Abschlussbericht

Erforschung des Einflusses der Umgebungsfeuchtigkeit auf die Morphologie der Partikeln und Partikelkontakte von an Einzelfasern haftenden mineralischen Partikeln mit hygroskopischen Eigenschaften

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Dr.-Ing. Qian Zhang,
Bergische Universität Wuppertal, Fakultät für Maschinenbau und Sicherheitstechnik,
Institut für Partikeltechnologie, Rainer-Gruenter-Straße 21, 42119 Wuppertal

Abstract

Es wurde experimentell online/inline mit bildgebenden Verfahren (ESEM) untersucht, wie die Morphologie der Partikeln und Partikelkontakte von an Einzelfasern haftenden Partikeln aus Mineralsalzen mit hygroskopischen Eigenschaften durch Änderung der Umgebungsfeuchtigkeit beeinflusst werden. Dazu wurde gasgetragene trockene Feststoffpartikeln mittels eines Atomizer-Systems generiert, die anschließend durch Filtration an definiert angeordneten Einzelfasern abgeschieden werden. Ein neu konstruiertes und gebautes Einzelfaser-Einspann-Minimodul ermöglichte hierbei eine direkte Probennahme sowie eine zerstörungsfreie Adaption der Probe für die nachfolgende Untersuchung im ESEM.

Einzelne Arbeitsergebnisse

Siehe Anlagen.

Fazit und Ausblick

Die Konstruktion der Einspannvorrichtung für Einzelnfaser (auch für andere Einzelkollektoren) konnte erfolgreich abgeschlossen werden. Aufgrund einer unerwarteten, längen technischen Störung des ESEMs konnte ein beträchtlicher Teil der geplanten Untersuchungen im ESEM leider nicht wie geplant in dem festen Förderzeitraum ausgeführt werden. Die erzielten Arbeitsergebnisse lieferten dennoch einen wichtigen Beitrag zu einem erfolgreichen Antrag eines laufenden DFG-Forschungsvorhabens (Thema: "Zum Einfluss einer zudosierten Fraktion von hygroskopischen Salzpartikeln auf das Betriebsverhalten von Oberflächenfiltern zur Gasreinigung durch feuchteinduzierte Deliqueszenz und Effloreszenz").

Veröffentlichungen (als Anlagen beigefügt)

Ein Vortrag wurde bei der internationalen Konferenz FILTECH 2015 (Köln, Febr. 2015) gehalten. Titel des Vortrags bzw. des Konferenzpapers: "*Experimental investigations into the effects of ambient humiduty on particle-loaded single filter fibers*".

Ein Poster-Präsentation wurde bei dem ProzessNet-Jahrestreffen der FG Agglomerations- und Schüttguttechnik (Magdeburg, März 2015). Titel der Präsentation: "*Untersuchung zum Einfluss der Umgebungsfeuchte auf die Morphologie der Partikeln und Partikelkontakte von an Einzelfasern haftenden mineralischen Partikeln mit hygroskopischen Eigenschaften*".

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Institut für Partikeltechnologie

Untersuchung zum Einfluss der Umgebungsfeuchte auf die Morphologie der Partikeln und Partikelkontakte von an Einzelfasern haftenden mineralischen Partikeln mit hygroskopischen Eigenschaften

Hintergrund und Aufgabenstellung

- Es ist bekannt, dass die Umgebungsfeuchte in vielen Verfahren der Partikeltechnologie die Eigenschaften der Feststoffpartikeln bzw. deren Wechselwirkungen untereinander sowie zu den mit denen in Kontakt stehenden Kollektoren/Wandungen beeinflusst.
- Hygroskopische Salzpartikeln zeigen – als Aerosolpartikeln in feuchter Umgebung – die typischen Deliqueszenz- und Effloreszenz-Eigenschaften, die eine Möglichkeit zur gezielten Beeinflussung der Partikelkontakte durch geregelte Feuchteänderungen darstellen.

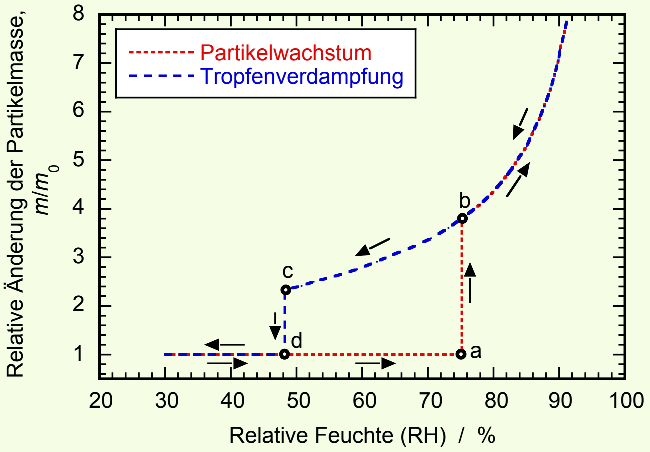

Abb. 1: Wachstum und Verdampfung einer NaCl-Partikel mit einer Größe im Submikronbereich in einer feuchten Gasumgebung bei 25 °C. Für eine schwebende Einzelpartikel in einem elektrodynamischen Gleichgewicht [1].

- Es wird experimentell online/inline mit bildgebenden Verfahren untersucht, wie die Morphologie der Partikeln und Partikelkontakte von an Einzelfasern haftenden Partikeln aus Mineralsalzen mit hygroskopischen Eigenschaften durch Änderung der Umgebungsfeuchte beeinflusst wird.

Methodisches Vorgehen

Schritt 1: Probenpräparation/Probennahme

Gasgetragene trockene Feststoffpartikeln werden mittels eines Atomizer-Systems generiert. Die Feststoffpartikeln werden durch Filtration an definiert angeordneten Einzelfasern abgeschieden. Ein neu konstruiertes Einzelfaser-Einspann-Minimodul ermöglicht hierbei eine direkte Probennahme sowie eine zerstörungsfreie Adaption der Probe für das nachfolgende Untersuchungsverfahren.

Schritt 2: Untersuchung in einem ESEM

Während die Umgebungsfeuchte (RH) im Grobvakuum-Bereich mittels Wasserdampf geregelt geändert wird, werden Partikeln und Partikelkontakte mit dem Mikroskop abgebildet.
(***ESEM***: ***E****nvironmental* ***S****canning* ***E****lectron* ***M****icroscope*)

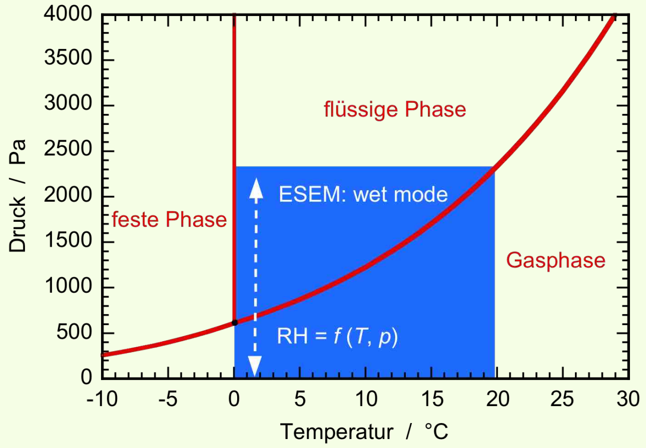

Abb. 2: Arbeitsbereich eines ESEMs mit Wasserdampf in der Probenkammer, dargestellt im Phasendiagramm des Wassers.

Probenpräparation

- Zur Probenpräparation mit Einzelfasern wurde ein Einzelfaser-Einspann-Minimodul konstruiert und angefertigt. Einzelfasern werden hier mithilfe von zwei Kämmen in paralleler Anordnung eingespannt. Über verschiedene Adapter lässt sich das Modul in eine modifizierte Labor-Filterprüfapparatur nach VDI 3926-1 (Typ 1) zum Anfiltrieren von trockenen Salzaerosolen einbauen.
- Die trockenen Salzaerosole werden mittels des Atomizer-Systems TSI 3076 generiert. Durch Variation der Konzentration der Salzlösung und der Filtrationsdauer können Einzelfasern mit unterschiedlichen Partikelbeladungen präpariert werden.
- Die Möglichkeit das Einzelfaser-Einspann-Minimodul direkt an den Kühltisch in der Probenkammer des ESEMs zu adaptieren ist gegeben.

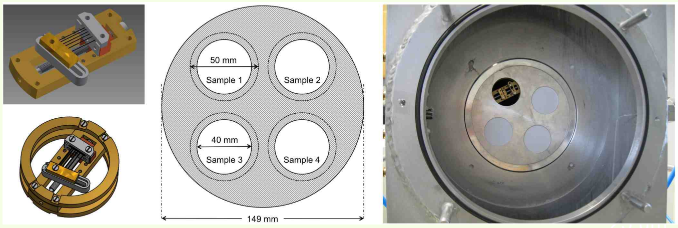

Abb. 3: Einzelfaser-Einspann-Minimodul mit verschiedenen Adaptern zum Einbau in die Labor-Filterprüfapparatur.

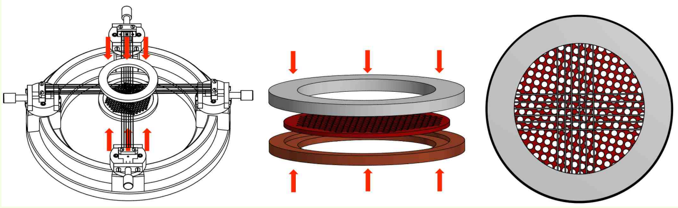

Abb. 4: Einspannen und Fixieren von Einzelfasern mit einem neuen Konzept. Die Endfixiereinheit lässt sich direkt an den Kühltisch des ESEMs adaptieren.

Erste Ergebnisse im ESEM

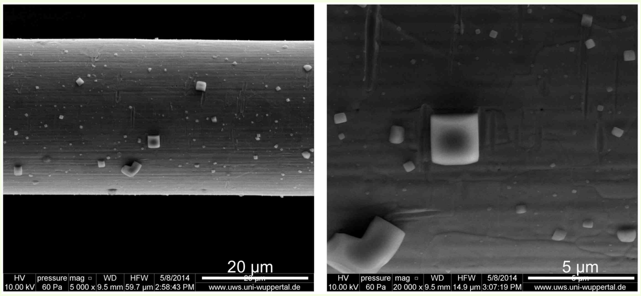

Abb. 5: REM-Aufnahmen einer partikelbeladenen Einzelfaser in zwei verschiedenen Vergrößerungen. Faser aus Edelstahl; Partikeln aus NaCl.

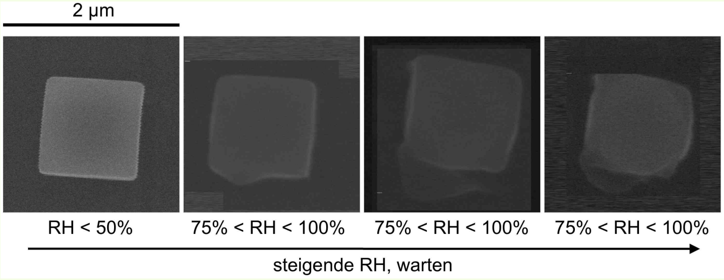

Abb. 6: ESEM-Aufnahmen einer NaCl-Partikel auf der Faseroberfläche. Von l. nach r.: steigender Druck in der Probenkammer (Kühltischtemperatur: 1 °C).

[1] I. N. Tang, Phase Transformation and Growth of Hygroscopic Aerosols, in *Aersol Chemical Processes in the Environment*, edited by K. R. Spurny, chap. 4, pp. 61–80, CRC Press LLC, Boca Ranta, Florida, 2000

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Dr.-Ing. Qian Zhang **www.ipt.uni-wuppertal.de** **zhang@uni-wuppertal.de**

EXPERIMENTAL INVESTIGATIONS INTO THE EFFECTS OF AMBIENT HUMIDITY ON PARTICLE-LOADED SINGLE FILTER FIBERS

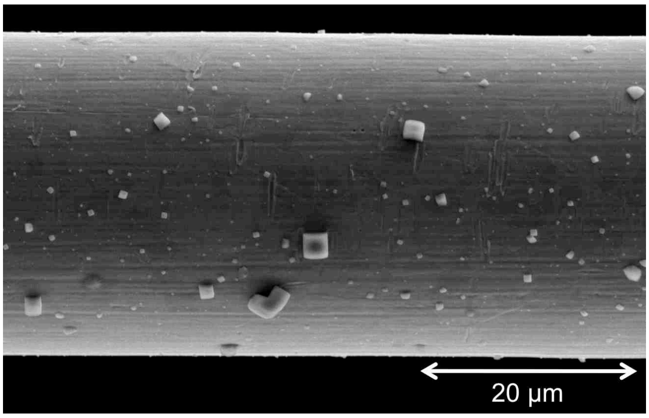
Dr.-Ing. Qian Zhang
University of Wuppertal, Faculty D – Institute of Particle Technology,
Wuppertal, Germany
e-mail: zhang@uni-wuppertal.de

Background and Aim

It is known in many processes of particle technology that the ambient humidity influences the properties of solid particles, the interactions between solid particles and their interactions with collectors or walls that are in contact with these particles. Many experimental observations, however, could not be clarified in a satisfactory manner with existing knowledge. An example of such a case is the phenomenon in separating mineral dust particles from gases with surface filters that the pressure drop across the dust-loaded filters can change abruptly at a moderate change of the humidity of the gas. And such change in the pressure drop can be reversed in some cases and cannot be reversed in others. This paper describes an experimental approach to investigate the effects of ambient humidity on particle-particle and particle-wall contacts based on particle-loaded single filter fibers.

Method and Results

The morphology of the mineral particles with different hygroscopic characteristics and particle contacts on single filter fibers is observed on-line/in-line through controlled changes in the ambient humidity using an Environmental Scanning Electron Microscope (ESEM). Additional information regarding the microscopic images could be gained by an accompanying EDX analysis. Sampling of the particle-loaded single filter fibers is done on a filter test rig by filtration of airborne particles, which are generated by a spray atomizer connected with a diffusion dryer for salt solutions or mineral particle suspensions. Single filter fibers, like metallic fiber, hydrophilic synthetic filaments or hydrophobic synthetic filaments, are fixed in a specific order like parallel arrays in a special designed fiber-holding unit, which is mounted in the filter holder of the filter test rig during the sampling of the particle-loaded fibers. After the sampling on the filter test rig is finished, the fiber-holding unit is demounted from the filter holder and directly mounted in the specimen chamber of the ESEM for observation and measurement. The relative humidity in the specimen chamber is increased step-by-step and, finally, beyond the deliquescence humidity of the salt. Two different self-designed single fiber-holding units and results with a single stainless steel fiber (Ø = 30 µm) and sodium chloride particles are presented in this paper.



KEYWORDS
Single Fiber, Particle Morphology, Hygroscopicity, Sodium Chloride, SEM

1. Introduction

It is known in many processes of particle technology that the ambient humidity influences the properties of solid particles, the interactions between solid particles and their interactions with collectors or walls that are in contact with these particles. Many experimental observations, however, could not be clarified in a satisfactory manner with existing knowledge. An example of such a case is the phenomenon in separating mineral dust particles from gases with surface filters that the pressure drop across the dust-loaded filters can change abruptly at a moderate change of the humidity of the gas. And such change in the pressure drop can be reversed in some cases and cannot be reversed in others.

Hygroscopic aerosol particles exhibit the properties of deliquescence and efflorescence in humid air. Tang et al. investigated the phase transformation and growth of hygroscopic aerosols by single particle levitation in an electrodynamic balance [1, 2, 3, 4]. The phase transformation, growth and evaporation process obtained for a single micro-sized pure NaCl particle in humid environment at 25°C are illustrated in Figure 1.

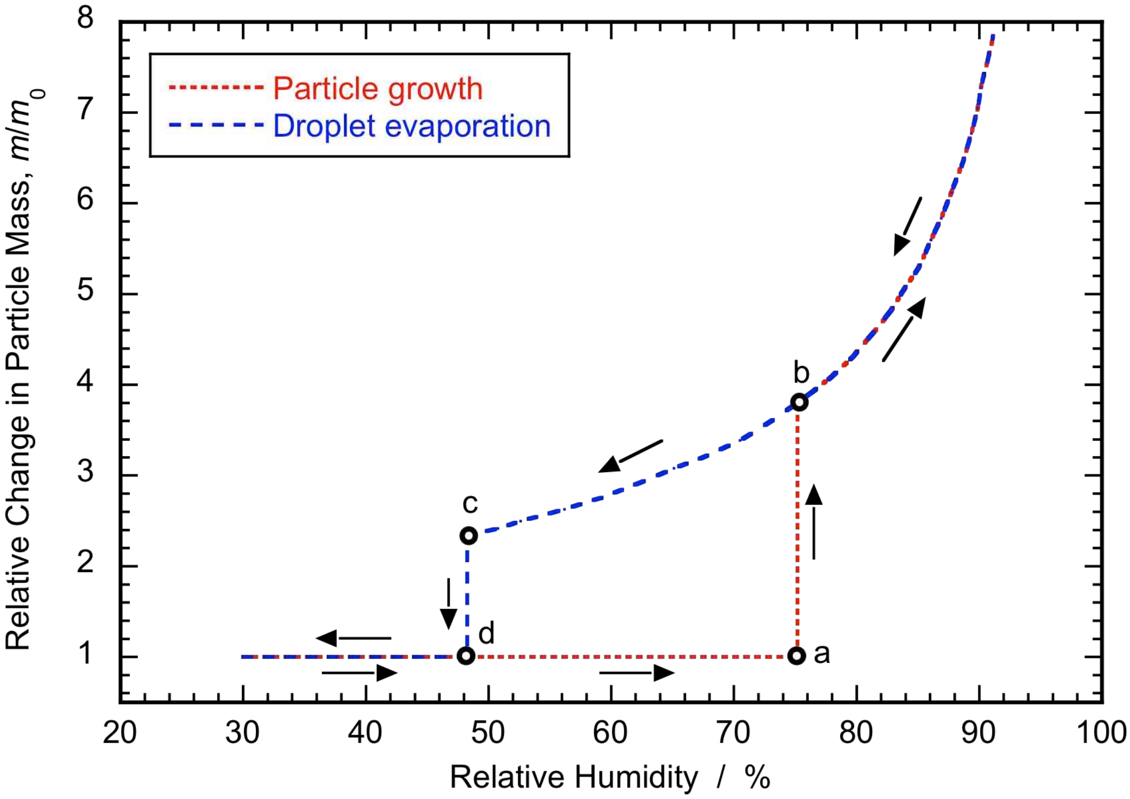


Figure 1 Growth and evaporation of a single micron-sized NaCl particle in humid environment at 25°C. Observation by single particle levitation in an electrodynamic balance [1].

Here, the particle mass change with respect to the dry salt mass, m/m_0, is plotted as a function of relative humidity (RH). As RH increases, a crystalline NaCl particle remains unchanged in mass until RH reaches its deliquescence point at 75,4% RH. Then, it deliquesces spontaneously (from point a to point b) to form a saturated solution droplet by water vapour condensation. The droplet grows continuously and smoothly by absorbing ambient water vapour as RH is further increased. As RH gradually decreases, the solution droplet loses its mass by water evaporation. Until point b is reached, the droplet mass change is exactly reversible. And then, it remains a solution droplet even beyond its saturation point and becomes highly supersaturated as a metastable droplet (from point b to point c) at RH much lower than the deliquescence point. As the droplet suddenly sheds all its water content and becomes a solid particle at about 48% RH, efflorescence occurs (from point c to point d). The hysteresis shown here represents a typical hydration behaviour exhibited by all hygroscopic aerosol particles. Tang pointed out, furthermore, that, for a pure single salt particle, the particle is either a solid or a droplet, but not in a state of partial dissolution [1].

If salt particles with hygroscopic properties are now deposited on walls, like in the case of dust-loaded filters, there will be no single, isolated aerosol particles, but always particle-wall contacts and / or particle-particle contacts. Due to new interfaces and heterogeneity of the particle composition, the typical phase transformation, growth and evaporation processes described above for hygroscopic aerosol particles may considerably change for these particles deposited on walls or in contact with other particles.

This paper describes an experimental approach using an imaging technique to investigate the effects of ambient humidity on particle-particle and particle-wall contacts based on particle-loaded single filter fibers.

2. Methods, Equipments and Materials

2.1. Methods

The experimental investigation is done in two separate steps:

- In the first step, particle-loaded single fibers will be prepared by filtration of airborne solid particles of hygroscopic materials in a relative dry environment (RH < 35% at 20…25°C). For a well-defined Arrangement of the single fibers, special clamping modules (fiber-holding units) were designed and manufactured.
- In the second step, well-prepared samples will be mounted into the specimen chamber of the ESEM and investigated by imaging deposited particles, particle-particle contacts and particle-fiber contacts during a controlled change in the ambient relative humidity of the specimen.

2.2. Equipments and Materials

2.2.1. Filter test rig and aerosol generator

A modified filter test rig built according to the VDI 3926-1, application 1, as a filtration device and an atomizing system as an aerosol generator were used for sample preparation.

Figure 2 shows the scheme of a standard filter test rig and the modified variant used here [5, 6, 7]. On the opposite side of the clean gas tube, through an opening in the wall of the crude gas channel, an aerosol channel connected with a particle feeding system was applied to supply airborne solid particles for filtration (Fig 2, right). Figure 3 shows the salt solution-atomizing system (TSI Model 3076). The particle size distribution of the generated aerosol depends on the concentration of the salt solution. The particle loading on the fibers is adjusted by controlling the duration of the filtration.

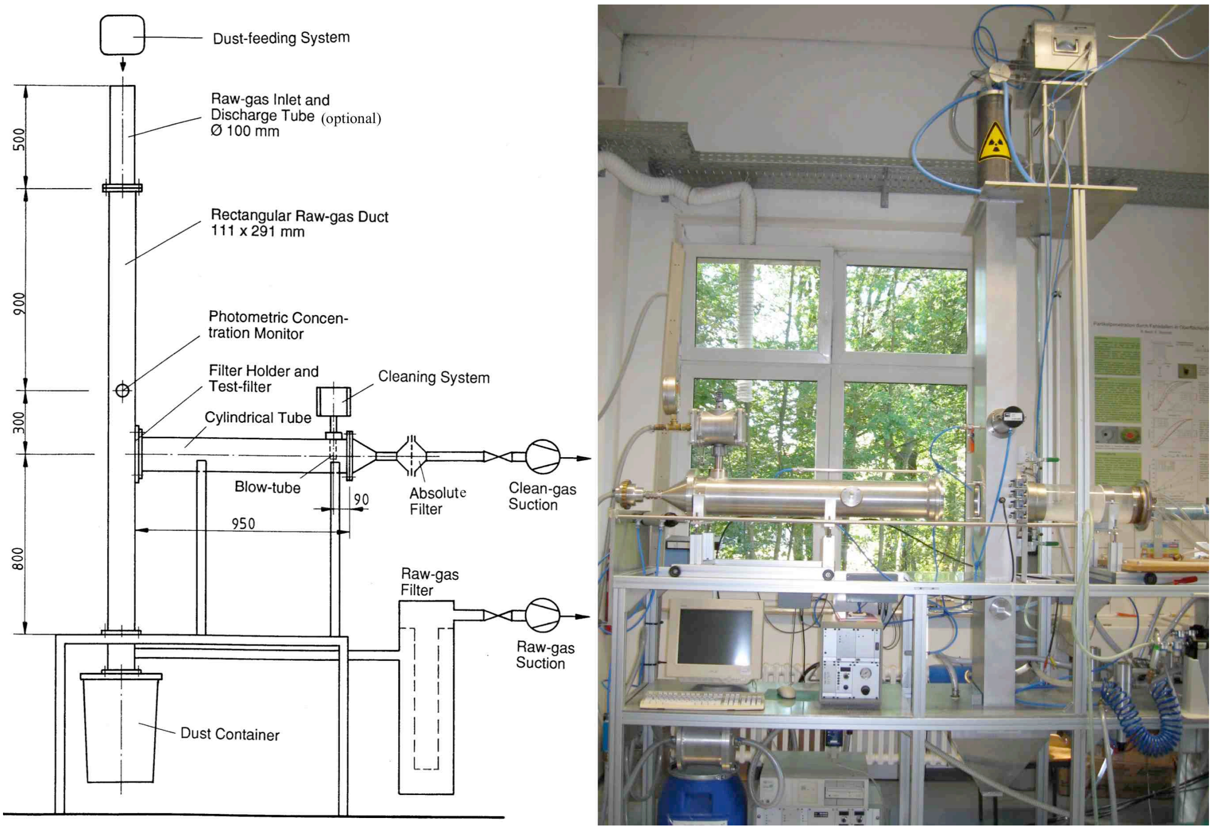


Figure 2 Scheme of a standard filter test rig according to VDI 3926-1 (l.) and photograph of the modified filter test rig used for the experimental investigations (r.).

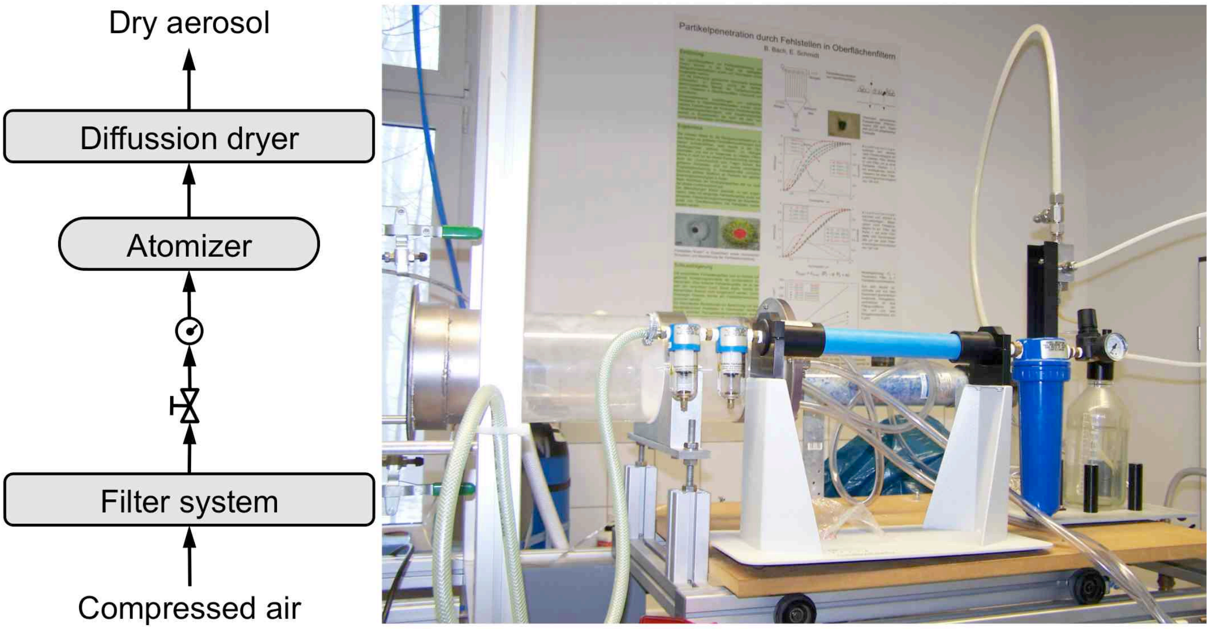


Figure 3 Operation scheme of the atomizing system TSI model 3076 (l.) and photograph of the atomizing system connected with an aerosol channel as a crude gas channel for filtration on the filter test rig (r.).

2.2.2. Environmental Scanning Electron Microscope (ESEM)

The Environmental Scanning Electron Microscope (ESEM) is a modified SEM and was developed in the 1980s [8]. The high vacuum, which is an essential condition for the normal SEM mode, is not necessary for the operation in the ESEM mode. Here in the so-called “wet mode”, the previously evacuated specimen chamber is vented to a pressure of some mbar of water vapour, so that, to a good approximation, only water molecules are present in the chamber. Using a Peltier cooling stage onto which the specimen is mounted, the temperature of the specimen can be modified and hold within a certain range. So the ambient relative humidity of the specimen is defined as a function of the specimen temperature and the specimen chamber pressure (RH = $f(T, p)$). In Figure 4, the operation range of an ESEM is shown schematically. At a certain temperature controlled by the cooling stage, the ambient relative humidity of the specimen can be modified by adjusting the chamber pressure.

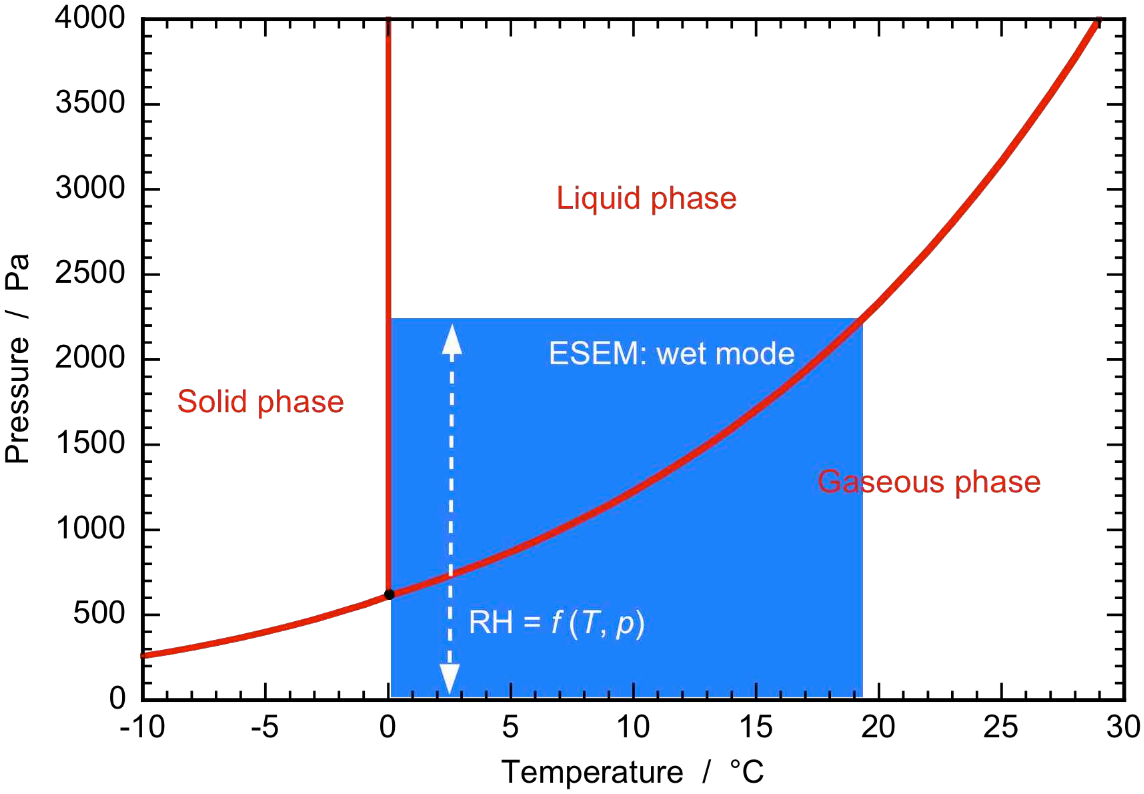


Figure 4 Phase diagram of water with the range of the wet mode of an ESEM by using water vapour for the gaseous environment in the specimen chamber.

2.2.3. Single fiber clampimg module

To create variants of particle-particle and particle-wall contacts based on particle-loaded single filter fibers, it is intended to vary the arrangement of single fibers and the particle mass deposited on the fibers. The sample variants for the investigation in the ESEM should include single particle contacts, formed dendrites, as well as a completely formed particle layer. For this purpose a clamping module was constructed for fixing / holding single fibers in a parallel array with different distances from each other, which is also used for preparing particle loading in the filtration device described above. A major challenge for this experimental approach is how to

mount the prepared single fibers with a sensitive particle loading into the specimen chamber non-destructively. It will be particularly important when a fragile particle layer is built on the fibers. Furthermore, a good temperature control of the sample on the cooling stage in the specimen chamber must be ensured.
Two clamping module variants A (see Fig. 5 and Fig. 6) and B (see Fig. 7 and Fig. 8) were designed and manufactured.

Self-designed clamping module variant A

At the variant A, single fibers (cut out from endless filaments) are mounted on two special combs with a defined distance between the teeth, which are individually fixed on two movable bars used for tensioning the fibers (Fig. 6). This compact single fiber-holding unit can be adapted both for mounting into the filtration device for particle loading and for mounting into the specimen chamber of an ESEM for further investigation (Fig. 5).

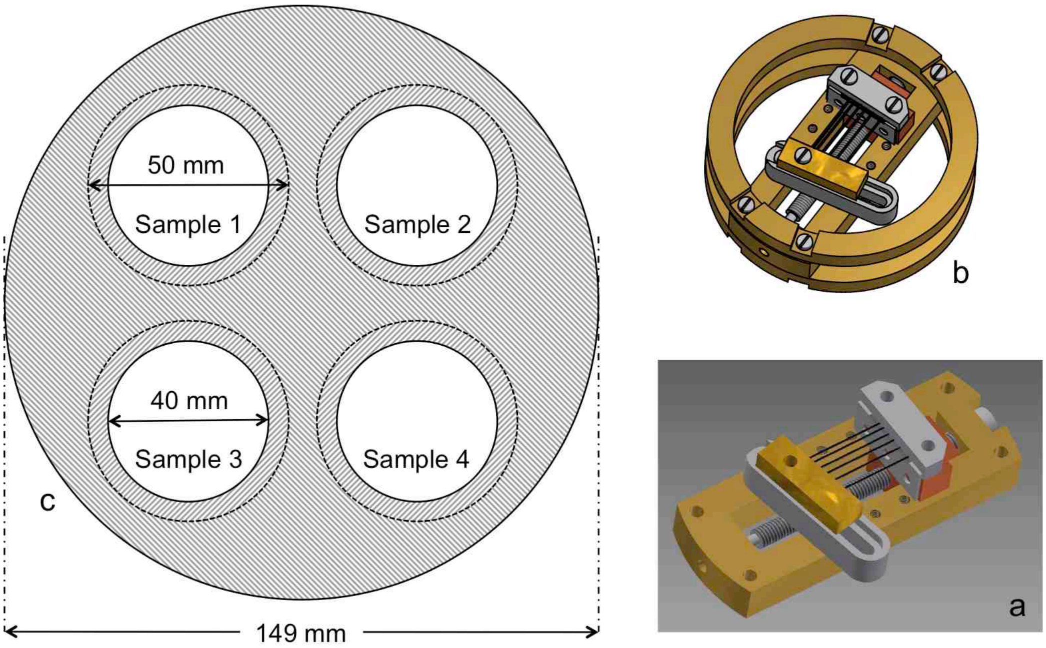


Figure 5 Scheme of the units of the self-designed clamping module variant A. Unit a: fixing and holding unit for single fibers; unit b: an intermediate adapter both for mounting into the apparatus for particle loading and for mounting into the specimen chamber of an ESEM (on the cooling stage); unit c: an adapter for the filter holder of the filter test rig (see Fig. 2) to hold unit b.

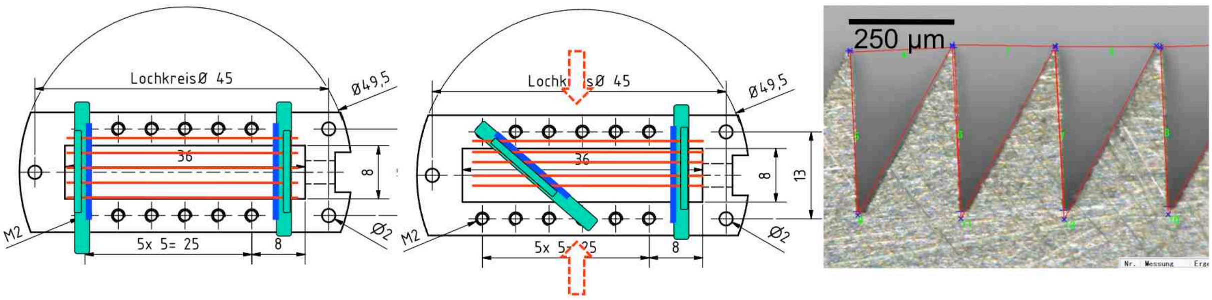
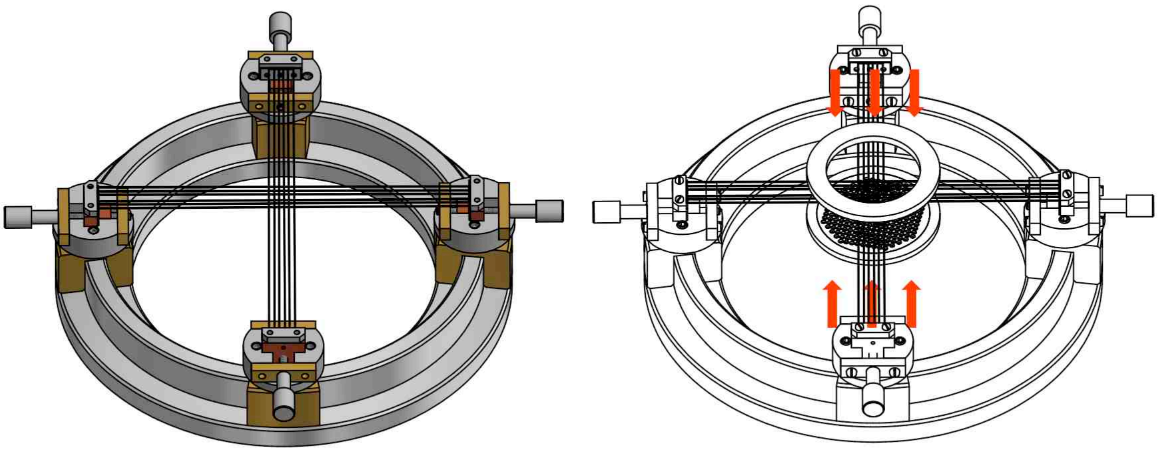


Figure 6 Scheme of the fixing and holding unit for single fibers of the self-designed clamping module variant A (l. and m.) with magnified comb for spacing fibers (r.).

Self-designed clamping module variant B

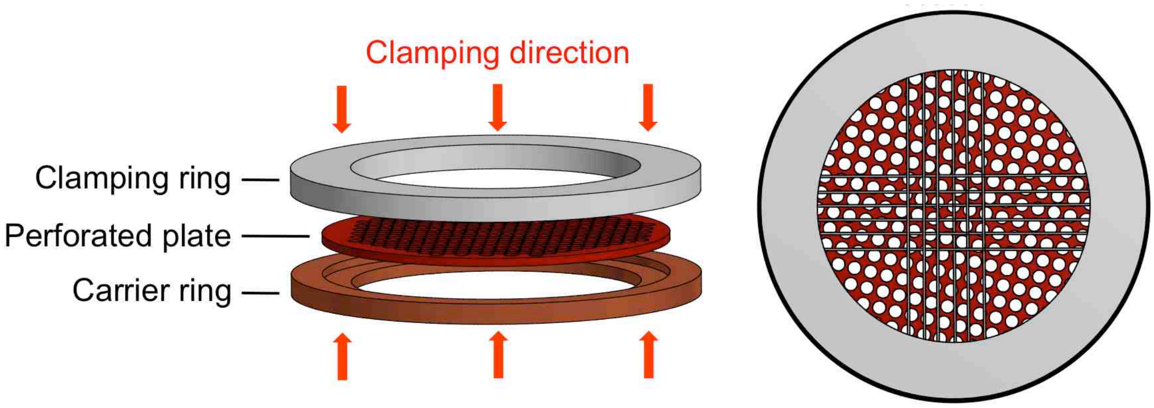
At the variant B, the step of arranging and tensioning the single fibers (cut out from endless filaments) is separated from the fiber-holding unit by a larger and more flexible unit (Fig. 7, a). After this step the fibers tensioned and fixed in a certain arrangement will be clamped into the final fiber-holding unit (Fig. 7, b) and then cut off from the larger holding unit. In the final fiber-holding unit the fibers are directly located on the flat perforated metal plate, so that the temperature of the specimen on the cooling stage should be controlled more precisely than at the variant A. And this will be particularly important when the fibers have poor heat transfer characteristics, like in the case of synthetic fibers.



a) Fixing single fibers

b) Clamping fiber arrays

Figure 7 Scheme of the two preparation steps for the self-designed clamping module variant B. Step a): single fibers will be fixed in two parallel arrangements, which can be crossed at a variable angle. Step b): single fibers fixed in a defined arrangement will be clamped into the final fiber-holding unit (cf. Fig. 8), which will be used for particle loading.



a) Side view (without fibers)

b) Top view (with clamped fibers)

Figure 8 Scheme of the final fiber-holding unit of the self-designed clamping module variant B, which will be used to collect particles by filtration in a suitable apparatus (see Fig. 5, c) and then be mounted directly on the cooling stage in the specimen chamber of an ESEM.

The clamping module variant B can furthermore be used in combination with a TSI Particle Size Selector Model 376060 for preparing particle loading [9]. In that case, the original TSI screens for selecting aerosol particles will be replaced by the fiber-holding unit of the clamping module variant B. One can also use an original TSI screen to substitute the single fibers in the fiber-holding unit to load particles, if a homogenous particle layer on the substrate is wanted.

3. First Results and Discussion

Figure 9 shows two SEM images of a particle-loaded single fiber in normal SEM mode with two different magnifications. Endless filament with a diameter of 30 µm from stainless steel was used to prepare the single fiber fixed in the holding unit of the clamping module variant A and sodium chloride was used to prepare the particles. It is clear to see that the crystalline NaCl particles are cube-shaped.

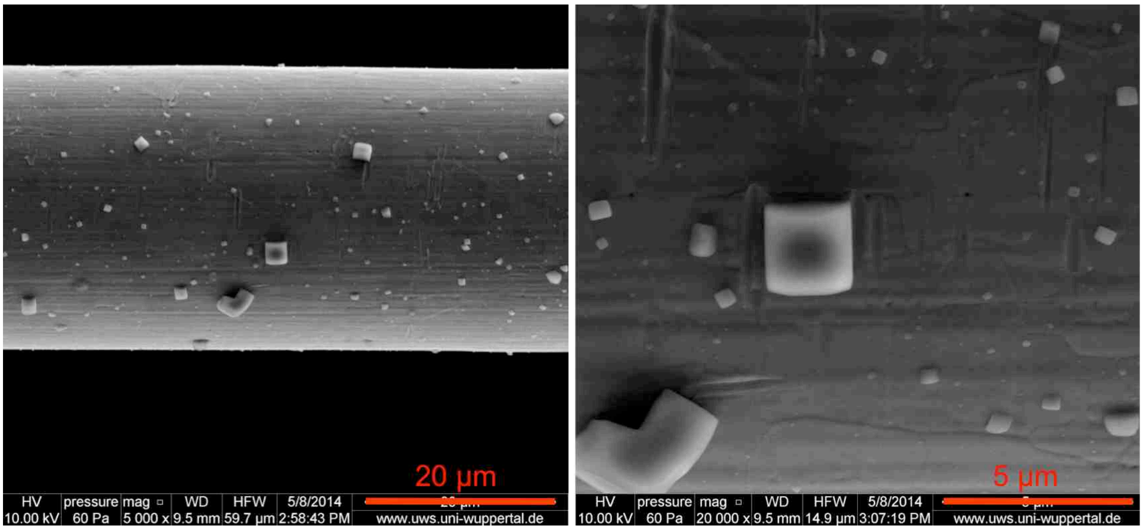


Figure 9 SEM images of a particle-loaded single fiber in normal SEM mode with two different magnifications. Fiber material: stainless steel, particle material: NaCl.

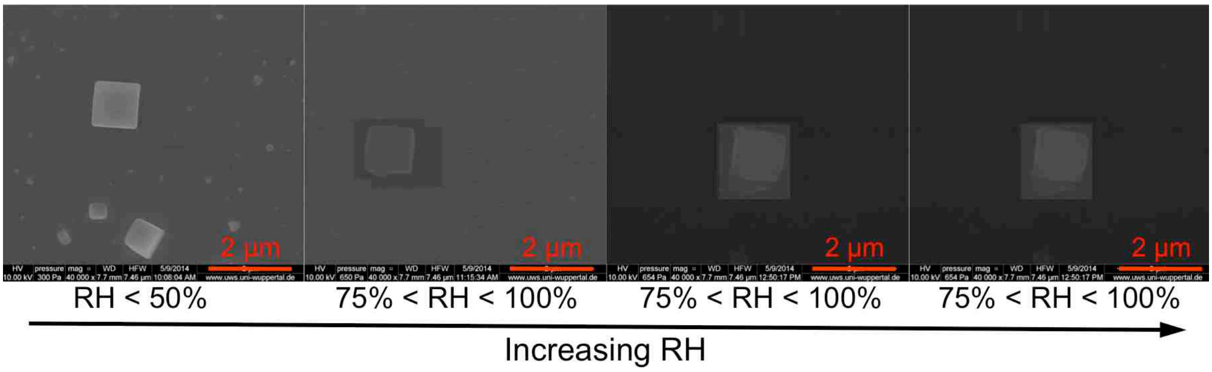


Figure 10 Images of a NaCl particle deposited on a single fiber (see Fig. 9) in ESEM mode with gradually increasing specimen chamber pressure using water vapour. Set temperature of the Peltier cooling stage: 1 °C.

Figure 10 shows the images of a selected NaCl particle in ESEM mode with gradually increasing specimen chamber pressure using water vapour. It is to see that the NaCl particle begins to change its form at a RH beyond its deliquescence point. The edges of the crystal become partly round; but no spherical shape is visible. So it can be

supposed that the NaCl particle shown here is in a state of partial dissolution. And this behaviour differs from that of a single, isolated NaCl aerosol particle. One reason for this may be that the particle-fiber contact constitutes a new interface, which influences the thermodynamics between the particle and the water vapour.
It is noted here that with the clamping module variant A the temperature control of the particle-loaded fiber may be not exact. For further studies the clamping module variant B will be used.

Acknowledgements

Financial support by the Max Buchner Research Foundation is acknowledged gratefully.

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