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Development of a high-speed bioaerosol elimination system for treatment of indoor air

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1. Introduction

In the winter of 2019, an outbreak of coronavirus disease 2019 (COVID-19) caused by severe acute respiratory syndrome coronavirus 2 (SARS-CoV-2) was confirmed in China. It spread worldwide the following year, causing severe damage to human health and the global economy. Subsequently, it was declared a pandemic by the World Health Organization [1–4]. COVID-19 is primarily transmitted through aerosols [5–10], and indoor air ventilation is recommended as a tentative countermeasure. However, indoor air ventilation has also resulted in increased heating and cooling costs in current air-conditioned societies [11].

SARS-CoV-2 and influenza viruses spread through human sneezing, coughing, and conversation [12], and the time required for the descent of these bioaerosols varies depending on their size. Aerosols in the size range of 1–10 μm, which is the size distribution of infectious aerosols, remain in the air for a long time [5,6,8,13,14]. By contrast, smaller particles generally dry out faster, resulting in inactivation due to the increased intraparticle salt concentration [15]. On the other hand, the rate of descent for particles larger than 10 μm is higher, reducing the likelihood of transmission via respiratory inhalation. The risk of viral infection can be reduced if the absolute amount of bioaerosols is reduced while they drift in air. Currently, high-efficiency particulate absorbing (HEPA) filters are used to treat viruses in indoor air [16–19]. However, in the case of HEPA filters, air passes through a submicron-sized mesh, resulting in a significant pressure drop and increased energy consumption [20]. Additionally, HEPA filters and prefilters for protection from dust and other contaminants must be replaced more frequently because they accumulate dust and become breeding grounds for bacteria and viruses [16,21]. Another possible countermeasure for COVID-19 is the continuous introduction of aerosols. Because this system exhibits excellent aerosol removal ability at a flow velocity of 5 m/s or higher, it is more suitable than other reactive air purification systems for treating large-volume spaces.

Keywords:
SARS-CoV-2
TiO2 photocatalyst
Bioaerosol
Windspeed
Indoor air

Abbreviations: COVID-19, coronavirus disease 2019; SARS-CoV-2, severe acute respiratory syndrome coronavirus 2; HEPA, high-efficiency particulate absorbing; UVA, ultraviolet-A; UVC, ultraviolet-C; AOP, advanced oxidation process; CFD, computational fluid dynamics; LES, Large eddy simulation; DES, detached eddy simulation; RANS, Reynolds-averaged Navier–Stokes; ISO, International Standard Organization; SCDLP, soya casein-digested lecithin polysorbate; UV, ultraviolet.

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aerosols is the use of ultraviolet-C (UVC), which damages viral RNA [22, 23] and can inactivate viruses contained in aerosols. However, for the efficient operation of UVC reactors, the flow rate must be controlled to irradiate UVC against the virus for the time necessary for complete treatment [20,22]. Other reactive treatment systems, including photocatalytic systems (advanced oxidation process, AOP), also require the inhaled virus to remain in the elimination system for a sufficiently long time for inactivation.

Table 1 presents the categories of bioaerosol removal and inactivation technologies. Our developed bioaerosol removal system follows a mechanism that effectively utilizes the kinetic energy of aerosol particles in high-speed air currents to treat aerosols, it is completely different from the existing technologies listed in the table, except for the use of photocatalysis. In other words, this system can be considered a novel bioaerosol removal technology.

For the equipment to be suitable for aerosol treatment, its performance must be equivalent to that of ventilation. Article 20 of the Building Standard Law of Japan specifies the required ventilation volume using the following formula [59]:

\[
\text{Required ventilation volume} = 20 \times \text{floor area of the room (m}^2\text{)/occupied area per person (m}^2\text{)}.
\] (1)

The occupied area per person is given by a coefficient that has a different value depending on the building classification. In terms of the total building floor space in Japan, schools ranked third (19.6%, 372 million m²) after offices (25.7%) and wholesale/retail spaces (24.8%) [60]. However, of these three, people spend the most time under high-population-density conditions in schools [61,62]. The relationship between COVID-19 and the school is evident in the tens of thousands of references using both of these keywords in scholarly articles in the search results on the Web of Science (www.webofscience.com), which indicate a high level of interest [63–72]. If we calculate the required ventilation volume for an average-sized elementary school classroom (72 m², less than 300 m³) in Japan, assuming 30 schoolchildren in the classroom, the required ventilation volume would be 600 m³/h. However, if an aerosol treatment system is included, it is not necessary to treat the entire volume. If the room is completely sealed, it cannot emit CO₂. Therefore, only half of this amount (300 m³/h) needs to be treated, assuming ventilation is used in conjunction. This is based on the ventilation rate of 0.5 times/h specified in Article 20.8 of the Building Standard Law of Japan [59]. The next issue would be determining the installation location of the device in the room. Several studies have shown that if the equipment is installed at an incorrect location, it would not function appropriately [16,17,73]. However, because performing a demonstration test in a large space is difficult, previous studies on the simulation of aerosol diffusion and processing by fluid analysis are highly useful [74,75].

Virus elimination using photocatalysis, which is representative of the AOP, has been studied extensively, and the photocatalytic treatment of bacteriophages has been established by the International Standard Organization (ISO) [76–78]. Reports on the photocatalytic elimination of SARS-CoV-2 are also rapidly increasing [53–58]; however, the inactivation of viruses is impossible unless bioaerosols are captured on the photocatalytic surface. Although several photocatalytic air purifiers are commercially available, most are photocatalyst-coated filter. A small group of products also exists that purifies air by passing airflow along the surface of the photocatalytic material [54]. However, selectively capturing and treating aerosols on photocatalytic surfaces using current photocatalytic filters or surface pass systems is difficult.

The size of bioaerosols is significantly larger than that of gas molecules (μm vs. Å). Therefore, when a high-velocity airflow containing aerosols flows through an elbow duct structure, large-mass aerosols cannot turn and collide with the elbow walls. If the number of elbows increases, the aerosols can be treated in stages. The captured virus can be inactivated and mineralized by photocatalysis if the duct wall is a

| Technology                              | References   | Note                                    |
|-----------------------------------------|--------------|-----------------------------------------|
| Elimination of bioaerosol               |              |                                         |
| HEPA Filter                             | Ruth 1935 [24] | Decrease in air supply due to continuous use |
|                                        | Olmsted 2008 [25] |                                       |
|                                        | Ryan 2010 [26] | Energy loss due to filter blockage      |
|                                        | Loderer 2012 [27] | Decrease in air supply due to continuous use |
|                                        | Hubbart 2012 [28] | Decrease in air supply due to continuous use |
|                                        | Lee 2015 [29] | Decrease in air supply due to continuous use |
|                                        | Mounavi 2020 [17] |                                       |
|                                        | Christopherson 2020 [18] |                                   |
|                                        | Nazarenko 2020 [20] |                                       |
|                                        | Zacharias 2021 [19] |                                       |
|                                        | Brousers 2021 [20] |                                       |
|                                        | Mounavi 2021 [21] | Decrease in air supply due to continuous use |
|                                        | Myers 2022 [31] |                                       |
| Acoustic agglomeration                  | Nelson 2013 [32] |                                       |
| Adsorption (Electrostatic)              | Volken 2002 [33] |                                       |
|                                        | Hong 2016 [34] |                                       |
|                                        | Jung 2020 [35] |                                       |
|                                        | Feng 2021 [36] | Water vapor reduces electrostatic adsorption |
| Adsorption (Adsorbent)                  | Li 2011 [37] |                                       |
|                                        | Pham 2015 [38] |                                       |
| Inactivation of bioaerosol              | UVC Kim 2018 [39] | UVC-LED                               |
|                                        | Welch 2018 [40] | Laser                                  |
|                                        | Lo 2021 [22] | Hg Lamp                                |
|                                        | Bono 2021 [23] |                                       |
|                                        | Muramoto 2021 [41] | UVC-LED                               |
|                                        | Herman 2021 [42] | Sunlight                                |
|                                        | Lai 2021 [43] | UVC-LED                                |
| O₂                                      | Masuda 1993 [44] |                                       |
|                                        | Gränshpun 2007 [45] | MS-2                                  |
|                                        | Kim 2018 [46] | MS-2                                   |
|                                        | Tiaossi 2020 [47] |                                       |
|                                        | Vyskocil 2020 [48] | Phage PhiX174 (HEP-036)               |
|                                        | Park 2020 [49] | Escherichia coli and Micrococcus luteus |
| Photocatalyst                           | Ishiguro2011 [50] | Qβ and T4 bacteriophages               |
|                                        | Daikoku 2015 [51] | Influenza virus                        |

(continued on next page)
photocatalyst [23,79,80]. However, an increase in the number of elbows (construction of multistage elbows) results in an increase in the pressure drop. Although the pressure drop can be reduced by reducing the flow velocity, the aerosol processing capability, which is an advantage of the elbow structure, is not expressed. In other words, obtaining the optimum solution for the number of elbow stages and wind speed is a significant challenge in the development of this system. Another difference between this multistage elbow structure and other reactive air purification systems is that they do not have a pre-filter to remove dust and other particles. Pre-filtering can reduce system performance owing to an increased pressure drop and result in uneconomical filter replacements [16,20,21]. In addition to bioaerosols, inhaled dust also collides with walls. However, if the body size is large (e.g., fiber fragments) compared to aerosols, even if it collides with the photocatalyst, it has high air resistance owing to its large size and is eventually discharged toward the discharge side by airflow. Fine particles, such as PM2.5, collide and adsorb onto the photocatalyst surface. If the particles are organic compounds, they eventually decompose into CO$_2$ via photocatalytic decomposition [81]. If they are inorganic particles, they remain on the photocatalytic surface; however, the presence of inorganic particles has little effect on the photocatalytic reaction.

Recently, UV-LEDs with wavelengths reaching the ultraviolet region have been commercialized and used for sterilization applications [20, 22]. Many UV-LEDs have sufficient intensity for photocatalytic excitation, and research on photocatalytic disinfection using UV-LEDs is in progress [81,82]. Compared with conventional black light bulb (BLB),
A mercury lamps, or cold cathode tubes, UV-LEDs have the advantage of an extremely small light-emitting element, and the design of the photocatalytic reaction system is not restricted by the shape of the light source (besides it has a long life of over 50,000 h for UVA-LED). UVA (near-UV) to UVC (deep-UV) LEDs have already been commercialized and are readily available; however, their price tends to be higher for shorter wavelengths. Although TiO2 can be excited in any wavelength range from UVA to UVC, UVA-LEDs with a central wavelength of 365 nm provide more than sufficient catalytic activity. Therefore, the requirement of expensive UVC LEDs is eliminated, which is a major economic advantage.

Thus, we developed a system and evaluated its performance to reduce the risk of infection. Rapidly treating bioaerosols floating in a large volume of indoor air, such as a classroom, is achieved by impinging on a photocatalyst in a high-speed air stream, inactivating the viruses by evaporating the aerosols, and finally photocatalytically mineralizing them with the UVA-LED irradiation.

2. Experimental methods

2.1. Design and fabrication of the multistage elbow photocatalytic reactor

The reaction device was designed to be portable indoors. The elbow duct structure was constructed from eight alternately connected navicular structures (Fig. 1A) fabricated from half-regular hexagonal columns with lengths of 302 and 40 mm on a side created from stainless steel (Fig. 1B). A 300 × 38 mm thermally sprayed photocatalytically coated aluminum plate (Kamaishi Electric Factory, Iwate, Japan) was fixed to the inner three sides of the navicular and elbow duct units (Fig. 1A). Each navicular elbow unit was combined with a UVA-LED unit (λ = 365 nm, JCN-FN60EUV-W-12, JCNet, Osaka, Japan) to irradiate the photocatalyst with excitation light on the inner three sides of the navicular elbow duct unit (Fig. 1A). The cross section of the system is shown in Fig. 1C. A low-temperature line heater (SRC1053, Hakko Electric Co., Ltd., Nagano, Japan) was also installed in this multistage elbow unit to prevent the photocatalytic activity from decreasing because of the high-humidity environment. Three fans were placed at the discharge side of the multistage elbow unit to draw air-containing aerosols into the unit by negative pressure. Because the fan used (MU12255-11D, Oriental Motor Co., Tokyo, Japan) was an axial flow fan, the rotational flows from the fans interfered with each other on the discharge side, and the exhaust speed decreased if three of the fans were arranged in parallel. Therefore, partition plates were placed on the fan discharge side to prevent mutual interference (Fig. 1D). Based on this design, we constructed the system shown in Fig. 1E. Hereafter, the unit consisting of this elbow duct structure will be referred to as the multistage elbow unit, and the aerosol processing equipment consisting of this unit will be referred to as the multistage elbow system.

The airflow conditions inside the multistage elbow were simulated using computational fluid dynamics (CFD) with a three-dimensional (3D) model of the multistage elbow unit modeled in Fusion 360 (Autodesk Inc., San Rafael, California USA) and Autodesk CFD (Autodesk Inc., San Rafael, California USA). In this CFD simulation, the boundary condition was 0 Pa (gauge pressure) at the intake, and a 5 m/s vertical at the discharge was used for a structure consisting of eight navicular units with three built-in 38 mm × 300 mm photocatalyst plates. The internal fluid was air (density 0.00120473 g/cm³, viscosity 0.0001817 Pa). A shear stress transport (SST) k-ω detached eddy simulation (DES) model with a high accuracy was adopted as the turbulence model. The meshes were generated using the automatic mesh size function of Autodesk CFD and made uniform. Six elbow bending angles were prepared: 90°, 100°, 110°, 120°, 130°, and 140°. Large eddy simulation (LES) is used for turbulence models because of its high reproducibility of unsteady flow; however, it has a very high analysis cost. Therefore, DES, a hybrid model of Reynolds-averaged Navier–Stokes (RANS) and LES with a low analysis cost, was used. Because of the high mesh dependence of DES, the mesh size was set to an automatic mesh size parameter of 0.5 (because in the 120° simulation, the results did not change significantly when set to a lower value of less than 0.5) and then a uniform mesh was applied. To reproduce the boundary layer flow, 10 wall layers were set for the surface subdivision and gap subdivision. The number of fluid elements in the gap was set to three and the number of individual elements was set to two. With these settings, the number of meshes generated was approximately 3.7 million (at 120°). The calculations were performed in 3000 steps with a time interval of 0.0002 s. The results of the CFD simulation are shown in the equipment design in Fig. 1.

2.2. Evaluation of aerosol removal performance using the multistage elbow system

2.2.1. Coloring experiment using dye aerosol

To visualize the aerosol removal behavior in the multistage elbow unit, we introduced dye aerosols into the multi-elbow system. A filter paper (No. 2, Advantech, Tokyo, Japan) of the same size as the three photocatalyst plates (300 × 114 mm) was placed on the three inner surfaces of each navicular elbow unit instead of the photocatalyst, and we observed the coloration condition of the filter paper. Aerosols containing rhodamine B (Wako, Osaka, Japan) aqueous solution (1 × 10⁻⁴ M) were generated using a glass nebulizer (Type V, AS ONE Corp., Osaka, Japan) and introduced into the multistage elbow unit for 30 min at 1.5 or 3.0 L/min.

2.2.2. Performance evaluation of each stage of the multistage elbow unit

The differential pressure between the navicular elbow units, wind velocity, and number of aerosol particles per unit stage were measured. A port for differential pressure and aerosol particle measurements was installed at the center of each navicular elbow unit. A micromanometer (Model 6850; Kanomax, Osaka, Japan) was used to measure the differential pressure. A hot-wire anemometer (Testo-425, Testo, Baden-Württemberg, Germany) was used to measure the air velocity. A particle counter (Aerotrak Handheld Particle Counter 9306, TSI Corp., Minnesota, USA) was used to measure the aerosol particles (0.3, 0.5, 1, 3, 5, and 10 μm particles/L). An ultrasonic nebulizer, NE-U780 (Omron, Kyoto, Japan), was used to generate ultrapure water aerosols, which were aspirated into a multistage elbow system.

2.2.3. Aerosol chamber removal performance test using the multistage elbow photocatalytic reactor

A 4.5 m³ (1.5 m × 1.5 m × 2.0 m) aerosol chamber was constructed to confirm the processing performance of indoor diffused aerosols using a multistage elbow photocatalytic reactor (Fig. 2). An aerosol supply outlet was installed at the center of the chamber, at a height of 1.5 m from the bottom. This value is the mouth height in the standing position, based on the average height of adults [83]. The sampling port of the particle counter is installed on one of the interior walls of the chamber. The sampling port height was configured at approximately 1 m above the floor level, which was assumed to be the height of the nose of a sitting human (calculated from the sitting height and sitting knee height, 42.1 Sitting height (erect), ISO 7250, 2017; 42.12 Lower leg length (popliteal height), ISO 7250, 2017) [84]. An ultrasonic nebulizer (NE-U780) was used to generate the aerosols. Because aerosols are heavier than air and settle at the bottom of the chamber without a circulator, the circulator was activated simultaneously with aerosol generation to equalize the aerosol distribution in the chamber. When the humidity in the chamber reached 100%, the aerosol discharge rate was the NE-U780 from 100% to 50% (to control the rate of decrease of the feed water to the nebulizer). After confirming that the aerosol concentration was stable, the circulator was stopped (to eliminate the effect of the circulator on the aerosol removal capacity of the multistage elbow system), and the operation of the multistage elbow system was started. Aerosol measurements were started simultaneously with the aerosol
discharge, and aerosol measurements of 0.3, 0.5, 1, 3, 5, and 10 μm particles/L were performed using a particle counter every 15 min (every 5 min for the first 15 min after the start of the operation of the multistage elbow system). The aerosol measurement using the particle counter was repeated three times, and the average value was used as the measurement data.

2.3.2. Photocatalytic SARS-CoV-2 inactivation experiment

The photocatalytic treatment performance of SARS-CoV-2 was tested at the Kanagawa Institute of Industrial Science and Technology (KISTEC). The photocatalyst used in this test was a WO$_3$-supported TiO$_2$ sprayed plate (50 × 50 mm), which exhibited the best performance based on the results of the ISO 22197-2 evaluation. The test virus was SARS-CoV-2/Hu/KngFJ/23875 and the host cell was a Vero cell (ATCC CCL-81, American Type Culture Collection). Eagle’s medium (EMEM) + 10% immobilized fetal bovine formation + antibiotics was used as the cell culture medium. EMEM was used as the maintenance medium. The maintenance medium + 0.75% cell culture agar was used as agar medium. Vero cells (100% confluent) were washed with the maintenance medium and inoculated with SARS-CoV-2. After 1 h of shaking every 15 min, the maintenance medium was added, and the cells were cultured for 5 days at 37 °C and 5% CO$_2$. The cells were then frozen and thawed twice and removed by centrifugation, and the supernatant was used as the viral stock solution. This virus stock solution was diluted 10 times with sterile water and used as the test virus solution (approximately 1–5 × 10$^6$ pfu/mL). The plaque method was used to measure the infectious virus titers. Vero cells were cultured in six-well plates (100% confluence). Subsequently, the medium was removed, and 0.1 mL of the virus recovery stock solution and staged dilutions were inoculated. After 1 h of shaking every 15 min, the agar medium (3.0 mL of agar medium) was overlaid and incubated for 5–7 days at 37 °C and 5% CO$_2$. After incubation, the cells were fixed in 10% formalin solution and the agar medium was removed. Finally, the number of plaques formed by the virus was measured by staining with methylene blue solution. The virus infection titer per sample was calculated from the number of plaques, dilution factor, and amount of soya casein-digested lecithin polysorbate (SCDLP).

The test virus was SARS-CoV-2/Hu/KngFJ/23875 and the host cell was a Vero cell (ATCC CCL-81, American Type Culture Collection). As pretreatment (removal of residual organic matter) before the antiviral test, WO$_3$-loaded TiO$_2$ thermal sprayed plates were subjected to UVA ($\lambda$ = 365 nm) treatment at an intensity of 1.0 mW/cm$^2$ for 48 h. For the virus elimination experiment, 0.25 mW/cm$^2$ ($\lambda$ = 365 nm) light irradiation for 30 min was conducted.

The sterile-treated photocatalyst plate was placed in a glass U-tube in

Fig. 2. Left: equipment layout of aerosol chamber. Right: photograph of aerosol chamber.
a Petri dish with the processed side up and inoculated with 0.15 mL of the test virus solution. The virus solution was adhered to the photocatalyst plate by covering it with an adhesion film. Additionally, a glass plate was placed on top of the plate for moisture retention. Thereafter, the cells were left in the dark or light irradiation conditions (0.25 mW/cm², λ = 365 nm). Samples were collected immediately after inoculation. The virus was then recovered from the photocatalyst plate and adherent film with 0.85 mL of the SCDLP medium. Next, a 10-fold stepwise dilution series of the recovered solution was prepared using the maintenance medium. Using the recovered stock and diluted solutions, viral infection titers were measured using the plaque method. The test was repeated three times, and the average value was calculated. An aluminum plate was used as the control for the photocatalytic plate.

Antiviral activity (V) was determined from the results of this test.

\[
V_L = \frac{\log(B_L) - \log(C_L)}{3} = \log\left(\frac{B_L}{C_L}\right),
\]

where \( V_L \) denotes the antiviral activity of the photocatalyst plate under UV irradiation (mW/cm²), \( B_L \) denotes the three-time mean value of the virus infection titer (pfu) after light irradiation of the photocatalyst plate (untreated specimen) for 1 h under UV irradiation condition L, and \( C_L \) is the three-time mean value of the virus infection titer (pfu) after 1 h of light irradiation of the photocatalyst plate under the UV irradiance condition L.

\[
V_D = \frac{\log(B_D) - \log(C_D)}{3} = \log\left(\frac{B_D}{C_D}\right)
\]

Here, \( V_D \) is the antiviral activity value of the photocatalyst plate; \( B_D \) denotes the three-time mean value of the virus infection titer of the photocatalyst plate (untreated test piece) under dark conditions (pfu). \( C_D \) is the three times mean value of the virus infection titer of the photocatalyst plate under dark conditions (pfu).

3. Results and discussion

3.1. Principle of multistage elbow system operation

The motivation for the design of the multistage elbow system was to establish a photocatalytic reaction system to efficiently irradiate the photocatalyst with excitation light. The resulting structure had a bent flow path to efficiently irradiate the photocatalyst with the excitation light. This bent structure was suitable for the elimination of aerosols.

According to ASHRAE Standard 34 (duct design), the elbow structure has completely different fitting loss coefficients depending on the bend angle and whether the bend is a smooth radius or mitered bend, with a mitered elbow generating a larger pressure loss than a smooth radius elbow [93,94]. In channels with mitered elbow structures, turbulence generation increases as the flow velocity increases [95,96]. Peter et al. reported that sufficient curvilinear motion for depositing large particles was not achieved when the bend angle was small [97]. By correlating with the pressure drop increase, it is easily predicted that particle accumulation will be more significant if the duct-bending geometry is a mitered elbow.

Therefore, considering the collection by particle collision to remove aerosols from the airflow, we deduced that the installation of many elbow points by zigzag folding of the fluid channel was suitable.

According to Tsuda et al. aerosol deposition mechanisms can be classified as (a) turbulent, (b) inertial, (c) gravitation, and (d) diffusional deposition [98].

The gravity efficiency (c) can be ignored because the flow speed of the system is extremely high (>5 m/s), and the diffusion deposition (d) shows that even with a temperature gradient of 100 °C/cm, the sedimentation rate is almost the same as that of (c) and can be almost completely ignored. Therefore, (a) and (b) represent the principles used in a multistage elbow-type system. The structure that generates the vortex and turbulence in (a) in the flow path structure includes pipes of different diameters and elbows.

Generally, aerosol deposition is more likely to occur in turbulent flows than in laminar flows [99], and inertial effects become more significant in the presence of large particles and strong turbulence [100]. In other words, aerosol particles maintain their straightness despite turbulence. In a structure with an elbow, a high movement velocity (i.e., a high flow velocity) causes collision with the wall. If the elbow angle is large, turbulence can be easily generated; however, a large angle causes an increase in the pressure drop, that is, attenuation of the flow velocity and an increase in energy loss. However, if this angle is minimized, the fluid flows smoothly and the energy loss (pressure drop) is reduced; however, the aerosol may simultaneously flow without hitting the wall. In other words, there must be an optimal angle for the multistage elbow. To determine the optimal angle, CFD simulations were performed when the elbow angle was changed every 10° between 90° and 140°. The results are shown in Fig. 3. We observed that when the elbow angle was greater than 120°, the pressure drops were smaller and turbulence was less likely to occur, resulting in near-laminar flow through the multistage elbow duct. In other words, the collision probability of aerosols on the elbow wall was relatively low. By contrast, when the elbow angle was smaller than 120°, the turbulence increased owing to the increased pressure drop, and the impact of the introduced particles on the duct wall increased. However, as the pressure drop increased, the air velocity decreased, resulting in a decrease in the air throughput per unit time (larger pressure drops resulted in a larger energy loss for ventilation). Thus, the optimal elbow angle was determined to be 120°. Based on these data, a photocatalytic aerosol elimination system with a built-in multistage elbow, as shown in Fig. 1, was designed and fabricated.

3.2. Visualization of aerosol deposition on photocatalytic surfaces in the multistage elbow system

The next problem under evaluation is the determination of the airspeed lower limit for a multistage elbow system. Theoretically, the faster the airspeed, the shorter the time required to clean the indoor air space; however, a powerful air-blowing capability (electricity) is required. To confirm the correlation between the airflow capability and aerosol removal capability, an aerosolized dye was introduced into the model to observe the coloration of the dye on the filter paper in place of the photocatalyst on the inner wall of the multistage elbow.

Fig. 4A shows the coloration for 30 min after the introduction of the dye aerosol and a 3D view of the distribution of the coloration intensity at a wind speed of 1.5 m/s. The measured results showed that strong deposition occurred in the fourth elbow unit. These results indicate that the main cause of deposition in the first elbow unit was the collision of aerosol particles from inertia caused by air intake, whereas aerosol deposition from vortex diffusion in turbulent flow was dominant in the elbow unit behind the first elbow unit. When the wind speed was set to 3.0 m/s, the coloration of the second elbow unit was stronger than that of the first unit, which was the entrance (Fig. 4B). This indicated that the maximum aerosol deposition moved to the intake side. To confirm the results of this actual measurement, we performed fluid analysis using CFD at a wind speed of 1–5 m/s. Consequently, we clarified that the introduced particles flowed along the wall without colliding with the elbow wall in the region near the intake side of the multistage elbow at a wind speed of 1 m/s. The effect of turbulence near the elbow side increased in the middle stage of the multistage elbow unit (Fig. 5A). When the wind speed was increased to 3 m/s, the introduced particles began to collide with the inner wall of the elbow unit near the intake. Because elastic particles are used in the CFD, the particles that impact the wall appear to leap. However, the main component of bioaerosol particles is water, and it is considered that the particles evaporate upon impact with a wall. The turbulence in the elbow unit on the side far from the intake increased (Fig. 5B). At a wind speed of 5 m/s, no significant difference was observed in the behavior of the particles compared to a wind speed of 3 m/s, except for an increase in the internal wind speed (Fig. 5C). This result indicates that the boundary at which the deposition
**Fig. 3.** Simulation of airflow in multistage elbow system for each elbow angle from 90° to 140°. The average airflow speed at the intake side was fixed to 5 m/s.

**Fig. 4.** Coloring condition of multistage elbow unit by introduction of dye aerosol (actual condition and 3D image of coloring). (a) 1.5 m/s average flow rate. (b) 3.0 m/s average flow rate.
of the introduced particles (aerosol) on the elbow unit wall became dominant was at a wind speed of approximately 3 m/s.

3.3. Decision on the minimum airflow in the multistage elbow system

Fig. 6 shows the dependence of the aerosol removal rate (discharge/intake ratio) of 0.3–10 μm in size on the air velocity when aerosols generated by a glass nebulizer were introduced at various air velocities into the multistage elbow unit. The aerosol removal capacity of all particle size regions was significantly low when the internal wind speed was low; however, the aerosol removal capacity of 1–10 μm particles increased rapidly when the wind speed exceeded 3 m/s. The removal capacity is approximately 100% at a velocity of 5 m/s. In other words, the inertial deposition of aerosols on the wall began at approximately 3 m/s. This result was in good agreement with the CFD results shown in Fig. 5.

3.4. Experiment in the aerosol chamber

Fig. 7 shows the temporal variation in each aerosol particle size in the aerosol chamber. When the aerosol discharge rate was set to 50%, the decrease in concentration from aerosol fall and evaporation was balanced by the increase in concentration from the continuous discharge, and the aerosol concentration was almost maintained at a constant level regardless of the particle size (Fig. 7A). By contrast, this stable condition changed dramatically with the operation of the multistage elbow system; a diameter larger than 1 μm was immediately reduced by 99.8%–99.9% after operating the multistage elbow system (Fig. 7B). After the aerosols were reduced, they did not increase despite the continuous supply by the nebulizer. This result indicates that the multistage elbow system can continuously process the aerosols.

Here, it is clear that continuous processing of aerosols can be achieved by the multistage elbow system; however, also another important factor is the expression of the photocatalytic performance that is not affected by humidity. This multistage elbow system has a built-in heating mechanism to avoid the effect of humidity on photocatalytic activity. Therefore, we evaluated the photocatalyst heating temperature in the multistage elbow system, the increase in temperature in the chamber, and the aerosol removal capacity. Fig. 7c and d shows the changes in the aerosol removal capacity, temperature, and humidity in the chamber when the heating temperatures of the multistage elbow system were 30 and 60 °C, respectively. When the power of the multi-stage elbow system was turned on, the temperature in the chamber increased owing to the heating of the heater, and the temperatures were +3.0 °C for 30 °C heating, +3.3 °C for 40 °C heating, and +4.9 °C for 60 °C heating. Approximately 20–25 min after heating, the relative humidity was <100%. This may have been due to an increase in the dew point caused by the increasing temperature in the chamber. From this heating experiment, we can conclude that the temperature increase in the indoor space was small even when heating was performed in the multistage elbow system, and the indoor temperature increase would be inversely proportional to its size if the space volume in which the

Fig. 5. Simulation of airflow in multistage elbow at various flow rates from 1 to 5 m/s (a) 1 m/s, (b) 3 m/s, and (c) 5 m/s.

Fig. 6. Elimination rate of aerosols with their sizes at various wind speeds by multistage elbow unit.
multistage elbow system is operated increases.

Table 2 presents the spatial aerosol removal capacity for each temperature of the multistage elbow system with photocatalyst heating.

| Aerosol size (μm) | Aerosol elimination % under each temperature |
|-------------------|---------------------------------------------|
|                   | 25 °C | 30 °C | 40 °C | 60 °C |
| 0.3               | 80.5  | 84.2  | 81.2  | 71.8  |
| 0.5               | 97.8  | 98.4  | 98    | 97.2  |
| 1                 | 99.8  | 100   | 99.9  | 99.7  |
| 3                 | 99.8  | 100   | 99.9  | 99.7  |
| 5                 | 99.8  | 100   | 99.9  | 99.8  |
| 10                | 99.9  | 100   | 100   | 99.8  |

Fig. 7. Time course of aerosol elimination in the aerosol chamber using a multistage elbow system with photocatalyst heating. (a) Without operation of the multistage elbow system. (b) Operation without heating (approximately 25 °C). (c) Operation with 30 °C heating. (d) Operation with 60 °C heating.

The space processing capacity of this multistage elbow system can be calculated using the processing air volume of the system. Fig. 8 shows the relationship between the actual measured values of the multistage elbow system treatment air velocity and the expected treatment volume per hour calculated from the air velocity. In Fig. 7, the shaded areas indicate that no aerosol removal was expected. As Fig. 7 shows, the aerosol removal capability of this system began to appear when the wind speed was 3 m/s or higher, and aerosols could be stably removed from 5 m/s. The volume of the classrooms in the aforementioned elementary schools was approximately 300 m³. If a blowing fan capable of securing a wind speed of approximately 12 m/s is selected, a treatment equivalent to full ventilation of this volume can be achieved in 1 h using one instrument. We used CFD to simulate how realistic this prediction is. The calculation method and the results are shown in Appendix. It was shown that air displacement in the classroom was almost completed before 1 h when the multistage elbow system was installed in the center of the wall behind the classroom with the wind speed set at 5 m/s. However, the CFD results showed that the air displacement tendency was different depending on the location of the multistage elbow system.
in the classroom. Thus, it will be necessary to conduct CFD with various factors in the future.

Anyhow, in response to the actual situation where the elimination of indoor aerosols is required owing to the spread of SARS-CoV-2 [102], this system is expected to significantly contribute as a countermeasure technology. However, this system is not intended for use in a completely closed room. The main reason for this is that ventilation is mandatory in major countries for controlling the increase in CO2 concentration [59, 103]. If a fan with high air velocity is used in a multistage elbow system, it would be possible to treat the entire volume of the classroom atmosphere; however, the fan operating noise and power consumption would consequently increase. Therefore, as mentioned in the introduction, because forced ventilation is mandatory for large spaces such as classrooms, it is sufficient if the aerosol treatment system can treat half of that volume in practice.

3.5. Performance evaluation of the thermally sprayed photocatalyst

The photocatalyst used in this multistage elbow system was a TiO2 photocatalyst deposited on an aluminum plate using thermal spraying. The photocatalytic activity was evaluated according to ISO 22197–2. Generally, the photocatalytic activity of the P25 standard sample exhibits an elimination performance of approximately 50% for 5 ppm acetaldehyde under ISO 22197–2. As presented in Table 3, our observed photocatalytic activity of the P25 standard catalyst at ISO 22197–2 was approximately 50% (slightly higher than this value). The activities of the thermally sprayed photocatalysts were approximately 50% higher than that of the reference (P25). Among them, we confirmed that the Fe3O4/TiO2 and WO3/TiO2 thermally sprayed plates were the materials least affected by changes in the relative humidity. It is known that photocatalytic activity is significantly affected by relative humidity [89–92]. However, because the multistage elbow system is equipped with a heating mechanism, we also determined whether the influence of relative humidity could be canceled by heating. The effect of the high-humidity environment was almost eliminated by setting the photocatalyst plate temperature to approximately 40 °C. As illustrated in Table 3, the activity of the standard photocatalyst (P25) decreased to approximately 60% when the humidity decreased from 80% to 50%. However, the thermally sprayed photocatalysts maintained activity above approximately 80% compared with the 50% humidity, except for the Cu2O-doped system. The effect of high humidity on photocatalytic activity was almost completely eliminated by heating the photocatalyst to approximately 40 °C.

3.6. Photocatalytic SARS-CoV-2 inactivation experiment

Among the thermally sprayed photocatalysts, the WO3/TiO2 photocatalyst, with exceptionally high catalytic activity, was evaluated for its ability to inactivate SARS-CoV-2 [104]. The antiviral activity results are listed in Table 4 and the antiviral activity is presented in Table 5. According to ISO 18061:2014 [76], an antiviral activity value of 2.0 or higher is considered effective, and the performance of the photocatalytic material presented in Table 5 is almost twice this value. Viruses are generally classified into nonenveloped and enveloped types. Among these, coronaviruses and influenza viruses are enveloped viruses, and they are easily inactivated by photocatalysis [78]. However, the probability of virus inactivation before UV irradiation was extremely high when the virus solution was dropped onto the catalyst plate in this experiment (Table 4). The reason for this is currently under investigation; however, it has been suggested that coronaviruses are vulnerable to environmental changes, and inactivation by drying may have progressed when moisture of aerosol was absorbed by the photocatalytic plate. The progress of inactivation before light irradiation was not the focus of this experiment, which was aimed at achieving virus inactivation by a photocatalytic reaction. However, if the bioaerosol is inactivated at the point of impact with the photocatalyst in the multistage elbow system, the bioaerosol removal capacity of this system is even higher. Of course, the inactivated virus on the photocatalyst will be decomposed into CO2 (and mineral components) and lost by photocatalysis. That is to say, the system is expected to endlessly maintain the bioaerosol removal capacity.

When the bioaerosol containing SARS-CoV-2 enters the multistage elbow system, it is deposited on the photocatalyst placed on the inner wall of the ventilation channel, and the viral infectivity titer is immediately reduced by UVA irradiation of several mW/cm2. Subsequently, the virus is decomposed through photocatalysis without being discharged from the system until it is completely mineralized. The use of this photocatalytic multistage elbow system enables high-speed processing of bioaerosols and cancels the increase in power load owing to the increase in pressure drop caused by the clogging of the filter and the requirement for filter replacement. Because this system is capable of high-speed and large-volume treatment, it is suitable for air treatment in classrooms and hospital rooms, where air-conditioning costs are likely to increase owing to ventilation. However, we predict that the system will not be practically applied unless a correlation between the CFD results and processing performance in real space is identified. Without clarifying this correlation, users will not be convinced. Particularly, we deduced that the simulated results are remarkably different depending

### Table 3

| Sample          | ISO 22197–2 Removal | %RH | Temp. | High S/RH Removal | %RH | Temp. | High %RH with Heating Removal | %RH | Temp. |
|-----------------|---------------------|-----|-------|-------------------|-----|-------|-----------------------------|-----|-------|
| P25             | 59.7%               | 50% | 25 °C | 38.1%             | 82% | 25 °C | 58.8%                       | 82% | 40 °C |
| TiO2            | 73.4%               | 50% | 25 °C | 59.2%             | 80% | 25 °C | 75.3%                       | 80% | 40 °C |
| Fe3O4/TiO2      | 78.1%               | 50% | 25 °C | 67.6%             | 82% | 25 °C | 77.7%                       | 81% | 46 °C |
| Cu2O/TiO2       | 74.2%               | 45% | 25 °C | 46.2%             | 70% | 25 °C | 69.0%                       | 71% | 39 °C |
| WO3/TiO2        | 75.4%               | 50% | 25 °C | 66.2%             | 81% | 25 °C | 76.7%                       | 81% | 40 °C |
on the installation position of the multistage elbow system in the classroom (in the center or corner of the wall) and location of the air conditioner and ventilation equipment. Additionally, as the fan used to generate high-speed airflow is noisy, sound control technologies must be developed. Our ongoing research seeks to solve these problems, and our findings shall be reported later.

4. Conclusions

Our developed photocatalytic multistage elbow system could almost completely remove aerosols using air velocities of over 5 m/s by forcing the aerosols to capture the photocatalyst placed inside the system through inertial force and turbulent diffusion. When the system was operated in a 4.5 m³ aerosol chamber, it immediately removed more than 99.8% aerosols with a particle size of 1–10 μm that were considered infectious; this performance was sustained for the duration of aerosol introduction. The performance of the system was equivalent to that of 1-h ventilation of a relatively large space such as a school classroom. The thermally sprayed photocatalyst used in this system almost completely thermally sprayed photocatalyst used in this system almost completely removed more than 99.8% aerosols with a particle size of 1–10 μm that were considered infectious; this performance was sustained for the duration of aerosol introduction. 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