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DOES THE FOREST ECHOE THE SAME YOU SHOUT AT IT? (AN APPLICATION OF A CONTROLLED WAVE REFLECTION TO NUCLEATION RESEARCH)

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Summary: *(The title is a paraphrase of a popular Czech proverb). This contribution concerns an important aspect of the experimental investigation of the droplet nucleation kinetics using a shock tube (ST). For the measurement of the nucleation and growth rates it is necessary to shape the time-course of the supersaturation of the vapor (at time scales of 10^{-5}s to 10^{-2}s). This may be achieved by shaping the ST: sudden or continuous changes of the tube cross section are considered. After opening of the diaphragm separating the high- and low- pressure sections, an expansion wave runs into the high-pressure section and it reflects from its end, again as an expansion wave. A shock-wave runs into the low-pressure section. The high-pressure section is filled with a mixture of a vapor and a carrier gas (typically He or Ar). After the passage of the expansion wave and its reflection, adiabatic cooling takes place in the low-pressure section, leading to a supersaturation of the mixture. From the fluid-dynamics point of view we deal with a quasi-one-dimensional non-stationary flow of a compressible fluid, including dissipative processes. In the moment of the diaphragm opening, we have two waves with a Heavyside profile, mowing ahead and back from the diaphragm. However, the "echo" from the shaped tube can have quite a different form. In other laboratories, two qualitatively different shapes of the ST have been developed, suitable for the investigation of the droplet formation kinetics. A third shape have been suggested, computed and sucessfully tested by the present laboratory. The future development of the experiment, directed mainly on atmospherically relevant processes, will require a different shapes of the pressure (and, hence, supersaturation) evolution. Therefore, we prepare a solution of an inverse problem of determination numerically the required shape of the low-pressure section of the shock-tube in order that required pressure signal is obtained at the place of measurement. As another innovation we are developing a "shock-wave trap" - a fast acting valve which separates the high-pressure section from the low pressure section in order to prevent return of the shock wave, limiting the droplet growth period.*

1. INTRODUCTION

The title is a paraphrase of a popular Czech proverb "Jak se do lesa volá, tak se z lesa ozývá.", converted into a question. The shout is a shock wave with a decaying tail running through the low-pressure section of a shock tube after rupture of the diaphragm, separating the high- and low-pressure sections. If the low-pressure section has a constant cross section and a flat end-plate, a listener at the diaphragm location hears the same "shout" echoed. However, modifying the cross section along the tube axis and/or adding local hydrodynamic resistors allows creating an almost arbitrary shape of the echoed pressure signal. This fact is used to implement experimentally the method of nucleation pulse

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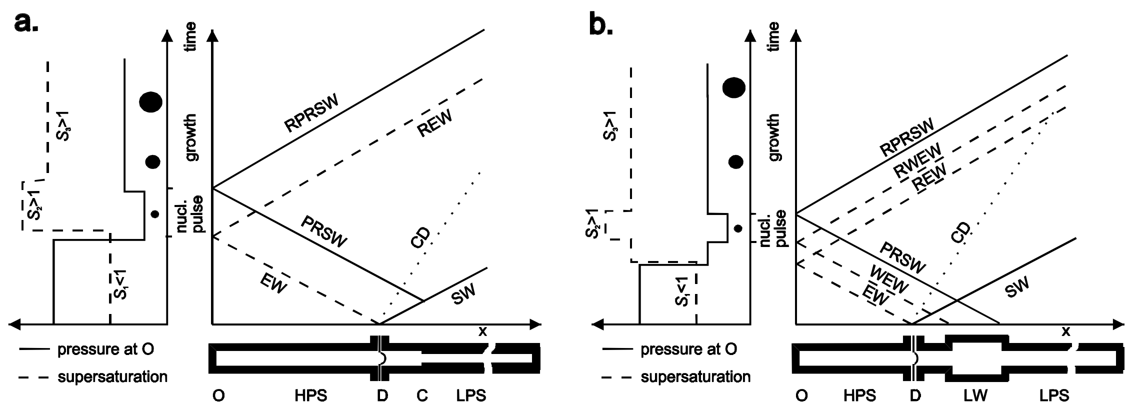
to study kinetics of homogeneous nucleation and growth of droplets in vapors. The value of the nucleation pulse method is in de-coupling the nucleation and growth processes, so that their rates can be determined independently, without additional assumptions. We present and original technique to generate the so-called nucleation pulse with a shock tube. The method has been tested successfully and is particularly suitable for investigation of rapidly growing droplets (i.e. for water vapor above the triple point.

The nucleation pulse technique is perhaps the most fundamental experimental approach to study droplet nucleation and growth. In general, the nucleation pulse is a short period of time, during which the system is so supersaturated that significant homogeneous nucleation takes place. However, the newly born nuclei are too small to be observed directly. Therefore, the nucleation pulse is followed by a long plateau, during which the system is still somewhat supersaturated, so that existing nuclei can grow, but the supersaturation is too low for the nucleation process to continue. The allowable duration of the nucleation pulse and the slope and roundness of its limits (in a supersaturation vs. time plot) are determined by the characteristic times of the growth. If the pulse is too long, the droplet distribution is too broad (polydisperse). Moreover, thermodynamic conditions can vary during the pulse (depletion and heating due to condensation of earlier nucleated droplets). Expansion cloud chambers use adiabatic expansion followed by a small re-compression to generate the nucleation pulse. At elevated temperatures (we consider temperatures about between 0 and 100°C), the growth of water droplets becomes very fast. The main reason is the exponentially increasing saturated vapor pressure. Under such conditions, the nucleation pulse must be rather short and have sharp limits. Devices generating the nucleation pulse based on gas-dynamical principles (like the shock tubes) avoid the necessarily slow moving mechanical parts and are, therefore, more suitable for the higher temperature range.



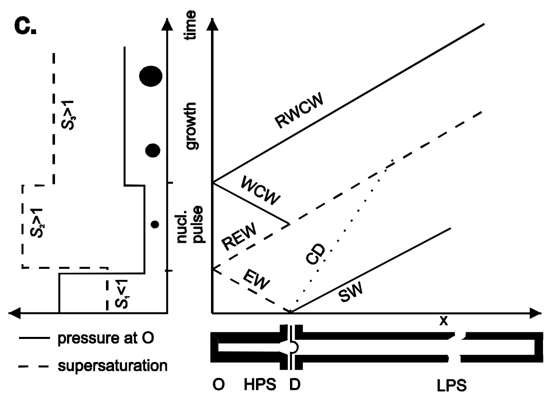


FIGURE 1. Three configurations of a shock tube to generate the nucleation pulse using
a) a constraint C in the LPS (4)
b) a local widening LW of the LPS (5)
c) larger cross section of the LPS (this work)
HPS/HPS…High/Low Pressure Section;
O…Observation place; D…Diaphragm;
CD…Contact Discontinuity;
(P)(R)CW/EW/SW…(Partially)(Reflected) Compression /Expansion /Shock Wave;
RPRSW Reflected PRSW;
(R)WCW/WEW...(Reflected) Weak CW/EW.

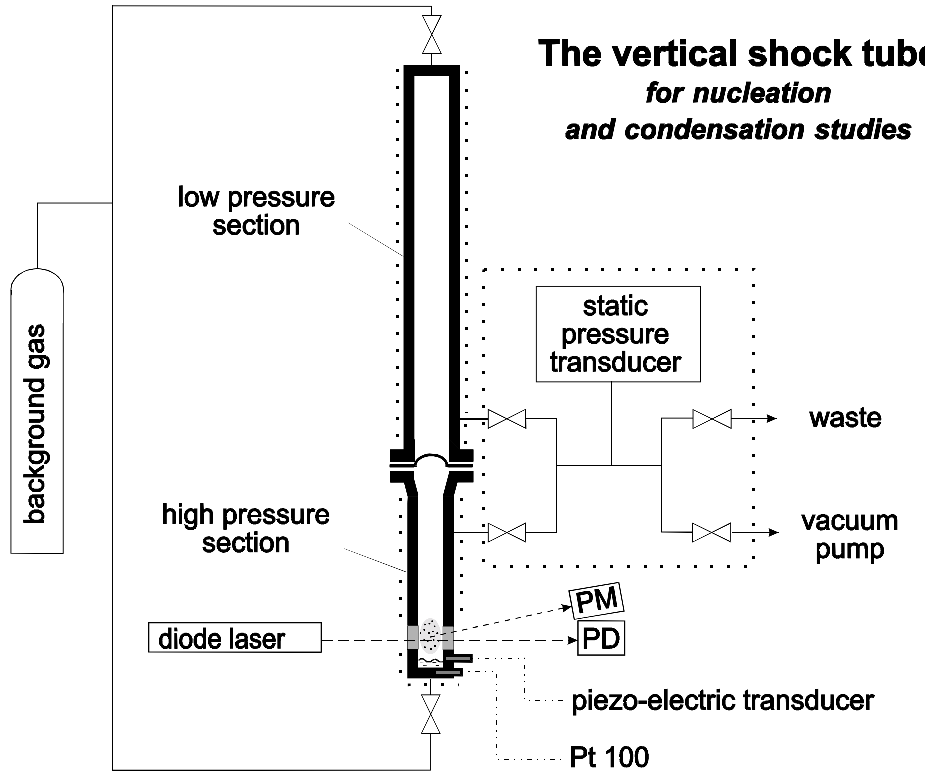


FIGURE 2. Scheme of the vertical shock tube developed for nucleation and condensation studies.

2. A NEW METHOD TO GENERATE THE NUCLEATION PULSE

For cloud chambers, the nucleation pulse was implemented by Schmitt (1) and Wagner and Strey (2). The last device has been continuously improved (2,3). The cloud chamber method can be considered quasi-static, since a sound wave caused by action of a piston or opening/closing a valve travels many times through the chamber and always brings only a very small change of pressure and temperature. The gas-dynamic based methods, on the other hand, rely on a combination of solitary waves propagating through a quasi-one dimensional tube after a thin diaphragm ruptures. The speed, magnitude and shape of these waves can be predicted with a good accuracy mathematically, starting from the initial conditions of the experiment and the tube geometry.

The first method to generate the nucleation pulse using a shock tube was invented by Peters (4, see Fig.1a). The vapor-background gas mixture is located in the high-pressure section (HPS) of a shock tube. Before the expansion, a thin diaphragm separates the high and low pressure sections (HPS and LPS). After the diapragm ruptures, an expansion wave propagates into the HPS, causing adiabatic cooling of the mixture and establishing supersaturated conditions. The observation place (O) is located near the bottom plate of the shock tube. Here the expansion wave reflects. The shock wave runs from the diaphragm place into the low-pressure section (LPS) and reflects partially on a constraint C. The partially reflected shock wave (PRSW), and its reflection (RPRSW), form the right edge of the nucleation pulse.

The Eindhoven group (5, see Fig.1b), working in the high-pressure range, developed a different technique. Here both the start and the end of the nucleation pulse are generated by reflections of the shock wave on a local widening of the LPS.

In Prague we have developed still another technique (see Fig.1c). The LPS has somewhat smaller cross section than the HPS. The nucleation is terminated by a weak compression wave (WCW, not a shock wave). WCW originates as a partial reflection of the reflected expansion wave (REW) at the place of broadening.

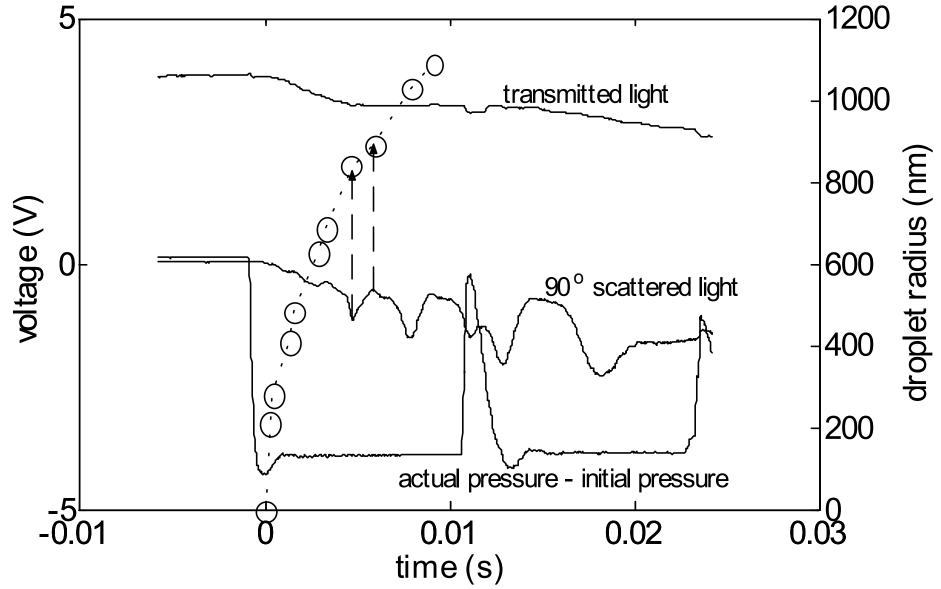


FIGURE 3. An example of pressure scattered and transmitted light signals in electrical units (water-argon, 265 K / 200 kPa nucleation temperature/pressure), combined with time-resolved mean radius of the droplets, evaluated using the Mie-Lorenz theory of light scattering. The first Mie-peak and first valley are almost unreadable because of the polydispersity.

3. The Apparatus

The high-pressure section (HPS) of our shock-tube is very short, some 180 mm, and it has an internal diameter of 46 mm. Yet the flow pattern is to a good approximation one-dimensional and it can be predicted using a simple mathematical model. The advantage of the shortness is that the cooling rate is very high (cooling of 50 K is achieved within about 0.3 ms with argon and even quicker with helium). The shock tube is oriented vertically (see Fig.2). This enables that a shallow liquid pool can be located at the bottom of the HPS. The HPS is thermostated using a circulation thermostat. The LPS is thermostated using electric heaters to prevent condensation of the liquid near the junction of the LPS and HPS. These facts enable that vapor-liquid equilibrium can establish in the HPS. After this equilibrium is established, the thin plastic diaphragm is punctured and it opens fully within a few microseconds. Near the bottom of the HPS, three windows are located, enabling illumination with a linearly polarized diode laser (red, 35 mW) and measurement of the transmitted and 90° scattered light using a photodiode and photomultiplier, respectively.

4. Results

Figure 3 shows an example of the signals recorded for an experiment with the water-argon system. The pressure signal shows peaks stemming from the reflections of the shock wave on the LPS end. Although the temperature is only about 265 K, the peaks and valleys of the scattered light signal become shallow and smooth, showing that the droplet cloud is significantly polydisperse. In fact, the 90° scattering was not found particularly suitable for higher temperatures. The reason is that the Mie-peaks at this angle are narrow-spaced and the pattern is very sensitive to polydispersity.

Preliminary results show that rather high cooling rates and sharp falling edge of the nucleation pulse can be achieved with the present method, enabling a well-defined start of the nucleation process. Although the nucleation pulse can be shortened down to a few tenth of millisecond, it is necessary to count with a polydisperse cloud of droplets. Therefore, we are currently improving the optical measuring methods in order to overcome this problem.

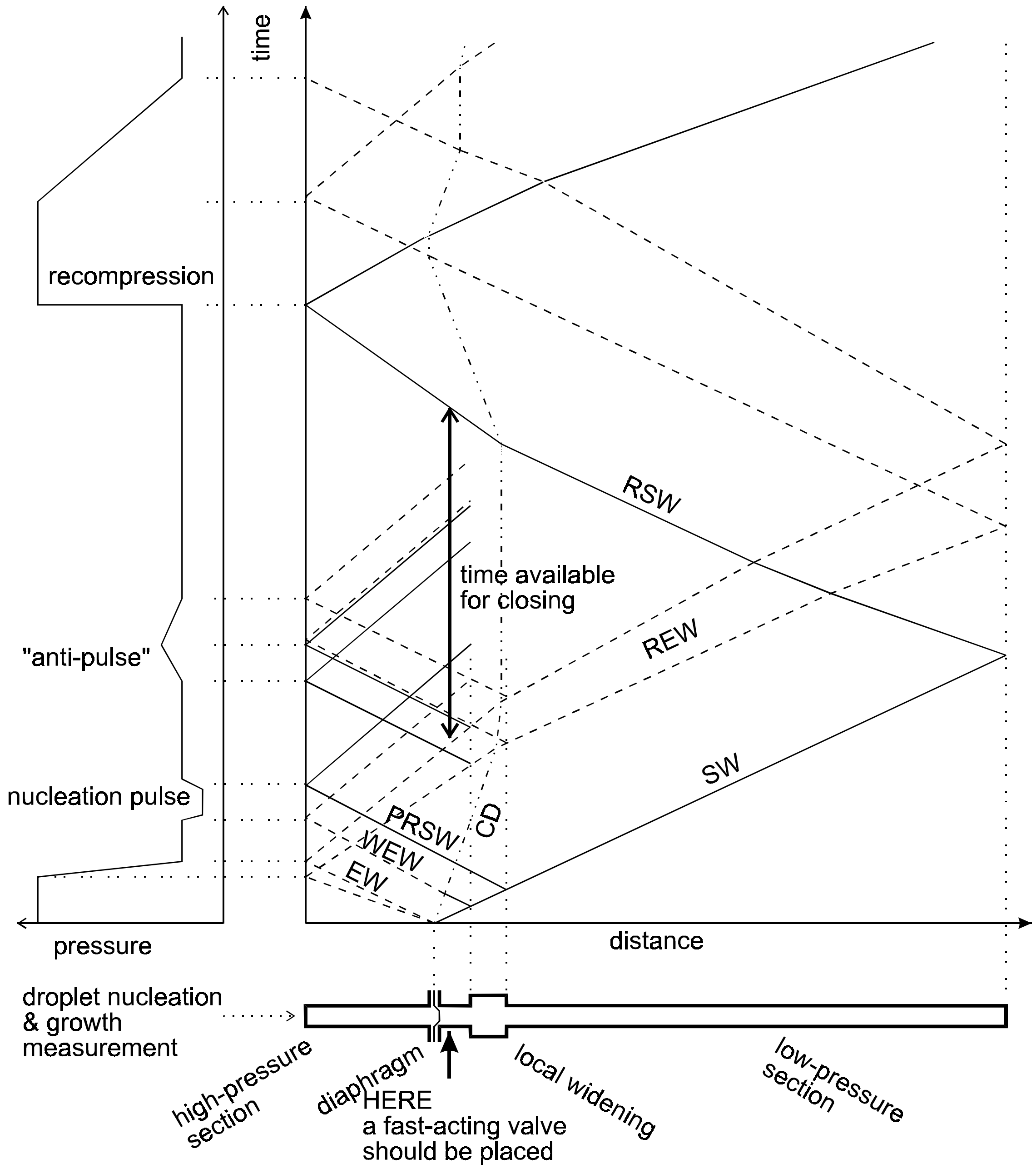


FIGURE 4. Non-idealities of the pressure evolution at the end of the high-pressure section. The tube design is taken over from Looimans and van Dongen [6]. We suggested an improvement to this design. The small hump, the "anti-pulse" disturbs somewhat, but it can be tolerated. Decisive is the recompression by the reflected shock wave (RSW). A possible remedy would be a fast acting valve, separating the high- and low- pressure section after the passage of the shock wave (SW) and reflected expansion wave (REW).

5. CLOSING REMARKS

In this paper we presented an extended presentation of the new technique to generate the nucleation pulse using a shock tube. This presentation provides more details than the original paper [8].

At present, we are preparing measurements of nucleation and growth rates in the mixture water, sulfuric acid and a carrier gas. These data is requested by climatologists and meteorologists, since it this process is assumed to be on of the main source of fine atmospheric aerosols. In order to be able to observe the droplets nucleated in this mixture, we need a long growth time for the droplets. However, with a shock-tube, the observation time is always limited by the impact of the reflected shock wave (see Fig.4). The available time is given by the time-of-flight of the shock wave from the diaphragm place to the low-pressure–section end-plate, and back to the low-pressure-section end-plate (the place of measurement). Hence, the limitation is basically by the length of the shock tube. Once the tube extends over the whole laboratory, other ways are to be looked for. What are the possibilities to eliminate this reflection? First we have to realize that not only the shock-wave, but also its decaying tail (the doubly-reflected expansion wave) have to be avoided. First we investigated passive method: the end of the high-pressure section would contain dissipative elements (e.g., orifices) and capacities (local widening), in a way not dissimilar to a silencer of an internal-combustion engine. Although numerical simulations have shown partial success of this idea, perfect damping of the waves seems very difficult. Instead, we considered an *active control* method: after the passage of the shock wave (SW) and the reflected expansion wave (REW), the high- and low-pressure section are separated again with a fast-closing valve. The time available for the valve closing is of the order of 0.01s, depending on the length of the shock tube and speed of sound of the carrier gas.

6. ACKNOWLEDGEMENT)

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