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Discussion about the thermal rebound of nanoparticles

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ABSTRACT

In the field of aerosol filtration, the theory of thermal rebound of nanoparticles developed by Wang and Kasper [10] has been subject of discussion for more than two decades since it does not fit the experiment. The main purpose of this theory is that the filtration efficiency of fibrous filters could decrease in the region of 10 nm and below because of an excessive impact velocity of particles. However and despite numerous lab experiments leaded by different teams, the phenomenon has never been clearly observed. We demonstrate in this paper why no thermal rebound effect has ever been measured experimentally. According to our approach, the decrease in filtration efficiency, if any, might only happen below 1 nm, even at high temperatures.

1. Introduction

For the last few decades, nanoscience and nanotechnologies have developed in such a way that some experts already speak of “the Industrial Revolution of the 21st Century” (Interagency Working Grouping on Nanoscience, Engineering and Technology [1], Keiper [2], Fundación de la Innovación Bankinter [3]). In parallel of this significant development, toxicologists warn us against the potential adverse effects of nanoparticles on both human health and the environment (Kreyling et al. [4], Biswas and Wu [5], Oberdörster et al. [6], Borm et al. [7]). Since 2 million nanotechnology workers are expected by 2015 (National Science Foundation [8], Roco [9]), this raises the problem of risk management.

For a long time, aerosols below 100 nm had been considered to be captured by Brownian diffusion. However, in 1991, a theory has triggered doubts about the filterability of particles smaller than 10 nm. Wang and Kasper [10] indeed developed a filter efficiency model for nanometer-sized particles which incorporates the effect of particle rebound from the fiber surface. It is interesting to note that such a concept of bounce probability is already well known for large particles [11,12] for which filtration efficiency typically decreases above 5 μm because of too high inertia. It can also be noted that for nanoparticles, Kops et al. [13] had proposed a first expression of this phenomenon in 1986.

Thus, according to the thermal rebound concept, filtration efficiency E could decrease below 10 nm because of a bounce of particles on the surface of filter medium due to a kinetic energy higher than the adhesion one. The problem is then substantial since the performances of protective equipment such as masks or hood systems are concerned. On the experimental point of view, the studies that have been carried out in the size range 1–20 nm since the early eighties give diverging conclusions. Table 1 provides a review of these research works. d_p and d_f are respectively the particle and fiber diameter (m), α is the packing density of the filtering media, U is the filtration velocity (m s^{-1}) and T, the temperature (K). These parameters are given in Table 1 when communicated by their authors.

Nearly no study results in evidence of thermal rebound observations. The few ones which could claim about a particle bounce measurement (Otani et al. [16], Ichitsubo et al. [18]) were shown by Alonso et al. [19] and Heim et al. [22] to be explained by experimental artefacts due to a mobility shift in size-classification devices used. Up to now, only the results of Kim et al. [23], who had no other choice but to work in extreme conditions to select particles as small as 1 nm with their DMA, have never been confirmed or refuted by other authors below 1.3 nm.

After fundamental reminds about the original concept, this paper discusses about the calculations of the thermal rebound effect in order to show that the current inconsistency between the theory and the experiments realized up to now could eventually be justified. The effect of temperature on the particle bounce is also studied in the last part.

2. A sticking problem

In classical filtration theories, the adhesion efficiency ε is assumed to be equal to unity, which means that all particles strik-

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Table 1
Literature experimental studies dealing with nanoparticle filtration and thermal rebound evidence.

Year	Authors	Particles and media type	Operating conditions	Results
1984	Scheibel and Porstendörfer [14]	Silver particles Stainless steel wire screens	$3.5\,\text{nm} < d_p < 130\,\text{nm}$ $d_f = 50\,\mu\text{m}$ $\alpha = 0.2798$ $U = 2.4\,\text{cm s}^{-1}$	E increases when d_p decreases
1990	VanOsdell et al. [15]	Silver particles Fiberglass fibrous filters	$4\,\text{nm} < d_p < 10\,\text{nm}$ $d_f < 1\,\mu\text{m}$ $\alpha \approx 0.07–0.08$ $5\,\text{cm s}^{-1} < U < 20\,\text{cm s}^{-1}$	E increases when d_p decreases
1995	Otani et al. [16]	Silver particles Stainless steel wire screens and tubes	$1\,\text{nm} < d_p < 10\,\text{nm}$ $d_f = 52\,\mu\text{m}$ $\alpha = 0.31$ $1.2\,\text{cm s}^{-1} < U < 3.6\,\text{cm s}^{-1}$	E increases when d_p decreases but penetration in tubes increases below 2 nm
1996	Skaptsov et al. [17]	WO_3 and MoO_3 particles Stainless steel wire screens	$3.1\,\text{nm} < d_p < 15.4\,\text{nm}$ $T = 295$, 316 and 337 K $U = 2.92\,\text{cm s}^{-1}$	E increases when d_p decreases but also when T decreases
1996	Ichitsubo et al. [18]	Silver and NaCl particles Stainless steel wire screens	$1\,\text{nm} < d_p < 7\,\text{nm}$ $d_f = 75\,\mu\text{m}$ $\alpha = 0.289$	E increases when d_p decreases and stagnates below 2 nm
1997	Alonso et al. [19]	Silver and NaCl particles Stainless steel wire screens and tubes	$1\,\text{nm} < d_p < 7\,\text{nm}$ $d_f = 75\,\mu\text{m}$ $\alpha = 0.289$ $6\,\text{cm s}^{-1} < U < 100\,\text{cm s}^{-1}$	E increases when d_p decreases
2004	Balazy et al. [20]	DEHS droplets Fibrous filters	$10\,\text{nm} < d_p < 0.5\,\mu\text{m}$ $U = 7.5\,\text{cm s}^{-1}$	E decreases below 20 nm[a]
2004	VanGulijk and Bal [21]	NaCl and H_2O particles Fiberglass fibrous filters	$7\,\text{nm} < d_p < 25\,\text{nm}$	E depends on the aerosol type
2005	Heim et al. [22]	NaCl particles Nickel wire screen, stainless steel filter and polypropylene wire screen	$2.5\,\text{nm} < d_p < 20\,\text{nm}$ $4.2 < d_f < 110\,\mu\text{m}$ $0.0022 < \alpha < 0.387$ $1.44\,\text{cm s}^{-1} < U < 6.48\,\text{cm s}^{-1}$	E increases when d_p decreases
2006	Kim et al. [23]	NaCl particles Fiberglass fibrous filters	$1\,\text{nm} < d_p < 100\,\text{nm}$ $d_f = 9.1\,\mu\text{m}$ $\alpha = 0.06$ $U = 2.5\,\text{cm s}^{-1}$	E decreases below 1.3 nm
2007	Kim et al. [24]	Silver particles Fiberglass fibrous filters	$3\,\text{nm} < d_p < 20\,\text{nm}$ $1.9\,\mu\text{m} < d_f < 4.9\,\mu\text{m}$ $0.039 < \alpha < 0.05$ $5\,\text{cm s}^{-1} < U < 15\,\text{cm s}^{-1}$	E increases when d_p decreases
2007	Huang et al. [25]	NaCl particles Fibrous filters for respiratory masks	$4.5\,\text{nm} < d_p$ filter B: $d_f = 13\,\mu\text{m}$ and $\alpha = 0.035$ $2.8\,\text{cm s}^{-1} < U < 8.6\,\text{cm s}^{-1}$	E increases when d_p decreases
2007	Steffens and Coury [26]	NaCl particles Polyester filter	$8.5\,\text{nm} < d_p < 94.8\,\text{nm}$ $d_f = 16\,\mu\text{m}$ $\alpha = 0.118$ $3\,\text{cm s}^{-1} < U < 25\,\text{cm s}^{-1}$	E increases when d_p decreases
2008	Shin et al. [27]	Silver particles Stainless steel wire screen	$3\,\text{nm} < d_p < 20\,\text{nm}$ $d_f = 90\,\mu\text{m}$ $\alpha = 0.3105$ $4.17\,\text{cm s}^{-1} < U < 7.04\,\text{cm s}^{-1}$ $296\,\text{K} < T < 500\,\text{K}$	E increases when d_p decreases and when T increases
2008	Mouret [28]	Cu and C particles Stainless steel wire screens, polyester and fiberglass Fibrous filters	$4\,\text{nm} < d_p < 80\,\text{nm}$ $1.3\,\mu\text{m} < d_f < 200\,\mu\text{m}$ $0.06 < \alpha < 0.39$ $5\,\text{cm s}^{-1} < U < 15\,\text{cm s}^{-1}$	E increases when d_p decreases
2009	VanGulijk et al. [29]	NaCl, $CaCl_2$, $(NH_4)_2SO_4$ and $NiSO_4$ particles Stainless steel wire screen	$6\,\text{nm} < d_p < 20\,\text{nm}$ $d_f = 40\,\mu\text{m}$	E depends on the aerosol type

[*] In an electronic communication to D.A. Japuntich referred in a 3M® technical data bulletin [30], Albert Podgórski, as a co-author of the Balazy's paper, has indicated that: "Unfortunately, those results were erroneous due to the aerosol spectrometer malfunction (WPS, made by MSP Corp.). Now, the instrument is upgraded by the manufacturer and we have also worked out a new experimental procedure; as a consequence, such a decrease of the filtration efficiency is no longer observed."

ing one fiber of the collecting media are collected. The thermal rebound model of Wang and Kasper is built introducing values of ε smaller than one, as the consequence of particle bounce on the fibrous surface. In other words, any contact between particle and fiber no longer systematically leads to collection. Mathematically, if the approach velocity of the particle is higher than the critical velocity V_{cr}, defined as the velocity above which rebound is to occur, the particle will not be trapped.