EPJ Web of Conferences **358**, 01001 (2026) https://doi.org/10.1051/epjconf/202635801001
EFM 2025

A study of the droplet nucleation in dependence on the gas mixture origin

Ondřej Bartoš[1*], *Adam* Huněk[1], and *Jan* Hrubý[2]

[1]CTU in Prague, Energy Engineering Department, Technická 4, Prague 6, 166 07, Czechia
[2]Institute of Thermomechanics, Czech Academy of Sciences, Dolejškova 5, 182 00, Praha, Czechia

Abstract. This paper presents the development and validation of the Fine Aerosol Generation Unit (FAGU), a novel instrument designed to support experimental research on nucleation phenomena, particularly at the interface between heterogeneous and homogeneous nucleation. The FAGU system enables precise and independent control of key aerosol parameters, including particle number concentration, relative humidity, and temperature. A core requirement in its design was the ability to generate aerosols composed of nanometer-sized particles of a single substance, with tunable particle density—an essential capability for reproducible and systematic nucleation studies. The aerosols produced by FAGU are intended for use in controlled experiments investigating unary, binary, and heterogeneous nucleation processes, such as those conducted in expansion chambers. To verify the unit's performance, a series of experiments were conducted using different aerosol and humidity sources, including a single-phase nozzle, a nebulizer, and surface evaporation techniques. These tests demonstrated the unit's ability to decouple and regulate aerosol concentration and humidity independently, offering a high degree of flexibility for tailoring initial conditions in nucleation experiments. The results confirm that FAGU provides a robust and versatile platform for generating well-defined aerosol environments, making it a valuable tool for advancing the understanding of particle formation mechanisms in both atmospheric and laboratory settings.

1 Introduction

Rapid adiabatic expansion processes, often accompanied by condensation of the working fluid, are encountered across a range of industrial applications, including steam turbines, compressed air systems, and natural gas processing. These phenomena are also increasingly relevant in emerging technologies related to carbon capture, transport, utilization, and storage. The underlying mechanisms of droplet formation are typically categorized into three nucleation pathways: unary homogeneous nucleation (involving pure steam), binary homogeneous nucleation (involving mixtures of vapors), and heterogeneous nucleation (occurring in the presence of aerosol particles). However, real-world conditions frequently give rise to transitional regimes in which the nucleation pathway cannot be distinctly classified.

This study contributes to a broader research initiative focused on nucleation processes in energy systems, with particular emphasis on these transient cases. The present investigation focuses on water vapor condensation influenced by additional gaseous compounds and/or aerosol particles. During rapid adiabatic expansion, water vapor commonly condenses into supercooled liquid droplets, a process that is highly sensitive to the surrounding chemical and particulate environment. While water is the primary working fluid examined, the methodology allows for the inclusion of other vapor pairs where their behavior may elucidate fundamental aspects of phase transition dynamics.

By exploring these transitional nucleation regimes, the research aims to refine the conceptual understanding of condensation under non-ideal conditions and to support the development of predictive models applicable to both conventional and emerging energy technologies [1, 2].

Humid air is a ubiquitous mixture of atmospheric gases and water vapor. Other examples of vapor – gas mixtures are natural gas, biogas or flue gas. The phenomenon of dropwise condensation, commonly occurring in nature and technology, is intimately connected with non-ideality of the gas phase. A key problem in the theory of nucleation (formation of droplets from free vapor molecules) is determining the distribution of clusters, i.e., the concentrations of dimers, trimers, tetramers, etc. At low temperatures, cluster concentrations can be directly related to virial coefficients, which define the real-gas equation of state (EOS) of the vapor. It was shown that less ideal gases promote nucleation by adsorbing on the surface of larger clusters and thus decreasing their formation work. At the level of the classical nucleation theory (CNT), this can be explained by a reduction of the surface tension of microscopic droplets following from the Gibbs adsorption equation. Thermodynamic effects of interactions of vapor and gas molecules are

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[*] Corresponding author: ondrej.bartos@fs.cvut.cz

characterized by virial cross-coefficients. It appears that an integrated study of the real-gas EOS for a vapor-gas mixture in conjunction with the study of nucleation can bring new insights, advancement of theoretical understanding of these phenomena and new quantitative models, applicable to various processes in nature and technology.

2 Experimental setup

The experimental setup was designed to regulate the concentration of aerosol particles while independently controlling the humidity of the carrier gas, as described by Hinds [3]. This capability is critical for accurate measurements within the expansion chamber.

The target aerosol particles are heterogeneous and expected to range from a few nanometers to several hundred nanometers in diameter. The most widely used method for generating particles of this size involves drying droplets formed from a sprayed solution. Consequently, the final particle size can be controlled by adjusting both the droplet size during primary atomization and the concentration of dissolved salts in the solution.

Fig. 1. The experiment setup.

The apparatus includes a nozzle for spraying the solution, after which the aerosol is mixed with a stream of dry, impurity-free air. To ensure air purity, an oil-free compressor supplies the airflow, which is subsequently passed through a filtration system ending with a HEPA filter rated for the specified flow rate.

The carrier gas flow rate is adjustable within the range of 0 to 50 L/min and is precisely regulated using a mass flow meter.

Figure 1 illustrates the complete experimental setup, while Figure 2 presents a schematic diagram of the system.

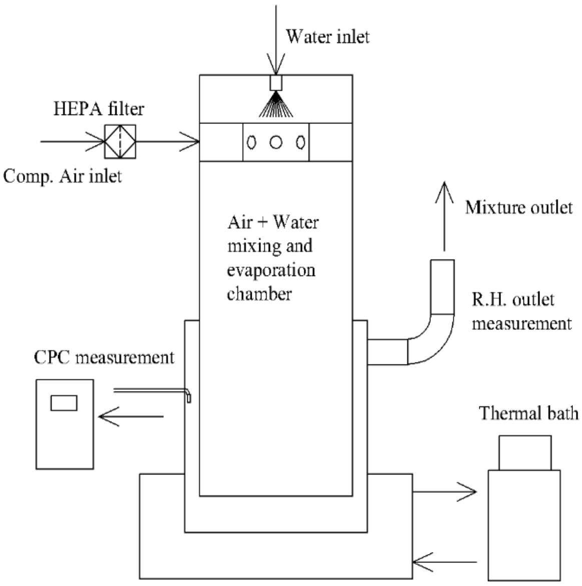

Fig. 2. The schema of the experiment.

After mixing, the aerosol-laden stream passes through a settling chamber, allowing sufficient time for the evaporation of excess moisture. The resulting mixture—comprising heterogeneous nuclei, carrier gases, and water vapor—then proceeds to the outlet of the FAGU. A combined humidity and temperature sensor is positioned at the outlet to monitor environmental conditions.

Before exiting the device, the flow passes through an isokinetic intake, which directs a portion of the aerosol stream into a Condensation Particle Counter (CPC, TSI Model 3756). This intake is also intended to serve as the future connection point for the expansion chamber.

If required, the outlet humidity can be independently adjusted. This is achieved either by modifying the free water level at the bottom of the device and by heating the device’s base to enhance water vaporization.

The entire process is almost isobaric with direct connection to the atmosphere. The correct functioning of the device is assumed when the outlet gas is not saturated with water vapor, and the relative humidity is less than 100%. It is necessary to remove all unbound liquid in the outlet gas, otherwise the result would be significantly distorted.

3 Atomizers

The primary aerosol source is a Schlick series 220 nozzle. This nozzle is single-phase and operates at a pressure of 0.6 MPa. The nozzle generates an aerosol, which for simplicity can be described by the Sauter mean diameter $D_{32} = 30\mu m$ [4]. However, for future measurements, the size of the droplets and the resulting heterogeneous nuclei is relatively large, especially for measurements at higher supersaturations. For this reason, the line was supplemented with a medical nebulizer, which produces droplets of approximately $D_{32} = 4 \div 6\ \mu m$ in size. It is impossible to use such large droplets while maintaining strict limits on the purity of the sprayed water to create heterogeneous nuclei of several nanometers in size. The course of the cumulative functions for both sources is shown in Fig. 3.

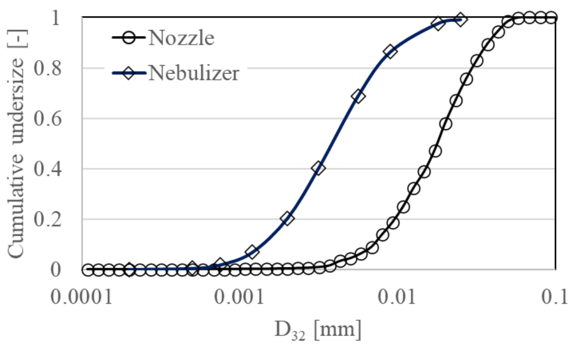

Fig. 3. The cumulative distribution for the nozzle and the nebulizer.

4 Results

This chapter will present the results of test measurements that were performed to verify the expected properties of FAGU. The main purpose of the FAGU device is to prepare a defined aerosol that will be used for further experiments, for this reason it is necessary to check the operation under different operating modes. Fig. 4 shows the course of the output humidity and temperature. At the same time, the numbers of detected particles in the CPC are plotted. No liquid was dosed into the FAGU, this is a dry measurement. The experiment was carried out for several temperatures from 25°C to 60°C. This measurement proves that the source of foreign particles in FAGU is not temperature dependent. These can be various residues of volatile substances or oils.

From the course of the measurement, it is clear that the numbers of particles detected by the CPC are almost zero, which also means that the entire track is well separated from the surroundings. The concentration of particles in the laboratory during the measurement is around 5000 #/cm^3, depending on the conditions.

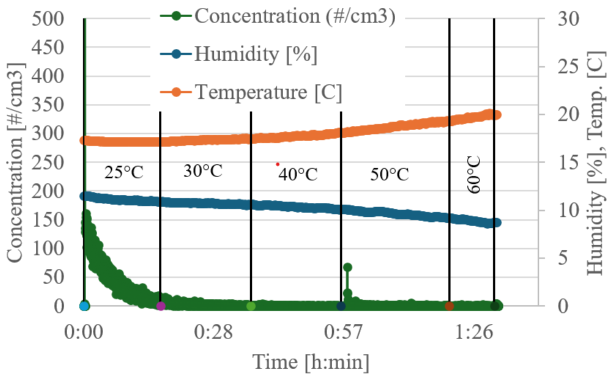

Fig. 4. The temperature, humidity and particle concentration profile for the dry measurement at rising temperature of the bath.

In Fig. 4. it is clearly visible the slow decrease of relative humidity which is related to the temperature change as the bath temperature increased. The increase of bath temperature does not affect the concentration of particles at the outlet of the FAGU. The decrease of concentration which is obvious for the bath temperature of 25C is caused by the fact that the FAGU space has not yet been completely washed with clean air.

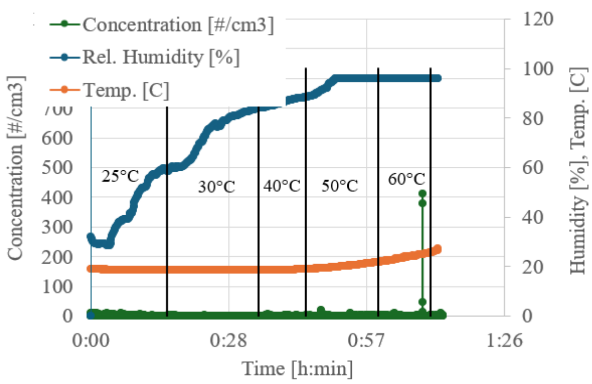

Fig. 5. The temperature, humidity and particle concentration profile for the wet measurement at rising temperature of the bath.

Fig. 5. shows an experiment similar to the previous one, where the FAGU bottom was filled with 400 mL of water with free surface. In this case, a rapid increase in relative humidity is visible, up to complete saturation at 50 C. The experiment was again carried out in such a way that the bath temperature gradually increased. The concentration of particles in the outlet gas is negligible. The possibility of changing the humidity of the gas mixture outlet independently of the injected water is a great advantage.

Fig. 6 shows an example of a measurement with a Schlick model 220 nozzle. The nozzle operates at a pressure of 0.6 MPa, the water flow rate is 13 ml/min. The flow rate of drops into the FAGU is limited by an aperture so that all drops entering the stilling chamber can be evaporated. The measurement shows that after the start of water injection, the humidity of the outgoing air increased. The air flow rate into the device was 12 L/min. The number of heterogeneous particles captured by the CPC reached 3000 #/cm3. Water was sprayed through the nozzle for 12 minutes, after which time there was a gradual decrease in humidity and a decrease in the recorded particles.

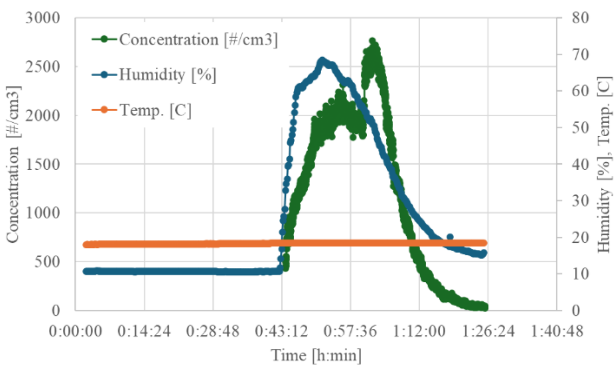

Fig. 6. The temperature, humidity and particle concentration profile for the measurement with the Schlick nozzle.

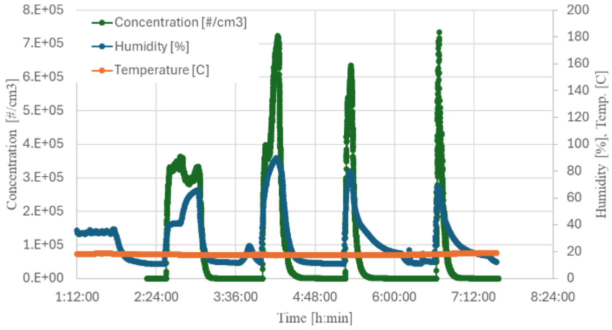

Fig. 7. The temperature, humidity and particle concentration profile for the measurement with the nebulizer.

In Fig. 6. the measurement with a nebulizer is presented. This measurement can be presented as the most suitable setting with the greatest possible variability of the resulting aerosol. The actual measurement can be performed by the following actions. At 1h43min, the FAGU flushing started. This was reflected in a decrease in the relative humidity of the outlet air. The air into the FAGU is first compressed in a compressor and then fed into the device through filters, therefore its relative humidity is lower than the humidity of the air in the laboratory. At 2:15, the CPC recording started, the concentration of impurities in the air was in the order of units per cubic centimeter. At 2:30, the nebulizer started, which ran for 30 minutes and then was turned off. While the nebulizer was running, the number of detected particles increased significantly to the level of 3.10^{5}#/cm^{3}. The relative humidity of the outlet gas increased to the level of 55%. After turning off the nebulizer, both the detected particles decreased to almost zero and the relative humidity in the air dropped to the original value before starting the nebulizer. The test had a similar course at 4:01, but the number of detected particles was higher at 7.10^{5}#/cm^{3}. This is probably related to the influence of the amount of liquid in the nebulizer reservoir. Based on this assumption, another experiment was performed that monitored the complete emptying of the nebulizer container. This experiment also proceeded similarly to the previous two, the only difference was in a slower decrease in relative humidity. This is caused by the additional air that the nebulizer consumes for its own operation. This air is also led through a HEPA filter so as not to contaminate the internal part of the FAGU. The last test at 6:37 was performed in a similar way, with the difference that the bath temperature was increased to 30C. However, this change had no visible effect on the result.

5 Discussion

Preliminary results from test measurements show very good properties of the device close to the planned ones. Replacing the nozzle with a nebulizer proved to be very suitable. Several nozzles were tested during the work. Single-phase ones had good properties for practical use, but in operation they proved to be very sensitive to clogging and the flow rate through the nozzle would be relatively large, which requires additional screens to drain excess water. Two-phase nozzles atomized the liquid better, but they bring practical problems with the mass balance of the air supplied. A nebulizer that produces a fine aerosol with an adequate amount of liquid and thus simplifies the regulation of the properties of the desired output mixture appears to be ideal. The amount of the air supply was solved by the calibration of the air flow. The nebulizer uses a volumetric displacement pump, the approach with a calibration is acceptable.

6 Summary

The results presented in this paper represent selected examples from the testing campaign of the FAGU. In the near future, the device will be integrated with an expansion chamber to facilitate detailed studies of nucleation processes. In parallel with the measurements at FAGU, it is necessary to verify the distribution functions of the produced droplets of both the nebulizer and the nozzle. For these measurements, the authors are equipped with several optical measuring devices [4].

We gratefully acknowledge the support Grant Agency of the Czech Republic Grant No. 22-28869S.

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