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Utilization of Medical Mask Wastes as Filter Material for Air Purification Units

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The COVID-19 pandemic significantly increased the amount of infectious medical wastes produced, with medical mask wastes being one of the largest contributors. Present research focuses on trying to turn medical mask waste into a functioning air filter by modifying it with CuO/TiO_2 to reduce the amount of infectious medical wastes laying around. Synthesis of CuO/TiO_2 was confirmed with FESEM-EDX, UV-Vis DRS and XRD techniques. The optimum amount of Cu added (1%wt of TiO_2) was determined by assessing the degradation performance of the modified medical mask wastes against an organic pollutant (methylene blue) and a biological pollutant (*S. aureus*). The filter was then integrated into a simple air purifying unit and complemented with a UV-C germicidal lamp and a plasma ion generator. The prototype of the simple air purifying unit was able to degrade 100% tobacco smoke in less than 15 min and 30.8% CO gas in 30 min.

Keywords: CuO/TiO_2, Medical mask waste, Air purification, Bacteria disinfection.

INTRODUCTION

Despite efforts being made to preserve energy supply and initiate clean energy transition, the ongoing issue of climate change, rapid population growth and global dependency on fossil fuels harnessed a direct negative impact on the world's air quality. The 2021 World Air Quality Report ranked combustion of fossil fuels as the primary contributor to particulate matter (PM) 2.5 pollution, while at the same time announcing Indonesia as the most polluted country in Southeast Asia and 17th place worldwide [1]. Furthermore, toxic gases (SO_2, NO_x, CO and CO_2) and $PM_{2.5}$ produced from burning fossil fuels also cause of death of 8.7 million people in 2018 [2].

Rapid transmission of air-borne droplets diseases such as COVID-19 and pneumonia with bacterial origin exacerbate the air quality problem into a dire public health concern. This continuing public health crisis mandates the need to ensure good air quality to be maintained not only outdoors but also indoors, as both COVID-19 and bacterial pneumonia have an increased risk of transmission in closed quarters especially in densely populated clinical or community settings. The prominent pathogenic microorganisms causing bacterial pneumonia has been shown to be associated with COVID-19 complications is known as *Staphylococcus aureus,* which sequelae can lead to necrotizing pneumonia, formation of abscesses, bacteremia, sepsis and eventually death [3,4]. Moreover, clinicians worldwide are also simultaneously combating another concern being antimicrobial resistant staphylococcal strain (also known as methicillin-resistant *Staphylococus aureus* (MRSA)), hence making prevention of transmission heavily sought after to reduce the medical burden [5]. In addition to the ongoing environmental crisis and global health concern, COVID-19 pandemic resulted in another problem which needs to be addressed. That being an increase in the amount of (potentially) infectious medical waste produced with medical mask wastes (MMW) being one of its largest contributors [6,7].

Thus, a solution to simultaneously solve all three problems stated above is proposed in this study. This study focuses on engineering simple air purification units that is effective against toxic gases and biological pollutants while using medical mask waste as a base material for the filters in said purifying units. The use of semiconductors like TiO_2 as a photocatalyst have been proved to be effective in degrading toxic gases and microorganisms before [8,9], but these semiconductors have an underlying weakness, which makes them unsuitable for air pollutants degradation application. That weakness is their weak

adsorption properties, which can be solved by pairing them with supports that have good adsorption properties [8]. Medical mask wastes have the potential to be an alternative support for air pollutants degradation application because of its ability to adsorb air pollutants and slow down air flow which can increase the contact between radical oxidative species (ROS) produced when a photocatalyst receives photon from a light source and the air pollutants.

To increase the performance of air purification unit against microorganisms, TiO_2 was doped with CuO, which has been proved to significantly increase antimicrobial properties of TiO_2 [10]. The use of copper as metal dopant to TiO_2 for air pollutants degradation has never been researched before unlike Ag and Fe [11]. To improve the overall performance of the air purification unit, a germicidal UV-C lamp, which can deactivate microorganisms and degrade pollutants by photolysis [12] and a plasma ion generator which can increase the amount of ROS produced were added into the unit [13].

This research aims to convert medical mask wastes into a functional air filters by modifying it using CuO/TiO_2 to reduce the amount of infectious medical wastes laying around. The optimum amount of Cu added was determined by assessing the degradation performance of the modified medical mask wastes against an organic pollutant (methylene blue) and a biological pollutant (*S. aureus*). The filter was then integrated into a simple air purifying unit equipped with a UV-C germicidal lamp and a plasma ion generator. The performance of the purifying unit engineered was evaluated by exposing it to tobacco smoke, a multicomponent pollutant that has similar components as the toxic gases produced from burning fossil fuels.

EXPERIMENTAL

Synthesis: CuO/TiO_2 was synthesized using a modified impregnation method. $CuCl_2{\cdot}2H_2O$ (Merck) and TiO_2 P25 (Evonik) were dispersed into double distilled water and then thoroughly mixed until a suspension was formed. The water content was evaporated from the mixture by drying it for 3 h at 110 ºC. The mixture was then ultrasonified for 10 min and calcinized for 2 h at 300 ºC. The composite obtained was finally grinded and poured into a 200-mesh test sieve.

Characterization: The existence of Cu in the composite was confirmed with FESEM-EDX (INSPECT F50) in high vacuum mode with these working conditions: 50000x magnifications, 30 kV voltage, working distance of 12.4 mm, spot size of 6.0 and point scan EDX mode. Formation of Cu crystal in the composite was analyzed with XRD (Empyrean Series 3 Panalytical) at 40 kV and 30 mA using Cu anode ($K\alpha = 0.15406$ nm). The effect of CuO addition towards composite's band gap energy was analyzed with UV-Vis DRS (Harrick Scientific Agilent Cary 600 UV-Vis DRS).

Medical mask waste (MMW) modification: Initially, MMW was subjected to modified with $US/H_2O_2/UV$ treatment process [14], where H_2O_2 was substituted with C_2H_5OH, another viable decontamination method [15]. To remove existing dust particles, debris and particulate matters from medical mask waste (MMW), it was soaked in distilled water and ultrasonified for 10 min. After drying the MMW for 30 min by exposing it to hot air, MMW was dipped into a 70% ethanol solution. After drying, each side of MMW was exposed to UV-C irradiation for 5 min. Now, MMW was modified by coating it with CuO/TiO_2 sol, which was made by dispersing the optimum amount of CuO/TiO_2 (1 g/L water) into distilled water, using the dip coating method. MMW was placed inside a dip coating device and then CuO/TiO_2 sol was poured. CuO/TiO_2 sol was drained from the device at a constant rate. The MMW was dried for 30 min following the dip coating process. These steps were repeated until three cycles of dip-coating were completed. The mixture was mixed for 10 min and then 2 mL of tetraethyl orthosilicate (TEOS) (Sigma-Aldrich) was added followed by the mixing for another 5 min.

Degradation studies: Methylene blue degradation was carried out in a cylinder shaped photoreactor with a volume of 500 mL using An 11 W YAMANO UV-C lamp. Methylene blue (400 mL) with an initial concentration of 50 ppm was poured into the photoreactor. The experiment was carried out for 2 h after the UV lamp and the magnetic stirrer were turned on. The absorbance of every sample taken was then measured using a UV-vis spectrophotometer.

Bacteria disinfection: One loop of dried culture of *S. aureus* (prepared by Qlab) was mixed with lysogeny broth, incubated at 37 ºC for 24 h and stored as a stock culture at 4 ºC. The sample used for the experiment, was prepared by mixing it with 25 mL of tryptic soy broth. The colonies from the stock were transferred into a NaCl solution and the concentration of bacteria was adjusted to 10^6 CFU/mL. A bacteria stock culture (1 mL) was added into a beaker glass where a sample had been immersed in distilled water prior. The beaker glass was then put into the photoreactor and the experiment was carried out for 2 h. The samples that were taken every 30 min were put into petri dishes and the petri dishes were filled with plate count agar (PCA) solution. After the PCA had solidified, the petri dishes were incubated at 37 ºC for 24 h and observed to estimate the total number of bacteria for every sample.

Toxic gas degradation: The air purification unit is configured in a way that maximizes the intensity of light received by the composite and the air residence time in the unit and minimizes pressure drop and turbulence. The experiment was carried out in a 0.343 m^3 test chamber, where the air purifying unit was put in beforehand. Ecom-D gas Analyzer's probe was then inserted into the test chamber. Tobacco smoke was injected periodically with a 100 mL syringe until the concentration of CO gas in the test chamber was 400 ppm.

RESULTS AND DISCUSSION

XRD studies: The existence of copper in the CuO/TiO_2 composite was confirmed through XRD patterns (Fig. 1). A new peak formed at $2\theta = 32.23°$ is due to a significant increase of intensity on the existing peak at $2\theta = 38.54°$, which indicate that Cu has been successfully doped in CuO/TiO_2 composite. This hypothesis has been confirmed with JCDPS Card 48-1548. Meanwhile, the anatase and rutile peak of TiO_2 can be seen at $2\theta = 25.25°$ and 27.38º, respectively which matched with JCDPS card No. 21-1272. The crystalline size was also calculated from

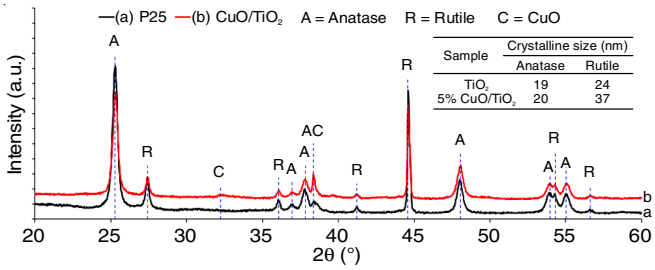

Fig. 1. XRD characterization results and calculated crystalline size for undoped TiO_2 and 5% CuO/TiO_2

the XRD characterization. The addition of CuO increases the crystalline size of the photocatalyst, which is caused by thermal treatment during calcination in the synthesis process [10].

Morphology studies: Aggregate formation and an increase of particle size as CuO loading is increased can be seen in Fig. 2a-c, which shows the FESEM results of composites with different CuO loading. Meanwhile, EDX scan result for 3% CuO/TiO_2 (Fig. 2d) was able to identify copper. The copper weight percentage obtained from 5 EDX point-scans shows that the mixing done during the synthesis process was not done thoroughly for 5% CuO/TiO_2, resulting in uneven spread of Cu in composite. A deviation from the intended CuO loading for every sample was also observed with 1% CuO/TiO_2, 3% CuO/TiO_2 and 5% CuO/TiO_2 having the average Cu weight percentage of 1.88%, 1.91% and 6.29%, respectively.

Electronic studies: The UV-Vis DRS spectra of composites with different CuO loading and their band gap values are shown in Fig. 3. It is observed that an increase in CuO loading results in an increase in the composites absorbance which shifts light absorption towards visible light (red-shifting). This can be explained as where the difference of average Cu weight percen-

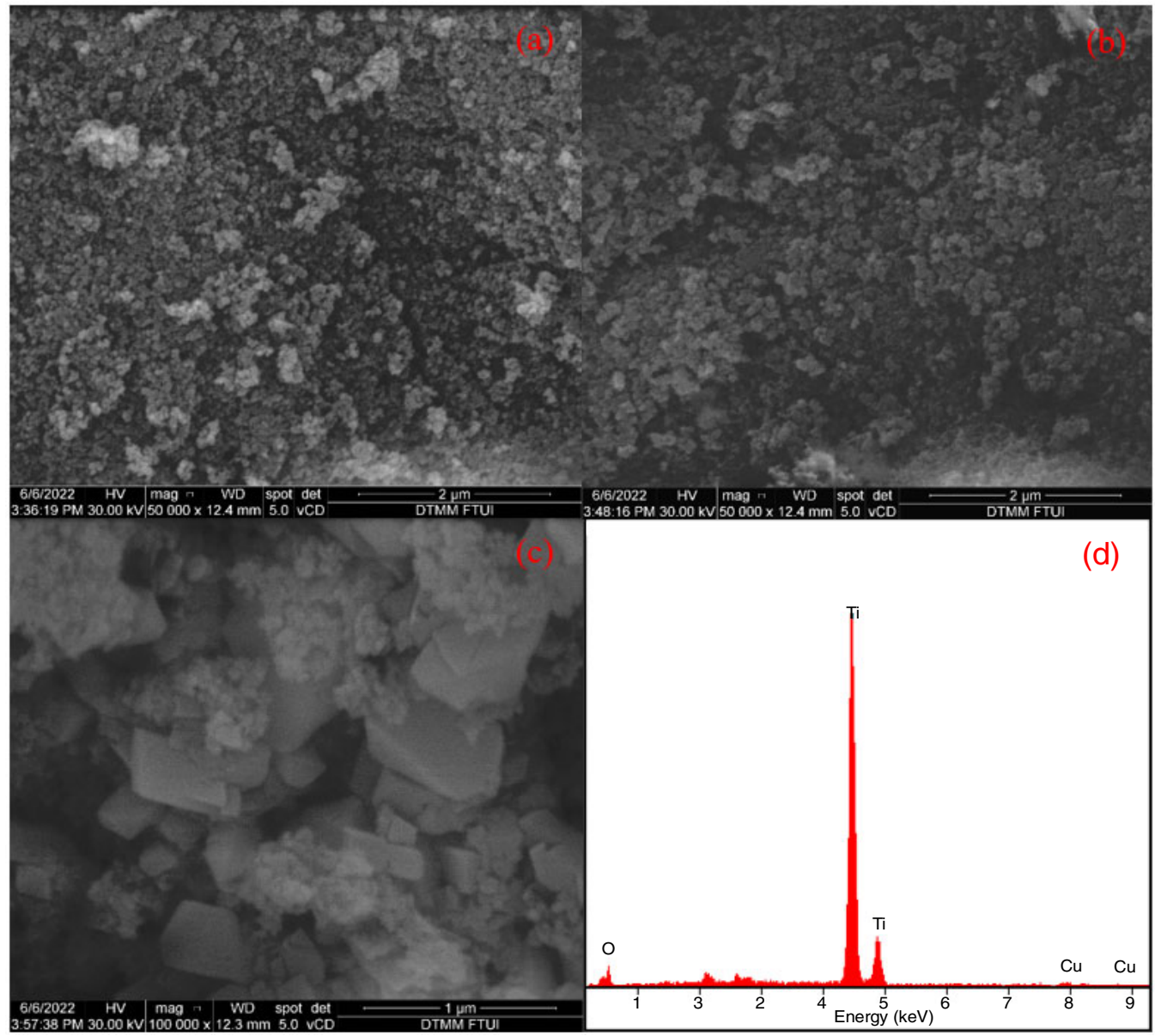

Fig. 2. FESEM-EDX characterization results (a) FESEM result for 1% CuO/TiO_2; (b) FESEM result for 3% CuO/TiO_2; (c) FESEM result for 5% CuO/TiO_2; (d) EDX result for 3% CuO/TiO_2

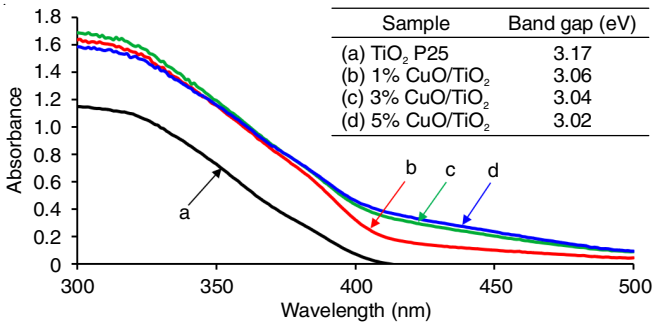

Fig. 3. UV-Vis DRS spectra for composites with different CuO loading and their band gap values

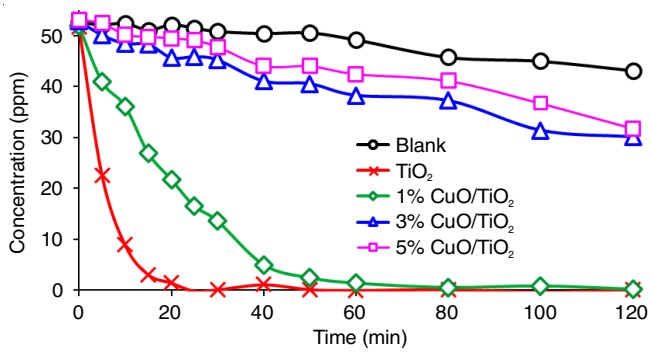

Fig. 5. Degradation of methylene blue

tage between 1% CuO/TiO_2 and 3% CuO/TiO_2 is very small (0.03%), however when the difference of average Cu weight percentage between 3% CuO/TiO_2 and 5% CuO/TiO_2 is more apparent, the distribution of Cu is very uneven on 5% CuO/TiO_2 indicated that not all Cu has been successfully doped. The same thing can be observed from the band gap values calculated using Kubelka-Munk equation where the decrease in band gap values is only significant when the loading of CuO does not exceed 1%.

Medical mask waste modification: The treatment which includes sonicating the MMW, dipping in 70% ethanol solution and exposing it to UV-C irradiation succeeded in removing contaminants in the MMW. The MMW modification was carried out using the dip coating method yields in 27-44 mg of composite coated on the surface of MMW. The successful modification procedure was also confirmed by the FTIR spectra (Fig. 4), where a new peak was formed because of the vibration of the Ti-O-Ti bond at 685 nm^{-1} on the modified MMW sample [16,17]. Several C-H peaks and a C-O peak indicated that MMW was made up of polypropylene and polyester. Other notable new peaks formed on the modified MMW sample at 1100 nm^{-1} formed by the vibration of Si-O-Si, which proved that TEOS was successfully hydrolyzed into SiO_2 and the weak signal at 938 nm^{-1} indicates that a Ti-O-Si bond was formed [18], which will increase the hydrophilicity and self-cleaning properties of MMW [19].

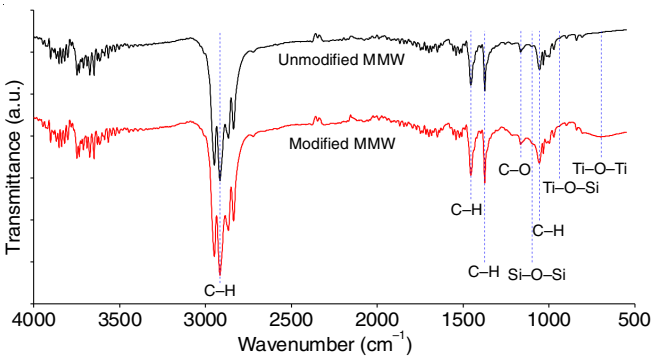

Fig. 4. FTIR spectra of an unmodified medical mask waste and a modified medical mask waste

Degradation studies: From Fig. 5, it is observed that 1% CuO/TiO_2 exhibit the best performance, which agreed with the reported works [20-22]. They explained that a lower loading of copper yields a better photocatalytic degradation performance, due to the high dispersion of CuO over TiO_2, which will effectively prevent the recombination of electron and hole, whereas at a higher loading of CuO will cause the shading effect due to the decrease of active sites of TiO_2, since they are covered with CuO crystals, resulting in the less ROS formation. This is detrimental to the experimental results, since in methylene blue degradation efficiency is dependent on the photocatalytic activity. The blank/unmodified MMW achieved a negligible %degradation compared to modified MMW showed that adsorption and photolysis didn't play a significant role in degrading methylene blue. Based on the degradation performance of modified MMW against methylene blue, the optimal loading of Cu is narrowed down into two options, 0% (undoped TiO_2) or 1%.

Bacteria disinfection: Bacterial disinfection investigation results against *S. aureus* are shown in Fig. 6. Among all the three samples, the antibacterial activity with the MMW sample modified by 1% CuO/TiO_2 achieved the best result. The addition of Cu managed to decrease the bacteria count by 12% because of its genotoxicity which enables Cu to damage the genetic make-up of the bacteria [23]. All the three samples showed a similar trend for *S. aureus* disinfection which starts with a decrease in bacteria count from 0 to 30 min due to adsorption in the solution into the MMW, then it was continued by an increase of bacteria count from 30 to 60 min due to the saturation of the adsorption sites with 1% CuO/TiO_2 being an exception due to the genotoxicity of Cu, from then onward a downtrend caused by photocatalytic activity. A decrease from

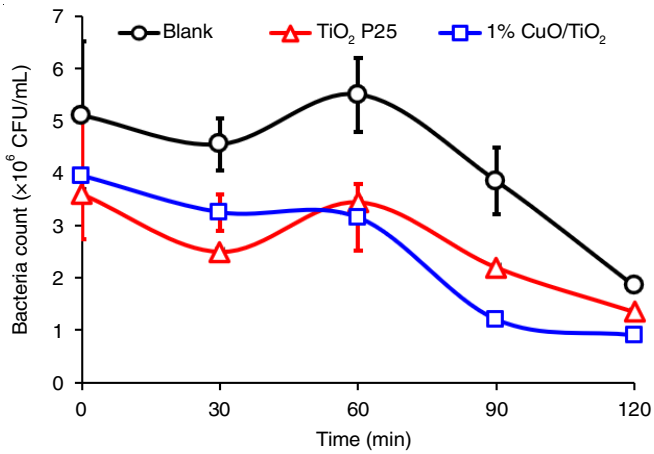

Fig. 6. Profile curve of *S. aureus* disinfection using medical mask waste as filter material

60 min to 120 min on the unmodified sample might have been caused by the photolysis from the germicidal UV-C lamp used since *S. aureus* is more vulnerable to physical treatments like UV-C irradiation due to its thinner cell wall [24].

Based on the bacteria disinfection test results, the optimum loading for Cu is 1% with the TiO_2 doped with a Cu loading of 1% having more or less the same performance in the methylene blue degradation test with the undoped TiO_2 and outperforming the undoped TiO_2 in the bacteria disinfection test.

Toxic gas degradation: The blank test was carried out without the purifying unit inside the test chamber resulted in the minimal change in the amount of tobacco smoke present in the test chamber and CO gas concentration which proved that the test chamber is free of leakage. Meanwhile, the results from the two tests (Figs. 7 and 8) showed the same trend when more components were activated, better results could be obtained since activating more components means including more process to degrade the existing pollutants in the test chamber. The best result for tobacco smoke degradation was achieved when the blower, UV-C lamp and plasma ion generator were activated with a result of 100% degradation achieved within > 15 min. Meanwhile for CO gas degradation, when all three components were activated, it took 30 min to degrade 30.8% CO gas in the test chamber. This result seems to be inefficient when compared with the results of Slamet & Ibadurrohman [8], where the purifying unit contained activated carbon as support, since TiO_2 managed to degrade 90% of CO gas in 10 min but this could also be caused by the difference in initial CO gas concentration where the initial CO gas concentration used in this research is 4 times higher than the previous one.

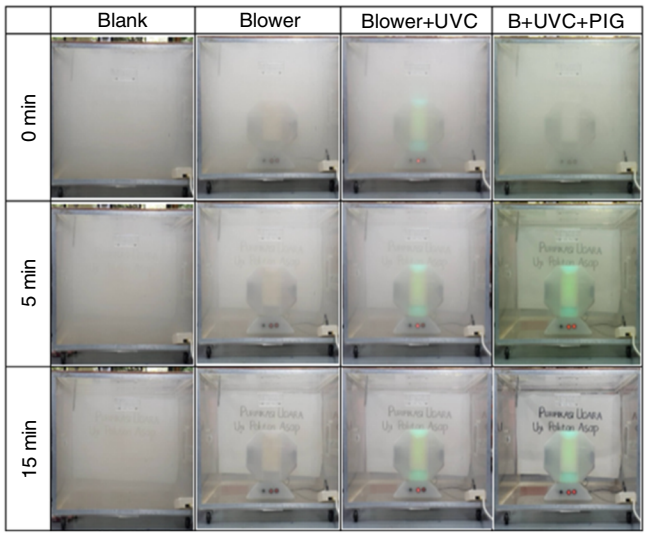

Fig. 7. Degradation of tobacco smoke

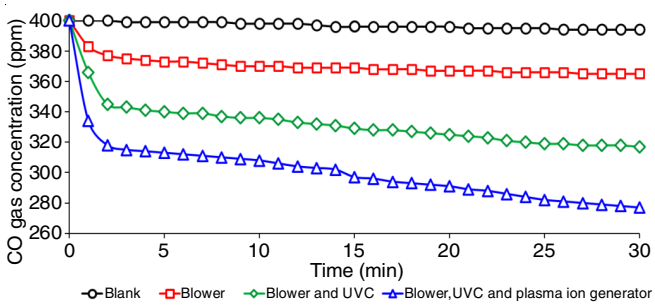

Fig. 8. degradation of CO gas

Conclusion

The CuO/TiO_2 composites were synthesized, characterized by FESEM-EDX, XRD, FTIR and UV-Vis DRS techniques and successfully applied in the medical mask wastes as filter material for air purification units. The modified MMW has succeeded in removing the contaminants which are existed in the MMW before the treatment. The optimum CuO loading based on the result of methylene blue degradation and bacteria disinfection results was 1%wt. of TiO_2. The prototype of the simple air purifying unit was able to degrade 100% tobacco smoke in less than 15 min and 30.8% CO gas in 30 min. Further production and cost-effective analyses are highly recommended to supplement the results, especially to calculate the potential implementation at the community level.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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