fractions are divided is called the cut size, x_t. Due to the fact that an ideal technical or analytical classification is not possible in principle, some coarse particles will unfortunately get into the fine fraction and *vice versa*. The grade efficiency function, $T(x)$, specifies for each particle size x of the feed material, which percentage gets into the coarse fraction. The grade efficiency thus varies between 0 and 1 as can be seen from Figure 3. In the diagram, x_t is the special cut size, $x_{t,50}$, which is the particle size that exhibits the same frequency in the fine and coarse particle fractions.

The quality of a classification is better when less particles get into the wrong fraction. For an ideal fractionation, $T(x)$ is a function like that shown in Figure 3. The more $T(x)$ deviates from the ideal curve, the poorer is the fractionation quality and this can be characterised by the sharpness of the classification process. One possible way to quantify the sharpness is via the standardised parameter, κ, which is the relation of $x_{t,25}$ to $x_{t,75}$. The numbers 25 and 75 characterise the percentage of the respective particle size from which 25% and 75% can be found in the coarse fraction.

It is possible to define the following:

- Sorting is the separation of a particle collection according to any dispersity attribute other than size like specific weight, colour, shape or others
- Separation is the total separation of one phase from the other, for example like in solid-liquid or solid-gas separation
- Splitting is the separation of a particle collection into parts of the same particle size distribution, as is practised in sample dividing before particle size analysis.

For these relevant classification processes three main tasks can be defined, which are distinguished systematically from each other according to:

- Removal of particles that are too coarse (degritting): For a certain cut size the largest particles of a monomodal material or peaks of a multimodal distribution, which can arise due to agglomeration of primary particles, have to be removed
- Removal of particles that are too small (desliming): The particle size distribution has to be cut off downwards from a certain particle size, x_t
- Narrowing of a particle spectrum into one or more size ranges (fractionation): In the ideal case a monodisperse material is the result, but in reality a narrow size distribution with well defined limits of allowed upper and lower particle size occurs.

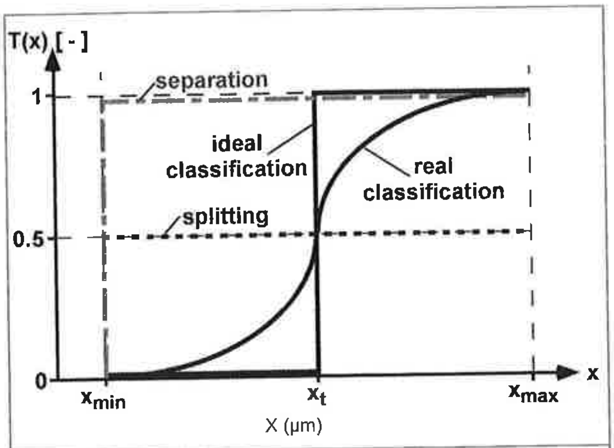


Figure 3: Grade efficiency function.

To be able to classify below a particle size of 1 µm special difficulties have to be overcome in comparison to the coarse particle range, these are:

- Due to the dominating surface forces in the particle size range of interest, the particles to be classified remain stable in the surrounding fluid (here, liquid) and do not agglomerate under the influence of adhesive forces
- The success and quality of a classification process must be measurable quantitatively and precisely
- The selected classification principle and process must be suited to fabrication of technically relevant amounts of product with a sufficiently precise limited particle size distribution and at reasonable cost.

ADHESIVE FORCES AS A LIMITING FACTOR FOR FINE PARTICLE CLASSIFICATION

For particle sizes of less than about 100 µm, adhesive forces like the van der Waals force, gain more influence and can lead to agglomeration, which hinders a classification process or makes it impossible. This is due to the fact that the van der Waals force, F_{vdW}, grows linearly whereas the mass force, F_G, grows with the third power of the particle diameter, x:

$$F_{vdW} \propto \frac{Ax}{a^2} \quad (1)$$

$$F_G \propto x^3 \rho_s g \quad (2)$$

where A is the Hamaker constant, a the distance between the particles, ρ_s the specific solids weight and g the gravitational acceleration. In the case of ideally smooth surfaces the so called 'contact distance' is supposed to be 4Å (0.4 nm). In reality this distance is increased by the roughness of the solid surfaces.

In principle, the van der Waals force between particles is stronger in a gaseous environment than in liquids. This is one reason for the limitation of air classifiers to particle sizes of about 10 µm, whereas wet classification gives more opportunity to get cut sizes below 1 µm. The resulting Hamaker constant, A_{sLs}, depends strongly on the combination of the particles (A_{ss}) and surrounding fluid (A_{LL} in the case of liquid). The resulting value of A_{sLs} greater when A_{LL} is smaller in comparison to A_{ss}:

$$A_{sLs} \approx \left(\sqrt{A_{SS}} - \sqrt{A_{LL}}\right)^2 \qquad (3)$$

When $A_{sLs} = A_{ss}$, there is maximal adhesion in a vacuum.

For fine particle classification special measures for dispersing must be taken to separate particles from each other. These may be mechanical forces generated, for example, by ultrasonic waves or a strong shear flow. Methods to stabilize particles are to mask them or to build up a repulsion potential by charging the particles electrostatically. For masking the surface of the particles has to be covered by a material (A_{mm}), which leads to a remarkably smaller Hamaker constant, A_{mLm}, than the pure solid surface, A_{sLs}. An example is the masking of solids dispersed in water by xylol, like that shown in Figure 4. The generation of the required repulsion potential can be realized by adjustment of the pH value or the ionic strength. Figure 5 shows the qualitative dependency of pH and ionic strength on the repulsive surface potential (zeta-potential) of particles. The isoelectric point marks zero repulsion which means maximal attraction.

Particles dispersed in polar liquids are influenced by attractive (van der Waals) and repulsive (electrostatic (Born)) forces. Around particles, which are carrying an immobilized surface charge (Stern potential), a diffuse double layer of counter ions forms. The DLVO theory describes the resulting force balance like that seen in Figure 6. As can be seen from Figure 6, the resulting 'potential wall' has to be made high enough to hinder the particles from coming close enough to each other to stick together. If particles are agglomerating in the secondary minimum, a mechanical dispersion can be realised relatively easily. If the particles are agglomerating in the primary minimum, a dispersion is very difficult. To overcome the repelling 'barrier', the kinetic energy of the particles, which are moving relative to each other, must be high enough. Table 1 illustrates an approximate estimation of the interaction energy due to different mechanisms depending on the particle diameter.

This data illustrates that for particles of 0.1 µm, shear forces due to stirring are much smaller than pure van der Waals attraction. This means that a mechanical deagglomeration in such a case is nearly impossible. On the other hand, one can see that in the case of enough kinetic energy, the repulsive barrier can be overcome and an agglomeration takes place. As a result depending on the specific conditions a certain amount of kinetic energy can lead to agglomeration or deagglomeration. This kinetic energy can be generated by the shear flow in the feeding zone of solid bowl centrifuges, in the flow channels of dynamic crossflow filters or by the action of stirrers or pumps.

Depending on the locally acting kinetic energy, the repulsive forces have to be made high enough to stabilise the particles. Unfortunately, in several cases such an intervention into the physicochemistry is not allowed due to unwanted interaction with additives that are already present. In addition, one has to take care for stabilization of the particles, not only before the classification to enable this process, but also after the classification to preserve the required product proper-

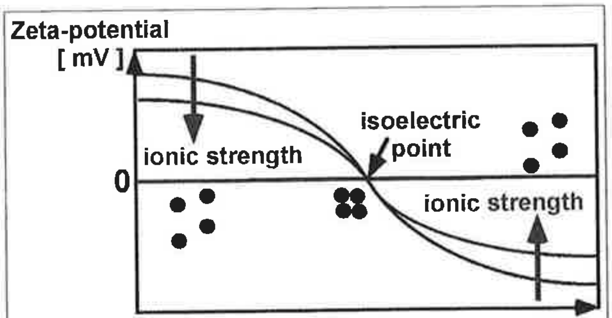


Figure 5: Influence of pH and ionic strength on surface potential.

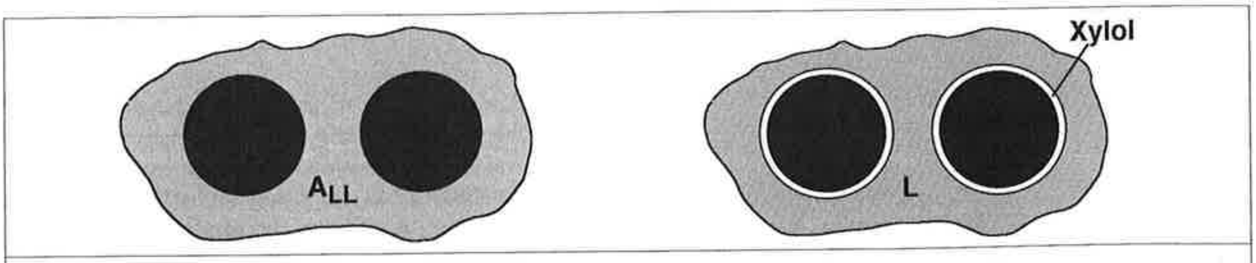


Figure 4: Masking of particles by xylol.

ties. This is a difficulty in the case of drying a product after wet classification or the handling of electrostatically stabilized particles after a dry classification. Last, but not least, an interesting idea especially for particles in the nanometre range may be that the van der Waals force is linearly proportional to the particle diameter, *x*, which means that the attractive force is decreasing with decreasing particle diameter. Thus, the repulsive forces required to hinder agglomeration can be lowered, which means that the stabilization of slurries towards smaller particles in the nanometre range may be easier than in the micron range.

PROCESS CONTROL AND VALIDATION OF CLASSIFICATION RESULTS

A special challenge for the realization of classification processes in the sub-micron range is the process control and validation of classification results. There are some analytical methods available to measure particles in the range between 0.1 µm and 1 µm, but they are normally very sophisticated, costly, time consuming and in some cases the danger of misinterpretation exists due to artefacts which have to be identified and excluded from the results. Laser diffraction methods are approaching their lower limit at about 1 µm. Small angle x-ray scattering is approaching its upper limit at about 0.1 µm. Methods of using ultrasound need sufficient slurry concentration whereas other techniques like dynamic light scattering (photon correlation spectroscopy (PCS)) need high dilution of the sample and time. The change of sample composition by dilution particularly emphasises the danger of changing the properties of the slurry and has to be managed carefully.

For precise measurements analytical disc centrifuges, for example, seem to be well suited to the 0.1 to 1 µm particle range. They operate offline, need low particle concentrations in the sample and the time for one analysis takes only some minutes. Figure 7 shows the general principle of operation. The particle sedimentation in the centrifuge is stabilised by a slight density gradient within the liquid. The particles settle within an optically clear, rotating disc. When particles are approaching the outer edge of the rotating disc they scatter a portion of a light beam that passes through the disc. The change of light intensity is continuously recorded and converted by the operating software into a particle size distribution.

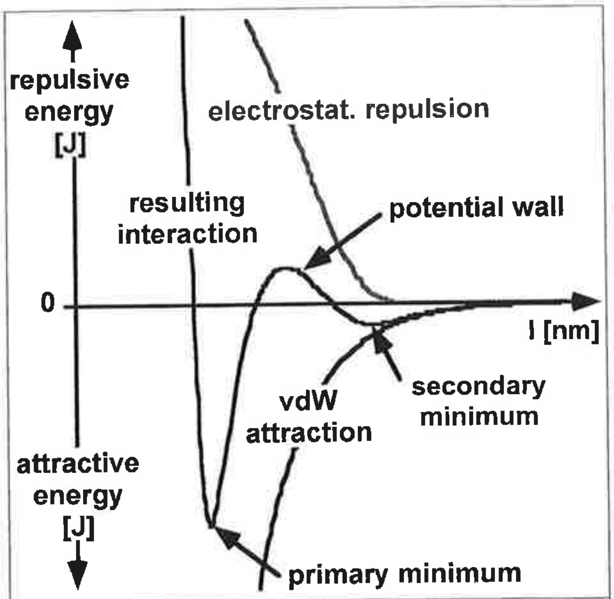


Figure 6: Adhesive and repulsive forces depending on the physicochemistry.

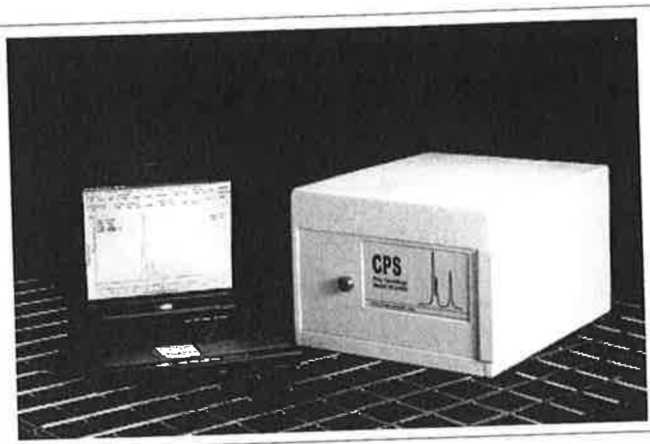
In conclusion, it can be stated that the particle size measurement for process control and product validation of industrial classification processes in the sub-micron range is distinctly possible, but exhibits some open questions that have to be answered. This means that in the future increased efforts are necessary to offer quick online or better inline particle size measurement tools for the sub-micron range.

TECHNICAL CLASSIFICATION OF PARTICLES < 1 µm

Today, two principles are primarily used for technical classification of particles in the range from 0.1 to 1 µm. According to Figure 8, fibrous depth filters represent one of these possibilities, which are especially used for the degritting of CMP slurries. The oversized particles

Interaction energy in kT	Particle size		
	0.1 µm	1 µm	10 µm
van der Waals attraction	≈ 10	≈ 100	≈ 1000
Electrostatic repulsion	$0\text{-}10^2$	$0\text{-}10^3$	$0\text{-}10^4$
Repulsion by Brownian movement	1	1	1
Kinetic energy due to sedimentation	10^{-13}	10^{-6}	10
Kinetic energy due to stirring	≈ 1	≈ 10^3	≈ 10^6

Table 1: Interaction energy between particles of different size[4].



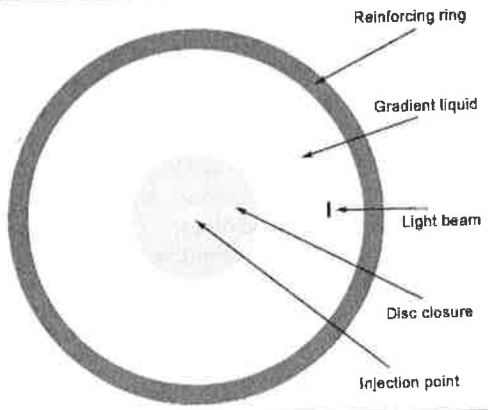
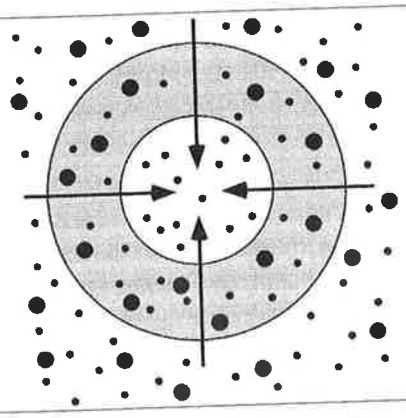


Figure 7: Front view of an analytical disc centrifuge (CPS Instruments).



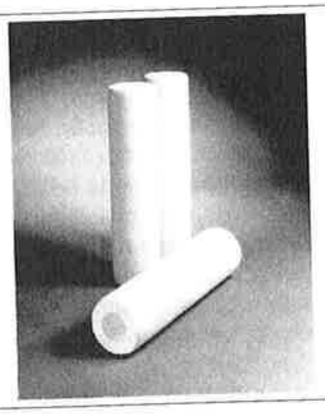


Figure 8: Depth filter to separate oversized particles from CMP slurries (Pall).

are separated safely in the structure of the filter whereby the main part of the slurry can pass through. Unfortunately, a number of small particles are separated too and the filters are not regenerable. The other principle is based on crossflow sedimentation in a centrifugal field. Tubular bowl centrifuges with high accelerations of some 10,000g, as shown schematically in Figure 9, are especially well suited to fractionate submicron particles. Nevertheless, their discontinuous operation and inadequate sharpness of cut lead to the necessity to improve the principle and/or look for new processes.

One possibility of improvement in the future could be the modification of centrifuges like disc stack separators to make the process continuous or to realize a cascade connection of centrifuges to improve the sharpness of cut. Other options could be to think about microporous membranes, or improved methods of counterflow classification in the centrifugal field. It could also be very interesting to check available analytical methods like chromatography to see whether they could be modified to separate technical amounts

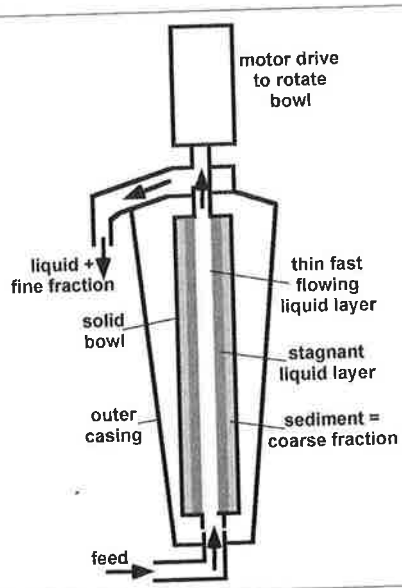


Figure 9: Principle of a tubular bowl centrifuge.

of material. In every case a strong need for progress in this field can be identified.

CONCLUSIONS AND OUTLOOK

The classification of particles greater than 1 μm can, as far as possible, be judged as an industrially solved problem and state of the art. However, classification of particles approaching 1 μm and below is still a challenge and in many aspects is only partially investigated and remains an unsolved task. The physically based problems for fractionation due to very small particle weight and strong particle interaction on the one hand, and more and more applications for such materials on the other hand lead to the need for intensified research and development to offer improved and/or new classification techniques for industry.

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EFFECT OF ACOUSTIC WAVES ON THE PERFORMANCE OF A MULTI-CYCLONE – FILTER SYSTEM

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Many air filtration systems include cyclones for pre-cleaning. The performance of the system changes with time because of filter clogging. The cyclones reduce the concentration of aerosol particles, but their sizes are reduced as well, which leads to deeper particle penetration into the filter. Acoustic waves enhance filtration efficiency and cause sedimentation of the very small particles on the filter surface. They also cause the filter layer to be more porous, which leads to a reduction in pressure drop across the filter. In the present study a cyclone-filter system, similar to that in heavy vehicles, was used to assess the effect of acoustic waves on the pressure drop across the filter. It was shown that a low frequency acoustic field reduces the pressure drop across the filter and increases the flow rate through the system, which leads to a reduction of the time between filter replacements or regeneration, ensuring improved performance.

INTRODUCTION

Filtration efficiency and pressure drop are the most important parameters that characterize air filters. These parameters depend on various features of the filter, such as porosity, size of filter elements, filter thickness etc. Another important parameter is the operating lifetime of a filter. Every filter undergoes clogging with time, the two main types being surface and depth. Depth clogging occurs when particles penetrate deep into the filter structure before capture. Surface clogging appears in high efficiency filters, when most of the particles are captured on the filter surface. The build-up of captured particles on the filter surface is known as the filter cake. The layer grows with time and so does the pressure drop across the filter. When the pressure drop reaches some critical value, the filter becomes clogged and it should be either replaced or cleaned. To increase the lifetime of filters the majority of air filtration systems include cyclones for pre-cleaning. The cyclone reduces the concentration of aerosol particles in the filter inlet, resulting in a thinner filter cake and lower pressure drop across the filter.

The filter loading process and filter cake formation have been considered by various authors. Endo *et al.*[1] proposed a theoretical model of cake formation in terms of bimodal aerosol loading. Schmidt studied the influence of cake compression on filter performance experimentally[2] and theoretically[3]. He considered periodically regenerable cake-forming surface filters and assessed the increase in pressure drop with filtration time as well as the effect of the dust cake structure. His theory was confirmed by quantitative experimental analyses of dust cake structures, including cake porosities, and particle and pore size distributions.

THEORETICAL BACKGROUND

Moldavsky *et al.*[4] studied the influence of acoustic