Experimental study on particle plugging in ultrafine capillaries

Mei Tian*, Xiaoyuan Han, Zongyu Gan, Yindong Wang and Ronghu Zou
Northwest Institute of Nuclear Technology, Xi’an, Shaanxi, 710024, China
*Corresponding author’s e-mail: tmhxy@sohu.com

Abstract. A practical procedure was used to study the plugging process of particles in ultrafine capillaries with bore sizes ranging from 10 to 20 μm and lengths from 10 mm to 40 mm under pressure differences from 60 to 400 kPa. The results show that the particle plugging in capillary is a popular phenomenon and plugging process is of great uncertainties. The ultrafine capillary could be plugged completely under a certain pressure difference after a long period aerosol exposure, and the plugging position was diagnosed to be at or near the leak path entrance. It was believed that pressure difference might play a special role in plugging situation as well as compactness of the deposit. In many cases, the deposit was not firm enough, and could be re-suspended. The possibility of complete plugging decreases gradually with the increase of the capillary bore, but partly plugging and unplugging of the capillary may occur repeatedly.

1. Introduction
Containment buildings act as barrier to the airborne transmission of radioactivity. A variety of tiny leak paths via shells or vessels would get rise to serious safety issues. Particle penetration and plugging through leaks has been investigated for a long time. Several early experimental works were carried out to study the particle penetration and plugging effects through capillaries with regular, well-defined geometry. Morewitz[1] provided a detailed review of these early works, which mostly focused on the correlation between the observed leakage rates or plugging times and experimental parameters. Williams[2] developed a theory of particle build-up on leak tube inner surfaces with time due to continuous deposition and some re-suspension, and compared the results from it with experiments of Nelson & Johnson[3], but the agreement between experiment and theory was not excellent, it was attributed to the uncertainties of the experiments and the assumptions in theory. Burton[4] and Morton[5] demonstrated the importance of pressure in regulating the rate at which the capillary was plugged by deposited particles. They observed that much of this deposition appeared to take place at the entrances of the capillaries. When the driving pressure is low, the leak-path appears likely to be plugged quickly. Luo XiaoWei[6] researched particle deposition in a laminar pipe flow, considering thermophoresis, gravity, lift force, diffusion, and convection. They employed a revised semi-implicit method (SIMPLER) to solve the aerosol transport equation, the concentration profile of particle at an arbitrary cross-section of the pipe was obtained for different deposition mechanisms, and the variations of particle deposition efficiency with particle size were given. A simplified mechanistic model for particle penetration and plugging in tubes and cracks was developed by Mitrakos et al.[7]. The model was verified against analytical solutions and validated by comparing with the experiments of Nelson & Johnson [3], Liu & Nazaroff[8] and so on.

When reviewing above researches, it was noted that few literatures experimentally investigated particle plugging in ultrafine leak paths, which is complex and of the great uncertainties. Capillary was
usually chosen as the model leak path to understand particle penetration behavior under worst-case conditions (e.g. straight leak-path). In this paper, particle plugging process in ultrafine capillary under pressure differences from 60kPa to 400kPa was obtained, and the plugging position was diagnosed for complete blocking capillary after aerosol exposure. It should be noted that the finds may be helpful in the treatment of aerosol leakage in a variety of nuclear or non-nuclear applications, especially with respect to the integrity of valves and fittings for the supply of particle-free gases and for the containment of particles in pressurized environments.

2. Experimental methodology

2.1 Experiment of aerosol exposure

A schematic diagram of the experimental apparatus is shown in figure 1. It consists of three parts: aerosol source, capillary subassembly and particle detector.

![Schematic diagram of experimental apparatus for aerosol exposure. All connection tubes are as short as possible.](image)

The smoke from a mosquito coil was used as the aerosol source in the experiment, the pressure in source container was controlled between 150 kPa and 500 kPa, and the capillaries were fixed horizontally. The number-size distribution of the aerosol source was measured by an electrical low-pressure impactor (ELPI, TSI, Inc.), which measures the airborne particle size distribution in the size range of 0.03-10 μm with 12 channels. The particle number concentration was monitored by an ultrafine condensation particle counter (UCPC, TSI, Inc., Model 3776), which is capable of counting particles larger than 2.5 nm in diameter. In order to reduce dilution, the sampling flow rate of 300 mL/min was chosen in the experiments. The detailed operating principle has been described elsewhere by Tian et al.[9]. In this study, the concentration of leaked particles from the target capillary was monitored in real time for a long time, usually more than 1 hours, and the measurement interval was 5 s. When no particle leaked from the target capillary any more, the exposure experiment was terminated and the gas leakage rate of the capillary after aerosol exposure would be measured.

2.2 Gas leakage testing

The gas leakage rate of a capillary after aerosol exposure was measured with the apparatus presented in figure 2. A capacitance diaphragm gauge (Inficon Inc., CDG025D) was employed to measure the vacuum pressure in the downstream vessel. The system is capable of detecting the gas leakage rates of values with a standardized leakage rate (SLR) as low as $5 \times 10^{-7}$ Pa·m³·s⁻¹.
Figure 2. Schematic diagram of the experimental apparatus for determining the capillary gas leakage rate

The flow in the capillary under the experimental conditions would nearly be in a viscous state, and the gas leakage rate ($L_a$) can be obtained by solving the following expression, which was derived from Poiseuille’s law for laminar flow (Poiseuille, see Lamb, H., 1945):

$$L_a = \frac{1}{2} (P_u + P_d) Q_v = \frac{\pi}{128} \frac{d^4 (p_u^2 - p_d^2)}{2\eta l_c}$$

Where, $P_u$ and $P_d$ are the pressure of the capillary entrance (upstream) and exit (downstream), respectively. $d_c$ and $l_c$ are the diameter and length of the capillary, respectively. $\eta$ is the gas viscosity (kg·m$^{-1}$·s$^{-1}$).

3. Results and discussion

3.1. Size distribution of aerosol source and deposition

The size distribution of aerosol generated by mosquito coil burning was measured with ELPI and the result was shown in figure 3. It can be seen that over 95% of mosquito-coil-smoke aerosol is less than 300 nm in size.

3.2. Particle plugging in ultrafine capillary

The aerosol concentration in the source container will decrease gradually due to various deposition mechanisms, such as particle diffusion, settling, and inertial deposition. The change curves of the aerosol concentration in the source container at pressures of 150 kPa, 200 kPa and 300 kPa are presented in figure 4. As shown, the aerosol concentrations have similar changes for different amounts of source pressure. If the concentration curve of the leaked particles is not consistent with the deposition curves showed in figure 4, even not smooth, it means that the plugging occurred.

Figure 3. Number-size distribution of mosquito-coil smoke aerosols

Figure 4. Change curves of the aerosol concentration in the source container at different pressures
The aerosol exposure experiments were undertaken using ultrafine capillaries with bore sizes ranging from 10 to 20 μm and lengths from 10 mm to 40 mm at upstream pressure in the range 150-500 kPa. It was found that many capillaries presented to be plugged partly or completely during test period, which indicates ultrafine capillary plugging was a popular phenomenon.

3.2.1 Complexity and uncertainty of the blockage
The plugging processes of three Φ20 μm × 10 mm capillaries in the similar pressure condition were shown in figure 5. The monitoring period of figure 5(a) was about 120 min. It can be seen that the curve of the capillary a was relatively smooth except occasional slight undulation and the penetration efficiency of particle through the capillary at the beginning and ending time of test was 99% and 100.3% respectively, which indicated that the plugging of the capillary was very slight. The experiment of figure 5(b) lasted 590 min, and the curve varied irregularly, sometimes the count of leaked particle rose or fell suddenly, this means that the capillary has been partly blocked or unblocked. It should be noted that plugging and unplugging of the capillary were happened alternately and the capillary was eventually completely blocked at about 526 min. A serial of surges in aerosol source pressure were tried and no aerosol leakage was found, the capillary was considered to be completed blocked; but the gas leakage rate of the capillary after aerosol exposure was tested is 1.92E-05 Pa·m³/s (P_a : 205.5 kPa, P_d : 35.28 Pa), only significantly less than the gas leakage rate 4.53E-04 Pa·m³/s before exposure, which meant that the capillary was only partly blocked. Perhaps the deposit was partly suspended again during the gas leakage rate measurement, which implied that the deposit was not compacted enough. This situation has been reappeared in many other experiments. figure 5(c) presented that the curve was smooth except a few fluctuation during the test period of 660 min, and was consistent with the curve of aerosol concentration in source container, which indicated that on serious plugging occurred in this capillary. It can be seen that the plugging situations of three same size capillaries were very different, which might be caused by small differences in inlet geometry of the capillaries; on the other hand, it is also believed that the capillary plugging has stochastic element to a certain extent.
Figure 5. Aerosol leakage through the capillaries with the size of Φ20 μm×10 mm under upstream pressure of about 200kPa

Sometimes the blockages were not so obvious on the curves. The concentration curves of the leaked aerosols through the Φ10μm×10mm capillaries under upstream pressure of 200 kPa and 300 kPa were showed in figure 6. The experiment of figure 6(a) lasted about 580min, and the curve was smooth and consecutive. It didn’t mean that there was no plugging. The penetration efficiency of aerosol in the capillary at the initial and end time were 44% and 83% respectively, which indicated that the plugging and the unplugging has happened, just because the deposition and re-suspension process was mild. Figure 6(b) presented that the curve was smooth during the first 120min, but then the complete plugging occurred rapidly; a surge in aerosol source pressure was tried and re-suspension of the deposit was found.

Figure 6. Aerosol leakage in capillaries with size of Φ10μm×10mm under 200kPa and 300kPa

Aerosol plugging process might be mild, particles deposit on the wall step by step till the capillary is plugged completely, such as figure 6(b); sometimes, plugging accumulates to some degree, deposited aerosols could be re-suspended, and the plugging process alternated with the purging process (see figure 5(b)) for a long time, and then complete plugging occurred or didn’t. Besides, the purging process of the deposit was often sudden and the leaked aerosol concentration increased sharply. It is worth noting that the size of the re-suspended matter may be much larger than the initial size before particle deposition and the re-entrained bulk could cause rapid blocking in an ultrafine capillary. The problems in this aspect involves a strongly stochastic element, which has been observed in experiments. As a typical case, figure 7(d) showed the sharp alternating between plugging and unplugging. It was also found in the experiments that the capillary was easily blocked completely when it had been used for aerosol exposure repeatedly.
3.2.2 Complete plugging of capillary

It was noted that some size capillaries occurred to be completely plugged under certain pressure. The plugging processes of some capillaries fully blocked were presented in figure 7. Figure 7(a), (b) and (c) showed plugging situation of three capillaries with same size of $\Phi 10\mu m \times 10mm$, and complete plugging occurred about 40–200min after aerosol exposure. For the capillaries with inner diameter of 15 $\mu m$ and 20 $\mu m$, complete plugging also happened after a long period of aerosol exposure (showed in figure 7(d) and (e)). Once the capillary was fully plugged, a serial of surges in aerosol source pressure were tried in order to unblock the plugged capillary, but no further penetration was observed. Then the capillaries were carefully removed from the facility after aerosol exposure, and re-mounted in the device (showed in figure 2) to obtain the post-test gas leakage rate of capillary.

The gas leakage rates for some fully plugged capillaries before and after aerosol exposure were listed in Tab.1. It was clear that the gas leakage rates for fully plugged capillary were far lower than before aerosol exposure. Furthermore, the plugged capillaries could not be unblocked by surge of upstream pressure from 200kPa to 500kPa. More attempts were performed about the capillary with size of $\Phi 15\mu m \times 40mm$. But no matter the capillary was fitted in positive direction and opposite direction, the gas leakage rates under different pressures were all lower than $5.0 \times 10^{-7} Pa \cdot m^3/s$; after the capillary was cut out 4.8mm in the entrance, the gas leakage rate was consistent with predicted value by formula 2.2,
Figure 7.plugging process of some capillaries fully blocked under about 500kPa
It implied that the plugging position should be at or near the leak path entrance. A reasonable explanation is the particles had deposited at the capillary entrance where the direction of gas flow would change. It seemed that these deposits were compacted enough, and so the gas leakage rates of the capillaries decreased significantly.

| Serial No. | Capillary size                                      | Pressure(Pa)** | Gas leakage rate (Pa·m³/s) |
|------------|-----------------------------------------------------|----------------|----------------------------|
| AIM46      | Φ10μm×10mm                                          | 101500         | 46.12 6.91×10⁻⁶ <5.0×10⁻⁷**** |
|            |                                                     | 503300         | 46.99 1.70×10⁻⁴ <5.0×10⁻⁷    |
| AIM89      | Φ10μm×10mm                                          | 102000         | 52.37 6.97×10⁻⁶ <5.0×10⁻⁷    |
|            |                                                     | 200000         | 54.32 2.68×10⁻⁵ <5.0×10⁻⁷    |
| AIM138     | Φ10μm×10mm                                          | 200400         | 31.91 2.69×10⁻⁵ <5.0×10⁻⁷    |
|            |                                                     | 353000         | 38.02 8.35×10⁻⁵ <5.0×10⁻⁷    |
| AIM123     | Φ15μm×40mm* (positive direction fitted)             | 100100         | 40.82 8.50×10⁻⁶ <5.0×10⁻⁷    |
|            |                                                     | 300000         | 41.33 7.63×10⁻⁵ <5.0×10⁻⁷    |
|            | Φ15μm×40mm (opposite direction fitted)              | 100100         | 40.37 8.50×10⁻⁶ <5.0×10⁻⁷    |
|            |                                                     | 299700         | 40.80 7.62×10⁻⁵ <5.0×10⁻⁷    |
|            | Φ15μm×35.2mm (Capillary entrance was cut 4.8mm)     | 100300         | 45.24 9.70×10⁻⁶ 9.70×10⁻⁶    |
|            |                                                     | 300200         | 55.01 8.69×10⁻⁵ 8.38×10⁻⁵    |
| AIM191     | Φ20μm×20mm                                          | 213200         | 46.33 2.44E-04 <5.0×10⁻⁷    |
|            |                                                     | 323700         | 47.36 5.62E-04 <5.0×10⁻⁷    |

* Positive direction means that pressure flow direction were identical in the measurement of aerosol leakage and gas leakage rate;
**: The gas leakage rate of capillary after aerosol exposure was tested under this pressure condition.
***: The gas leakage rate of capillary before aerosol exposure was calculated with formula 2.2;
****: Detection limit of the method for gas leakage rate with the device showed in figure 2 is 5.0×10⁻⁷ Pa·m³/s.

Some capillaries with size of Φ10μm×10mm were tested under Pd 200kPa and 300kPa, the results showed that the blockages were not so serious, sometimes the complete blockage occurred too (see figure 6), but it seemed that the deposit was more likely to be re-suspended. It was believed that the pressure difference might play a special role in plugging situation. Maybe when the upstream pressure was about 500kPa for Φ10μm×10mm capillary, the gas flow rate is more convenient to particles deposition and the deposit was tight enough so that the particulate couldn’t re-suspended due to strongly adhesive forces.

3.2.3 Influence of the capillary dimension on plugging
With the increase of capillary diameter, the possibility of complete blockage for fine capillary decreases
gradually. Figure 8 showed the typical concentration curve of leaked particles from the capillaries with I.D. 10 μm~20 μm and 10mm long under $P_2$ 300~315kPa. It could be seen that the complete plugging occurred at about 250min for $\Phi10\mu m \times 10mm$ capillary (see figure 8(a)), and about 808min for $\Phi15\mu m \times 10mm$ capillary (see figure 8(b)). For $\Phi20\mu m \times 10mm$ capillary, the aerosol exposure experiment lasted about 64 hours and no complete blockage occurred; figure 8(c) just presented first 950min, it could be found that partly plugging and unplugging happened again and again. Figure 9 showed the plugging process of the capillary with 50μm bore, and it is clear that partly plugging occurred, but the plugging was not serious and deposited aerosols were re-suspended easily. Under same pressure condition, the gas leakage rate of the capillary with larger bore would increase obviously and the capability of gas flow re-suspending aerosol is enhanced, so complete blockage didn’t occurred.

Figure 8. Plugging process of the capillaries with bore from 10 μm to 20 μm under similar pressure difference
Figure 9. Aerosol leakage through the capillary with the size of Φ50μm×9mm under P₀ 200kPa

4. Conclusion
The experiments proved that the plugging of particles in the ultrafine capillary was a popular phenomenon. Capillary plugging situation may be affected by pressure difference, capillary size and entrance shape, and so on. Plugging process is very complex and of great uncertainties, even for same size capillaries in similar pressure condition. After a long period aerosol exposure, the ultrafine capillary can be plugged completely at a certain pressure difference, and the blockage is diagnosed to be at or near the leak path entrance. It is believed that pressure difference might play a special role in plugging situation as well as compactness of the deposit. In many cases, the deposit can be re-suspended. The possibility of complete blocking decreases gradually with the increase of the capillary bore. For capillary bore larger than 20μm, no complete blocking was observed, but partly plugging and unplugging happened repeatedly. More quantitative research is not yet available in our laboratory. In addition, the effect of particle size on the plugging needs further study.

The capillary tube is just an ideal leakage path with regular, well-defined geometry, the actual leaks may occur in various forms, such as thin irregular cracks, non-circular-shaped orifices, annular passages, and labyrinthine paths through mechanical packing. As a straight leak path, the experimental results demonstrated the most conservative blockages of ultrafine leak paths. The irregular leaks would be more advantageous to aerosol deposition. Complete plugging of micro leak path has positive significance to the safety of unclear containment or sealing of non-nuclear device and parts.

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