Filtration of Viral Aerosols via a Hybrid Carbon Nanotube Active Filter

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Abstract

Exposure to expired particles and droplets carrying infectious viruses is a primary transmission pathway for respiratory diseases. Removal of particles and droplets via filtration from a volume can drastically reduce the exposure to viruses, but viruses may remain active on filtration surfaces as potential resuspension or fomite risks. Here, we report the development of macroscopic carbon nanotube air filters synthesized using ultra-thin carbon nanotube electrically conductive membranes, mechanically supported by a porous polyester backing. Filtration efficiencies were measured up to 99.999%, while ultra-thin materials with low areal density (0.1 g m⁻²) exhibited pressure drops comparable to commercial HEPA filters. These electrically conductive filters are actively self-sanitized by thermal flashes *via* resistive heating to temperatures above 80 °C within seconds or less. Such temperatures were proven to achieve full deactivation of a betacoronavirus and an adeno-associated virus retained on filter surfaces. A filtration unit prototype equipped with a CNT filter module (~1.2 m²) was shown to achieve air purification of 99% of a room within 10 minutes at 26 air changes per hour.

KEYWORDS: anti-viral, carbon nanotubes, air-filtration, bio-aerosols, multi-functional materials

1. Introduction

The global pandemic caused by the severe acute respiratory syndrome coronavirus 2 (SARS-CoV-2) has had a devastating effect on both human lives [1] and the global economy [2,3]. To combat this over the long term, means must be developed to disrupt the transmission of the disease by limiting the primary vectors for viral spread. Respiratory liquid aerosols (with droplet sizes $\leq 5 \mu$ m) are thought to be an important primary vector for many viruses, including coronaviruses, and are produced through expiration while coughing, speaking and breathing [4–6]. Respiratory particles that can penetrate into the lung can also remain suspended for hours and migrate over tens of meters via advection and diffusion, thus posing a hazard for indoor environments [7–10]. These aerosols can contain active SARS-CoV-2 virions for at least three hours [11], contributing to high infection rates in enclosed and crowded spaces [12–14]. To mitigate these risks, air

filtration in poorly ventilated (< 3 air changes per hour) or air-recycling dominant environments has been proposed as a means to limit disease spread [15,16].

To reduce ambient concentrations of viral particles, filtration media must allow for high fluxes of gases at low static pressure loss with high rates of particle removal and viral trapping or deactivation. Nanofiber-based filtration can achieve high aerosol filtration efficiency while exhibiting low-pressure drops. The increased surface area and the ability to lower gas flow drag due to the 'slip' mechanism enables non-zero velocity on the surface of the nanofibers [17]. As such, the smaller the diameter of the nanofiber, the better the filter's performance. Carbon nanotubes (CNTs), with diameters of several to a few tens of nanometers, are therefore ideal candidates as effective nanofiber filters. Techniques using CNTs to produce aerosol filters involve coating pre-existing filters with nanotubes, either by dispersion coating[18] or in situ CVD growth [19–23]. Another approach involves attaching free-standing CNT membranes to conventional microfiber backings [24,25]. Although these techniques offer filters with improved performance compared to conventional filtration media, the potential for broader impact has been limited due to complicated and costly synthesis techniques. Much progress has been made recently on advanced filtration materials. However, most of the focus was dedicated to the reusability of commercial face coverings by functionalizing them with innovative nanomaterials [26-31]. While these technological solutions show great promise in personal protection, they cannot be directly implemented to filters used in heating, ventilation, and air conditioning (HVAC) systems as they lack the filtration efficiency, ease of implementation over large filter areas, and proven ability to have pathogen disinfection capabilities under the dark conditions of a closed filtration module.

Here we report the first mass-producible air filter to possess HEPA-like pressure drop and efficiency, which is simultaneously self-sanitizing. The active filtration system is based on an electrically conductive CNT mat that can rapidly be sanitized via resistive heating. The free-standing CNT material has 10-100 nm sized pores that serves as the filtration media and electrically conductive network. To enhance the structural stability of the CNT mats and enable ultrathin-films with high filtration efficiency and low pressure drop, the CNT aerogels were collected onto mechanical supports of perforated polyester with ~100 μ m

sized holes. These hybrid CNT-polyester filters meet all relevant metrics required for HVAC filtration (i.e. filtration efficiency, pressure drop, heating rate, disinfection capabilities and price). This capability of actively neutralizing viral and other pathogenic threats is classed as "active", in contrast with a "passive" filter that captures pathogenic threats without neutralizing them. The multifunctional aspect of this filter allows its implementation in an innovative HVAC filtration system that can act as an advanced alternative to current solutions, especially being able to tackle the well-known surface persistence of the SARS-Cov-2 [32,33]. The mat is produced via the direct CNT spinning method by the floating catalyst CVD (FCCVD) process [34,35]. Tests showed the filter has an efficiency equivalent to High-Efficiency Particulate Air (HEPA) filters, independent of its thickness, while air permeability followed a Darcy's law-related trend. As opposed to standard microfiber filters, no apparent minimum of filtration efficiency was detected for any particle size. Although CNT-based filters show some inherent antiviral and antimicrobial capabilities [21,36] they are still prone, like any commercially available filters, to become biologically contaminated, posing a threat of re-aerosolization of pathogens or fomite transport [37,38]. A recent publication showed that resistive heating of nickel filter foams can neutralize airborne SARS-CoV-2 virions and anthrax spores [39]. Although effective in sterilizing, this approach required continuous applied power and relatively large energy consumption. Here we report the development of CNT filters with high permeability, high capture efficiency, and low thermal mass that can be sterilized insitu by Joule heating. Thermal analyses demonstrated that these filters can act as fastresponse heating elements. Furthermore, virus infectivity trials confirmed that total viral inactivation due to thermal exposure is achievable in a matter of seconds. Modeling indicates that adsorbed aerosols should readily desiccate once the power is applied, assuring that efficient energy management is achievable.

2. Methods

2.1 Filtration efficiency

The filtration efficiency tests were done on disc-shaped samples (d=25 mm) inserted into a conductive cassette blank (SureSeal cassette blanks, SKC). An electrically conductive housing was essential to minimize electrostatic losses, particularly for particles smaller

than ~50 nm. For firm mounting and to ensure proper circumferential sealing, the disc samples were sandwiched between a stainless-steel mesh support and a silicone rubber Oring (OD=25mm; ID=20mm). The tests were done using particles with mobility diameters in the range of 6-2500 nm. Tests done in the range of 6 - 100 nm used a silver (Ag) challenge aerosol generated by a bespoke particle generator which produces silver vapor that later recondenses into nanoparticles. The Ag is only used to test filtration performance and is not added to the filter more generally to affect biological activity or otherwise. The Ag aerosol is generated from a Ag powder evaporated inside a quartz test tube within a dedicated furnace. The generator was heated to a temperature range of 1280-1320 °C, running at a nitrogen flow of 2.2-2.5 standard liters per minute (slpm; HEPA filtered, BOC). The Ag nanoparticles were size-selected to discrete, nearly-monodisperse (geometric standard deviation ~1.05) mobility diameters of 6, 10, 15, 25, 50, 75, and 100 nm using a TSI-Differential Mobility Analyzer (DMA) with a 3085 DMA column and a 3080 electrostatic classifier. Analysis of filtration performance for particles in the range of 100-2500 nm used a Dioctyl sebacate (DOS, Sigma Aldrich, purity \geq 90%)) aerosol droplets created by a single-jet collision nebulizer (CH Technologies) running at nitrogen flows of 0.5-1 slpm. DOS droplets were size-selected by a Cambustion-aerodynamic aerosol classifier (AAC) to discrete and again nearly-monodisperse mobility diameters of 300, 500, 1000, and 2500 nm. As the AAC classifies particles using a particle's aerodynamic diameter, the appropriate conversion from aerodynamic to mobility diameter was used as shown in equation S5. Ag and DOS particle concentrations downstream of the cassette were analyzed by TSI-Ultrafine Condensation Particle Counters (UCPC) 3025A and 3776 respectively. For additional visual representation please refer to Figure S1. Particle concentration measurements were done for each selected size in a specified sequence of (1) running an empty cassette used as a blank run; (2) running a cassette loaded with a polyester backing (in the case of a hybrid filter); (3) running a cassette loaded with a designated CNT filter. Each measurement was repeated on three different samples and done with a nitrogen flow of 0.3 slpm through the filtration cassette. Filtration efficiency was calculated by the following equation:

$$E(d_p) = \frac{C_{\text{upstream}}(d_p) - C_{\text{downstream}}(d_p)}{C_{\text{upstream}}(d_p)} \times 100\%$$
(1)

where $E(d_p)$ is the fractional particle filtration efficiency, $C_{upstream}(d_p)$ and $C_{downstream}(d_p)$ are the number concentration of aerosolized particles measured without and with the filter, respectively.

2.2 SEM imaging

Two CNT filter samples for SEM imaging were produced to exemplify different particle capture mechanisms of the filter. The first was prepared by collecting aerosolized Ag nanoparticles (with a size range of 5-120 nm) for 45 minutes. The second was produced by applying a 10 μ L droplet of aqueous suspended 2 μ m polystyrene beads (Merck) diluted by a 1:100 ratio with deionized water (DIW), the droplet was air-dried at ambient temperature. The filter surface was imaged using a MIRA3 field emission gun-SEM (Tescan). Imaging was done at an acceleration voltage of 1 kV using the E-T SE detector (polystyrene beads) and 5 kV using the In-Beam SE detector (Ag nanoparticles) at a working distance of 3-5 mm. No conductive coating was added.

2.3 Filter pressure drop

The filter pressure drop tests were done on the same disc samples and cassettes described above. The volumetric flow was controlled from 0.1 to 6 slpm using a mass flow controller (Alicat) and suction was provided by a scroll vacuum pump (nXDS, Edwards). The pressure drop across the filter was measured using a differential pressure manometer (HD750, Extech Instruments) connected to the cassette inlet and outlet. All measurements were corrected by subtracting the inherent pressure drop of the blank filter cartridge.

2.4 Electro-thermal Analysis

Electro-thermal analyses of the free-standing CNT and PET-backed "hybrid" materials were carried out with a FLIR T650sc infrared camera (640×480 px resolution, $7.5-14 \mu$ m spectral sensitivity, 24mm f/1.0 optics) and a bespoke heating jig. The jig consisted of a sample holder with two adjustable parallel brass bar electrodes to which samples of different sizes (lengths between 75 and 120 mm, and widths up to 50 mm) could be clamped. The electrodes were connected to the terminals of a DC power supply (EX2020R,

AIM-TTI instruments). To minimize the effects of forced convection due to stray air currents, no fans or blowers were used in the vicinity of a running experiment and the jig was situated in a deep glass container.

Temperature versus power measurements were recorded by manually stepping the voltage applied to a 75x10 mm CNT strip while monitoring the average temperature within a 420 by 55 pixels square (encompassing most of the strip) with the camera's built-in software. At each setpoint, the current was recorded directly from the power supply console. The voltage step size and maximum voltages depended on the resistance of the sample (inversely proportional to its areal density). Each experiment was repeated on at least three different samples. Still images captured during the heat-up experiments were used to assess the heating uniformity of samples using both the "FLIR tools" software for a qualitative visual examination and a custom MatLab script for pixel-by-pixel quantitative analysis (code included in the SI appendix) to export pixel temperature information from the images.

The dynamic heating and cooling of the samples were characterized by recording thermal videos while manually switching the power supply on and off. The voltage was selected so samples would reach a stable temperature of around 80 $^{\circ}$ C (or 130 $^{\circ}$ C) when on. A Matlab script (SI appendix) was then used to extract the average temperature of the sample (from a 420px by 55 px crop of the frame) and the timestamp of each frame in the video. For each case, the results from a minimum of 10 heat-up (and, when relevant, cool-down) cycles were averaged to get the reported results.

2.5 Viral thermal inactivation and infectivity tests

Tests were done on mouse coronavirus (MHV-A59) as a surrogate. MHV-A59 is a betacoronavirus, within the same group as SARS and SARS-CoV-2. Murine 17 clone 1 (17Cl-1) cells were grown in Dulbecco's Modified Eagle's Medium (DMEM) low glucose supplemented with 5% (v/v) fetal calf serum (FCS), 6% (v/v) tryptose phosphate broth (TPB), 1x L-glutamine, 1x non-essential amino acid and 100 U penicillin ml⁻¹ and 100 mg streptomycin ml⁻¹ at 37 °C with 5% CO₂. Recombinant murine hepatitis virus (MHV) strain A59 recovered from full-length infectious MHV-A59 cDNA clone a gift from Dr Nerea Irigoyen, University of Cambridge, UK and propagated in 17Cl-1 cells supplemented with 0.2% bovine serum albumin (BSA) and 50 μ g/ml DEAE-dextran [40]. Virus stocks were aliquoted and frozen at -80°C, prior to being thawed and titrated to determine the tissue culture infectious dose 50 (TCID50) using the Reed and Muench method as described below [41]. To assess viral inactivation on CNT filters, 1 cm wide 7g m⁻² CNT strips were mounted on a dedicated heating jig (Figure 3a). 5µl droplets of MHV virus stock at a concentration of $\sim 8 \times 10^7$ infectious units mL⁻¹ (TCID50), were pipetted along the strip a total of four drops. The droplets were left undisturbed for a minute to let natural adsorption occur. Voltage was applied to the CNT strips and the CNT strips were heated to various temperatures (RT, 30, 45, 60, 80 °C) for a period of 90 s. In sequential experiments, fifty 0.4 μ l droplets (a total of 5 μ l) of MHV virus stock were pipetted along the strip and heated for different periods (0, 5, 10, 15, 30, 45, 60 s) at a temperature of 80 °C . As a control, 5 µl droplets of virus stock were distributed onto similar sized strips of non-adsorbent material used in disposable lab coats (43 gsm spunbond-meltblown-spunbond, non-woven fabric) and incubated at room temperature for the same period of time. Following incubation and heating, CNT strips (or control materials) were cut into four equal sized sections (~1 cm \times 1 cm) and transferred to elution tubes containing 500 µl of DMEM supplemented with 2.5% FCS and 3% TPB, vortexed vigorously for 10 s and then placed on ice for at least 5 minutes prior to titration by TCID50. To titrate the virus stocks and to assess the residual infectivity present in eluted samples, a TCID50 assay was performed as follows: ten-fold serial dilutions of the virus stock or the eluted viruses were prepared in DMEM supplemented 2.5% FBS and 3% TPB. 50 µl of these dilutions were inoculated onto monolayers of 17Cl-1 cells grown on 96-well plates and incubated at 37 °C a 5% CO₂ incubator. Plates were scored for cytopathic effect (CPE) after 48 hours and the virus titres expressed as TCID50 ml⁻¹ values by the Reed–Muench method [41]. Statistical analyses were performed on triplicate experiments using the two-tailed Student t- test (Prism 8 version 8.4.3). Figures were generated using Prism 8 version 8.4.3.

2.6 Droplet Evaporation – experimental and modeling

Experiments for visualizing the droplet evaporation process were run on a 110×40 mm 7 g m⁻² CNT strip substrate. The sample was placed in the strip-heating jig described above

using a gauge length of 75 mm. The CNT mat was heated to an average temperature of 80° C by applying a voltage of 4.35 V exerting a current of 1.76 A, which equates to an areal power density of 0.255 W cm⁻². DIW droplets were pipetted on the surface of the substrate with volumes of 0.1, 0.4, 1 and 5 µL. Each evaporation run was repeated at least three times. The image and video acquisition of the droplet evaporation was carried using a Dino-Lite AM4113T USB microscope (AnMo Electronics Corporation) at a magnification of X45. Image analysis was done using the Dino-Lite software.

A computational model was developed to simulate the diffusion-controlled evaporation of a water droplet on a CNT substrate with COMSOL Multiphysics (version 5.5). The model adopted a 2D axisymmetric geometry that revolved into a cylindrical domain including the CNT substrate, the water droplet and the ambient air. The overall height of the domain was 1,600 times the height of the droplet, to reduce the influence of evaporation on the ambient conditions, which were maintained constant at 25° C and 60% relative humidity. The radius of the domain was 40 times the base radius of a 0.4 µL droplet unless otherwise specified, to be consistent with the relative length scale used in the experiment. More details about other simulation parameters, along with the governing physics and the boundary conditions used in the simulation are covered in the supplementary information.

2.7 Performance evaluation of a CNT filter-based prototype

The filtration unit was placed in a chamber with a volume of 8 m³ made of plexiglass that was interconnected with Rexroth frames (BOSCH). A background scan of the particle concentration within the chamber was taken before each measurement. A 20-jet collision nebulizer (CH Technologies) was positioned on the chamber's floor, filled with a 20% w/w NaCl (>99.7%, Fisher Scientific) in DIW solution (volume 300 mL). Nitrogen (HEPA filtered, BOC) was delivered to the nebulizer, through an MFC, at a flow rate of 37 slpm atomizing the solution and filling the chamber with NaCl nanoparticles recorded to have a count median diameter and geometric standard deviation of 119 nm and 2 nm respectively (Figure S2). Filtration experiments were performed with a flow rate of 143 and 200 m³ hr⁻¹ and without any active filtration to determine the natural decay rate. After a two-minute aerosolization period, the flow rate to the nebulizer was stopped and the filter unit was

turned on externally. Each experiment was repeated at least 3 times. All particle concentration measurements were done using a TSI-UCPC model 3776.

3. Results and Discussion

3.1 Pressure Drop

Ultra-low areal density (<1 g m⁻²) CNT-polyester materials were developed to enable robust structures with high gas permeability. CNT aerogels were spun onto a porous polyester backing material (PET apertured spunlace, N.R. Spuntech Inc.; Figure 1a) via a continuous facile process. Upon collection, a bilayered hybrid material is formed, as shown in Figure. 1b, consisting of a thin CNT membrane (several hundred nanometers to several micrometers thick, Figure 1ci) on top of a 0.4 mm thick porous polyester backing (Figure 1ci). The hybrid composition was designed to minimize airflow resistance while maintaining mechanical integrity, ease of handling, and high filtration efficiency of the material. The CNT layer in the hybrid material is sufficiently thin that light is readily transmitted when visible backlighting is used to reveal its fine structure (Figure 1cii). The advantage of the synthesis and deposition process is that it does not require any posttreatment, thus preserving the single-step nature of the original CNT synthesis process. To assess the permeability of the free-standing CNT filters (>1 g m⁻²) and polyester backed filters (<1 g m⁻²) , the pressure drop was measured and permeability was determined by Darcy's law [42] for laminar flow through a porous medium,

$$U = \frac{\kappa}{\mu L} \Delta p = \frac{\kappa \rho}{\mu \rho_s} \Delta p = k \Delta p, \qquad (2)$$

where U is the air's velocity perpendicular to the face of the filter, K is the CNT filter's intrinsic permeability, ρ is the CNT bulk density, μ is the dynamic viscosity of air, L is the CNT membrane thickness, and Δp is the pressure drop developed across the filtration matrix. While CNT film thickness is inherently variable when the length scales of film thickness and pores are of similar orders of magnitude, the areal density ρ_s serves as a reliable surrogate for scaling. The CNTs' intrinsic permeability can be combined with the filter's areal density, bulk density and air viscosity to produce a coefficient, the filter permittance, $k \equiv K\rho/\mu\rho_s$ which directly relates the flow through the filter to the corresponding pressure drop. As expected, the hybrid filter permittance varies inversely with areal density, giving absolute value of the power-law fit near unity (a = -0.95) (Figure 2a). The intrinsic permeability of the material is remarkably stable, $K=6.01 \times 10^{-17} \pm 8 \times 10^{-19}$ m², over the samples of varying areal densities (0.1 to ~14 g m⁻²; Figure S4). Therefore, minimizing the CNT layer thickness in the hybrid filter provides a means for reducing the pressure drop for a given flow rate, while the filtration efficiency is maintained.

3.2 Filtration Efficiency

Although CNT aerogels have been characterized and evaluated for water purification and oil-water separation, [43,44] there has been no extensive research on their air filtration capabilities. Therefore, a wide spectrum of particle sizes (6-2500 nm) was chosen to assess the material's filtration capabilities and to find its so-called, most penetrating particle size (MPPS). Solid Ag nanoparticles were used as a test aerosol for sizes between 6 nm and 100 nm, while for sizes between 300 nm and 2.5 µm, low volatility dioctyl sebacate (DOS) oil droplets were used. This range covers the sizes of typical viruses (AAV ~20 nm [45] to SARS-CoV-2 ~100 nm [46]) to aerosolized droplets that contain the virus (~0.5 to >5 μ m [47]). As shown in Figure 2b, the CNT filters exhibit high and nearly constant filtration efficiency (>99.95%, the inverse of penetration) across the range of particle diameters for all CNT areal densities at 0.2 g m⁻² or above. The CNT filters with $\rho_s \ge 0.2$ g m⁻² have efficiencies that are comparable to H13 class HEPA filters. SEM images indicate the extremes of the particle filtration from diffusive collection for small particles (e.g. < 100nm, Figure 2c) to interception/impaction at larger sizes (> 1 μ m, Figure 2d). The hybrid filters exhibit no MPPS at the transition from diffusive to interceptive filtration, in contrast to what is typically observed for classical microfiber filtration materials (see FFP3 mask in Figure 2b), which exhibit a minima in filtration efficiency at the MPPS. Penetration values remained constant even when the CNT filters were "thinned" by more than an order of magnitude. Only the thinnest material produced with an areal density of 0.1 g m⁻² showed a significant increase in the penetration ratio (Figure 2b). At such low areal densities, defects in synthesis or handling lead to macroscopic defects in the CNT mesh resulting in a significant drop in filtration efficiency. Further work may lead to thinner films that maintain >99.95% filtration efficiency with improved deposition and bonding techniques.

The high filtration efficiency without an apparent MPPS is a result of the nanostructure of the CNT filter (i.e. bundles of several to few tens of CNTs) that are orders of magnitude smaller (10-50 nm) than the microfibers (0.8-20 μ m) [48] used in traditional filters. Traditional filtration curves, such as the experimental and theoretical model for a 3M FFP3 filter medium (grey full and dashed lines in Figure 2b), exhibit a characteristic minimum filtration efficiency that tends to be between 100 and 500 nm of 99.97% in compliance with manufacturer specifications [48]. In this size range, neither diffusion due to Brownian motion nor interception due to particles coming within one particle radius or less from a filter fiber are fully effective. As opposed to microfibers, the solidity and surface area of CNT filters are greater than traditional filters, allowing for overlap in filtration by interception and diffusion regimes (Figure S5). This results in a material that does not exhibit an MPPS, but rather where filtration efficiency is dictated purely by material defects in its homogeneity.

The filter quality factor [49] is a common means of assessing the ratio of the filter's filtration efficiency in comparison to its inherent pressure drop,

$$Q_f = \frac{-\ln(P)}{\Delta p} \tag{3}$$

where Q_f is the quality factor and *P* is the penetration ratio at the MPPS. The quality factor of the 7, 0.2 and 0.1 g m⁻² filters is 5.07, 45.56 and 39.75 kPa⁻¹, respectively, which are in line with other CNT-based filters reported in the literature [17] and within a factor ~2 of a HEPA H13 filter (Camfil). Although the 0.1 g m⁻² material shows an EPA E10 class filtration efficiency, it can still be adequate for aerosol filtration in air-recycling systems as the ultimate pathogen removal efficiency is a function of both pressure drop and filtration efficiency. For recycling air filtration systems, the removal function has a relatively weak dependence on filtration efficiency when recycled at a constant volumetric flow rate (Figure S6). Nevertheless, further research and characterization were focused on the 0.2 g m⁻² polyester-backed hybrids and the free-standing 7 g m⁻² CNT filters which displayed better inherent homogeneity.

3.3 CNT filters as efficient and fast-response heating elements

As the CNT filters are electrically conductive, it is possible to deactivate viral components by thermally denaturing captured pathogens through resistive heating [50]. To assess the power consumption-to-heat ratio of both free-standing and hybrid CNT filters, CNT sample strips were mounted on a bespoke heating jig (Figure 3a) and analyzed using an infrared thermal camera. As heat loss from the filters is approximately proportional to the surface area, the power consumption per unit area is comparable for different hybrid filter areal densities and resistances. As seen in Figure 3b, the behavior of both inspected materials is linear for temperature increases of $\Delta T < 100$ °C with gradients that represent the power density normalized heating of 242 and 273 °C cm² W⁻¹ for areal densities of 7 and 0.2 g m⁻², respectively. The linear dependence of ΔT with applied power suggests that heat conduction and convection are the primary mechanisms ($\propto \Delta T^1$) for heat loss, rather than radiation ($\propto \Delta T^4$) which will dominate at larger temperature differences between the filter surface and environment. The higher normalized heating for the hybrid material suggests that it loses slightly less heat than the bare, free-standing, CNT filters, primarily because of the thermal insulation of the backing and the reduced thermal conductance of the thinner CNT mat. CNT strips used in the analysis typically had a resistance of 4 Ω (7 g m⁻²) and 150 Ω (0.2 g m⁻²); thus maintaining a surface temperature of 80 °C. Voltages of 3 and 16 V were used to produce currents of 0.75 and 0.11 A in the 7 and 0.2 g m⁻² cases, respectively. Generally, it was found that a power density in the range of 0.20-0.25 W cm⁻² can reach and sustain a temperature of 80 °C which is higher than the 70 °C required to inactivate many viruses, such as, adeno-associated virus, hepatitis E virus, or SARS-CoV-2 [51–53].

To assess the filter's heating uniformity, thermal imaging was used as summarized in Figure 3c. The insert shows thermal images of the two materials at different average temperatures while the histogram gives the pixel by pixel (each pixel being roughly equivalent to a 100 μ m × 100 μ m square on the sample) temperature of the filters. The thermal uniformity of the filters with a mean temperature value of 82 °C in both cases is quantified by the standard deviation of 3 and 6 °C for the 7 and 0.2 g m⁻² samples, respectively. The increased thermal homogeneity of thicker samples is due to the greater

cross-sectional area of the conductive CNT materials (120-140 W m⁻¹ K⁻¹) [54]. To eliminate the chance of having cooler filter domains, a higher setpoint temperature was also analyzed. As seen in the dark red histogram, when the setpoint was adjusted to 130 °C the coldest point in the mat did not fall below 100 °C. Additional tests verified that the filtration efficiency of the 0.2 g m⁻² hybrid filter does not degrade due to heating cycles that simulate a period equivalent to year-long use (Figure S7).

The filter thermal response time provides an upper bound to the rate at which viruses can be deactivated. The thermal response was assessed using a frame-by-frame analysis of the samples' mean temperature in thermal videos while being heated to varying setpoints. As seen in Figure 3d and S8, when the setpoint is 80 °C both materials show short characteristic times of heating of <6 s due to the combined material heat capacities of the CNT filters (~800 J kg⁻¹ K⁻¹) [55] and ultra-low areal density (0.2 to 7 g m⁻²) resulting in a low areal heat capacity ($< 6 \text{ J m}^{-2} \text{ K}^{-1}$). The low heat capacity of the filters is desirable as it allows the films to achieve temperatures sufficient for viral deactivation with lower power consumption, leading to a quick and efficient flash sterilizing. While it takes 3.54±0.24 s for the 0.2 g m⁻² polyester-backed hybrids to heat from 30 to 70 °C (red line), the 7 g m⁻² free-standing filters do the same in only 0.48±0.24 s (blue line). The slower response of the hybrid CNT-polyester material is due to the additional thermal inertia of the polyester (~10-fold increase). However, by increasing the setpoint to 130 °C (dark red line), the 0.2 g m^{-2} hybrid shows a comparable response rate to the 7 g m⁻² material. These results confirm that once the correct heating parameters are established, viral deactivating temperatures above 70 °C can be achieved in <1 s to ensure that heating time is not the rate-limiting factor for viral deactivation.

3.4 CNT filter thermally induced virus inactivation

To assess the viral deactivation of the CNT filter, viral inactivation tests were run using a mouse coronavirus (MHV-A59). This is a beta-coronavirus, within the same group as SARS-CoV-2 and SARS, that can be handled outside of a containment level 3 laboratory. Firstly, experiments were run to find a "deactivation temperature" showing a significant drop in virus infectivity. 7 g m⁻² CNT strips were mounted on the heating jig (Figure 3a) and 5 μ L of virus containing droplets were pipetted along the CNT strip. Experiments were

also conducted on a reference sample (disposable lab coat), a control sample (0 V) and four more samples at varying voltages producing a surface temperature not lower than 30, 45, 60 and 80 °C (1.3, 2.0, 3.2 and 4.0 V respectively). As portrayed in the reference column (0 V) seen in Figure 4a, the experimental protocol was adequate for detecting viral infectivity using samples acquired from CNT mats. Even by applying low voltages, an evident decrease in viral infectivity was observed at temperatures less than 70 °C (typical deactivation temperature of SARS-CoV-2 and SARS virus [51]). This phenomenon suggests that direct surface oxidation may be an additional deactivation mechanism as found previously for water-borne virus filters [56]. At an applied potential of 4 V, a fourorder magnitude drop in infectivity was observed, reducing the virus titer to the limit of detection (LOD). The full deactivation of the virus at this voltage is a result of full droplet evaporation witnessed within 90 seconds, resulting in virion exposure to surface temperature higher than 70 °C. In order to determine the minimal time needed to achieve total deactivation when applying a 4 V potential, smaller, virus-loaded droplets (0.4 μ L) were pipetted on the CNT strips while a heating cycle was run for 5, 10, 15, 30, 45 and 60 s. As seen in Figure 4b, even though full evaporation was not achieved after 10 seconds, there was an evident decrease in infectivity due to possible surface oxidation resulting in loss of function of ~90% from the initial viral load. Heating for 30 and 45 s led to the evaporation of the droplets and was associated with a significant deactivation level of about one order of magnitude. A heating period of 60 s led to full deactivation (LOD), which was likely a result of prolonged exposure to the deactivation temperature. Further viral deactivation experiments were done on adeno-associated virus 9 (AAV9) using the above method. Full deactivation of AAV9 contained in 0.2 µL droplets was achieved after 30 s when the CNT substrate was heated to 80 °C (Figure S9).

The results above demonstrate that the principle of resistive heating for self-sterilization of the CNT filters is valid for a virus from the same group as the SARS-CoV-2. For experimental sensitivity, the droplets used in these experiments were loaded in extremely high concentrations of the active virus, many orders of magnitude higher than estimated to be exhaled by individuals actively spreading the virus [57]. As such, it is assumed that in a real-life application the pathogen loading should be lower and thus making it easier to sterilize to an acceptable level. Experimentally, we have shown that full deactivation is

achievable on millimeter-sized droplets. These droplets require significantly higher energy for evaporation in comparison to micro-sized aerosols ($<5 \mu$ m). Incorporating a theoretical understanding of the dynamics of aerosol evaporation on a CNT mat gives insight into the timeframe needed for full evaporation and thus total deactivation of virus-containing aerosols.

3.5 Droplet and aerosol drying on a heated CNT substrate

To further bound the timescales associated with viral deactivation, we studied the evaporation process of surface-bound aqueous droplets with experimental and computational methods. Α computational model simulated the diffusioncontrolled evaporation of a droplet on a heated CNT mat. The model was validated for droplets in the continuum regime using light microscopy imaging on several millimetre to sub-millimeter water droplets undergoing evaporation on a heated CNT mat (Figure 5a). The measurements showed that there is no change in the droplet base area during most of the evaporation period, indicating that water droplets on the CNT surface follow a "constant contact radius" (CCR) evaporation mode which is described further in the literature.[58,59] The CCR evaporation mode was embedded in the model under consideration of convection within the droplet and diffusion of water vapor from the interface to the ambient environment.

Figure 5a shows that there is good agreement temporally between the measured and modeled evaporation process for a measured 0.4 µL water droplet. The minor overestimation of the evaporation time by the model (19 s) as compared to the experimental result (15 s) can be explained by modeling inaccuracies due to the absence of the account for water infiltration into the CNT mat. An agreement between experiment and theory for 0.1, 0.4, 1 and 5 µL droplets is visualized in Figure 5b, in which the red circles (experimental) corroborate the black (modeled) squares. The results are in line with the reported power-law temporal scaling for CCR evaporation dynamics (evaporation time t_f and initial volume V_0 related by $t_f \propto V_0^{2/3}$) [60]. As portrayed by the CCR power law (dashed black line) it can be inferred that the evaporation time of micro- and nano-droplets should be less than several milliseconds. These results fit well with an analytical quasisteady-state model developed by Hu and Wu [60] for the evaporation of small sessile

droplets under isothermal conditions. Our results, derived from cases where the interface temperature was ~70 °C, lie well between the 50 to 70 °C isotherms of the Hu and Wu model portrayed by the green lines (Figure 5b). Although our model required adaptations to better fit the current scenario in which a constant volumetric heat is generated at a known rate by the CNT substrate, such an agreement gives confidence to its applicability.

After establishing the validity of the model, we assessed other parameters influencing the evaporation time. The filtration period, directly affecting the aerosol surface concentration has a significant impact on the evaporation time. As seen in Figure 5b, the higher the aerosol surface concentration (represented by squares turning from red to orange, blue and turquoise), the longer the evaporation period becomes, reaching fractions of seconds for aerosols with an initial diameter of 5 μ m. As the schematic in Figure 5c depicts, the simulation showed that as more droplets are captured by the CNT filter, the energy flux to each surface-bound droplet decreases. The reduced energy flux leads to a lower droplet interface temperature and lengthened evaporation time. However, as seen in the dynamic thermal behavior (Figure 3d) and illustrated in Figure 5d even with an evaporation timescale on the order of a few hundreds of milliseconds, all aerosols should evaporate well within a 5 s, 130 °C flash pulse (yellow zone). Once all aerosols completely evaporate, the virions will be directly exposed to a surface temperature higher than 80 °C, which has been shown to deactivate the modeled virus [51]. These results indicate that viral deactivation can be achieved using short flash pulses, ensuring the high thermal efficiency of the active filter system. It also gives appropriate timescales for optimization between flash heating intervals and pulse duration to balance energy consumption and viral deactivation of in-use filters.

3.6 Performance evaluation of a CNT filter based prototype

As the CNT aerogel synthesis process can produce CNT mats in large quantities, it was possible to produce a full-scale CNT hybrid filtration module to be fitted to a conventional recirculating filter unit. As illustrated in Figure 6a, the system was designed to draw ambient air by a centrifugal blower (RG175/2000, ebm-papst UK) and to direct the air, outwards, through a cylindrical filter module. The filtered air is recycled back to the ambient air thus reducing the airborne particle and droplet concentration in the

environment. As shown in Figure 6b, the filtration module was produced by fitting a $\sim 1.2 \text{ m}^2$ of 0.2 g m⁻² CNT hybrid on a cylindrical stainless steel coarse mesh so the CNT membrane faced inwards, ensuring mechanical membrane support. The module assembly was then fitted into the filtration unit (Figure 6c). As illustrated in Figure 6a, the efficiency of particle reduction was measured within an enclosed volume (~8.0 m³) after introducing a significant concentration ($\sim 3 \times 10^5 \, \text{# cm}^{-3}$) of NaCl nanocrystals acting as a model aerosol. The count geometric mean diameter of the nanocrystals was adjusted to be ~120 nm (Figure S2) to correspond to a typical MPPS for filter media, thus representing the most challenging test aerosol (also approximate size of SARS-CoV-2 virion [46]). To evaluate the system's efficiency in purifying enclosed environments, it was operated using two flow rates (143 and 200 $\text{m}^3 \text{hr}^{-1}$) which correspond to 16 and 23 air changes per hour (ACH) of the internal volume which is in line with the current guidelines for isolation rooms which are >12 ACH [61]. The decay rate of the suspended particles was monitored using a condensation particle counter. To properly decouple the decay rate resulting from active filtration from the total rate, the natural decay rate (achieved due to leaks, diffusive losses, etc.) was subtracted from the total decay rate. From Figure 6d a characteristic exponential decay is apparent. Lowering the pollutant number concentration to background levels (~ $6.5 \times 10^3 \, \text{# cm}^{-3}$), only by filtration, took approximately 15 and 11 minutes using a flow rate of 143 and 200 m³ hr⁻¹ respectively. In comparison, by extrapolation of the green line, natural decay should do the same within ~350 minutes. The experimental results (solid lines) correspond well with the theoretical model (dashed lines) that assumes the suspended particles are fully mixed within the enclosed volume (see equation S8). A slight deviation is seen at very low particle concentrations ($<3x10^2 \text{ # cm}^{-3}$), due to minor leaks from the ambient environment inwards to the confined volume. Overall, these results show a promising application for these hybrid CNT materials in a full-scale filtration system.

4. Conclusion

The results of this study show that active CNT hybrid filters exhibit excellent filtration efficiency (HEPA H13 level) while maintaining a low pressure drop. The active filter can be flash heated to 130 °C within seconds, leading to full viral inactivation as shown by trials done with AAV9, and betacoronavirus heated to 80 °C on a CNT substrate.

Implementing the filter material in a large filtration module ($\sim 1.2 \text{ m}^2$) installed in a prototype unit showed that a 10-fold decrease in air contamination in several minutes is achievable. Such units, deployed in ill-ventilated and crowded environments (i.e. offices, public transportation, leisure and recreational centers etc.) whether in conjunction with HVAC systems or as independent units, can have a material impact on the means to fight the viral spread of airborne diseases, not only COVID-19 but others such as seasonal influenza, which has been sown to inflict a total economic burden equivalent to \$87.1 billion, in the United States alone [62].

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Figure legends



Figure 1. The hybrid CNT filter. (a) The adapted direct spinning method using a collection bobbin covered with the polyester backing for the in-situ production of the CNT hybrid filter material. (b) An illustration showing the concept of the active hybrid CNT filter. The CNT filter can retain SARS-CoV-2 virions and aerosols containing them. The hybrid can be actively sterilized via resistive heating enabled by applying a potential between two electrodes. (c) Photographs showing (i) The upper layer of the hybrid that is made from a micrometer thin non-woven CNT mat. (ii) The lower layer is made from a porous polyester backing. (iii) The fine structure of the hybrid as revealed by a backlight.



Figure 2. Permittance and filtration efficiency of the CNT filter. (a) CNT filter permittance (blue circles, left axis) and penetration (red squares, right axis) as a function of CNT mat areal density. The permeability trend line (blue) follows a slope of -0.946, fitting well with the theoretical value of -1. The penetration values differ slightly with areal density, only the 0.1 g m⁻² material shows an increase, associated with macroscopic defects. (b) Filtration efficiency as a function of particle diameter shows that CNT filters (apart from the 0.1 g m⁻² material) exhibit constant filtration efficiency across the full particle range, with no apparent 'U' shape profile. The filtration efficiency is comparable to an H13 class HEPA filter. FFP3 mask material shows an experimental (solid grey line) and theoretical (dashed grey line) classical 'U' shape behavior with a most penetrating particle size (MPPS) residing in the range of hundreds of nanometres. (c) SEM image showing the CNT filter surface after Ag nanoparticles (5-120 nm) aerosol filtration. Due to Brownian motion, nanoparticles, even smaller than the apparent pore size can be efficiently retained. Scale bar, 500 nm. (d) SEM image showing polystyrene microbeads (2 μ m) deposited on the filter surface. Microparticles are significantly larger than the filter's pore sizes and are mechanically sieved. Scale bar, 2 μ m. . Error bars denote standard deviations using at least three different samples.



Figure 3. Electro-thermal behavior of the CNT filter. (a) The bespoke heating jig used for the electro-thermal experiments. Its design allowed using CNT strips with various dimensions and assured firm electrical contact on both ends. (b) Temperature increase (ΔT) as a function of areal power density shows a linear correlation at this temperature range. Slopes of 242.01 and 272.96 °C cm² W⁻¹ respectively for the free-standing 7 g m⁻² and the 0.2 g m⁻² polyester-backed hybrids. The regions bordered by dashed lines enclose 90% of data points (±4.65 °C for 0.2 g m⁻² and ±4.42 °C for 7 g m⁻²). (c) Filter's areal heating uniformity shown by thermal imaging reveals a homogenous pattern (inset). Histogram made by pixel by pixel temperature analysis shows mean temperature values of 82°C in both cases, the standard deviation is of 3 and 6 °C for the 7 (red bars) and 0.2 g m⁻² (blue bars) samples respectively. 0.2 g m⁻² set to a temperature of 130°C (dark red bars) is shown to assure no domain is colder than 100°C. (d) The heating response time shows it takes 3.54±0.24 s for the 0.2 g m⁻² polyester-backed hybrids to heat from 30 to 70°C (red line), and the 7 g m⁻² free-standing filters do the same in only 0.48±0.24 s (blue line). By increasing the setpoint to 130°C (dark red line), the 0.2 g m⁻² hybrid shows a comparable response rate to the one shown with the 7 g m⁻² material.



Figure 4. Viral infectivity due to thermal exposure. Results show the percentages (%) of remaining infectivity levels of the surrogate coronavirus on CNT substrates heated to various temperatures or heating periods. (a) 5 μ L virus-containing droplets were heated for a period of 90 s. Control is based on the infectivity level of the stock solution. RT (room temperature) shows the infectivity levels of samples that did not undergo active heating. Full inactivation is seen when the CNT mats were heated up to 80°C. (b) 0.4 μ L virus-containing droplets were heated to a temperature of 80°C. Full inactivation is seen after a period of 60 s. Error bars represent the mean ± SEM of at least four biological samples analyzed. Statistically significant values are represented as: ns=not significant, *=p≤0.05,**=p≤0.005,**=p≤0.001.



Figure 5. Droplet and aerosol drying on a heated CNT substrate. (a) Images of evaporating a 0.4 μ L droplet on CNT filter at T = 80 °C at advancing τ , taken as the ratio of time and full evaporation time, t_f . (top line) Images confirm a "constant contact radius" (CCR) evaporation mode with $t_f = 15$ s and scale bar, 1 mm. (middle line) Plan view of FEM numerical results for droplet surface height corresponds to the experimental conditions. (bottom line) Cross-section of the FEM results of droplet temperature profile with $t_f = 19$ s. (b) Plotted results of t_f as a function of the initial droplet volume, $V_0^{2/3}$ (equivalent initial droplet diameters on the secondary x-axis). FEM results assume a contact angle of 70° with upper and lower error bars denoting contact angles 100° and 30°, respectively. Experimental measurements (red circles) agree with FEM results (black squares). The linear correlation (black dashed line) indicates aerosols ($\leq 5 \mu$ m) will evaporate in $t_f < 1$ ms, which agrees with the analytical quasi-steady-state Hu and Wu model. Colored lines represent different surface concentrations based on filtration time (1, 5, 15 and 60 min for red, orange, blue and turquoise respectively). (c) Illustration representing the CNT filter thermal behavior as a function of aerosol loading with higher concentrations leading to lower temperatures. (d) Plot of measured temperature response of an 0.2 g m⁻²CNT hybrid filter with a 5 s heat flash set to 130 °C.



Figure 6. Developing and testing an active filtration prototype unit. (a) An illustration showing the prototype test setup. The unit was placed in an enclosed volume (8.7 m³). NaCl nanocrystals (with a geometric mean diameter of 119 nm) were introduced to the chamber through a 20-jet collision nebulizer. The aerosol concentration was continuously monitored using a condensed particle counter (CPC). (b) A photograph showing the construction of the filtration module based on the 0.2 g m⁻² CNT hybrid filter material. (c) A photograph of the internal parts of the active filtration prototype unit. Surrounding, contaminated air is drawn to the upper chamber by a centrifugal blower, then blown downwards into the filtration module internal volume. The air is purified by passing through the filter outwards and consequently recycled back to the environment. (d) A decay plot showing an exponential decay behavior was recorded during active filtration with flow rates of 134 (blue) and 200 (red) m³ hr⁻¹. Lowering the pollutant number concentration to background levels took ~15 and 11 min using a flow rate of 143 and 200 m³ hr⁻¹ respectively. The experimental results (full lines) fit nicely with the theoretical model (dashed lines) dealing with decay rates of pollutants in fully mixed confined volumes. Filtration decay rates were decoupled from the measured total decay rates by normalizing those with the natural decay rate (green line).

Author Contribution

Liron Issman: Project administration, Investigation, Data Curation, Visualization, Writing - Original Draft, Brian Graves: Investigation, Visualization, Writing - Original Draft, Jeronimo Terrones: Investigation, Visualization, Methodology, Writing -Original Draft, Myra Hosmillo: Investigation, Writing - Original Draft, Rulan Qiao: Software, Writing - Original Draft, Michael Glerum: Investigation, Writing - Original Draft, Shuki Yeshurun: Conceptualization, Resources Martin Pick: Conceptualization, Funding acquisition, Ian Goodfellow: Investigation, Writing - Review & Editing, James Elliott: Supervision, Writing - Review & Editing, Adam Boies: Supervision, Writing -Review & Editing, Funding acquisition.

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Competing interests

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