



A Review, Methods for removal and adsorption of volatile organic compounds from environmental matrixes

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ABSTRACT

The volatile organic compounds (VOCs) have toxic effects on human health and environmental matrices. So, determination and removal of VOCs from the environmental samples such as water, wastewater and air are very important as they exert toxic effects on human. Many chemical techniques such as; analytical methods for sorbents (extraction, adsorption), sole gel method, pervaporation, regenerative catalytic oxidation (RCO), recuperative catalytic oxidation (CO), adsorptive concentration-catalytic oxidation, photocatalytic oxidation (PCO), ozonation-catalytic oxidation and non-thermal plasma-catalytic oxidation, have been used for removal and reduction of VOCs from different matrices. This review study has been conducted to collect the adsorbents and applied chemistry methods which have been recently used in different works for the elimination of VOCs from air and water samples.

1. Introduction

Volatile organic compounds (VOCs) enter the environment through various sources, leaving severe environmental and health impacts [1]. The production of industrial wastewater is the main origin of VOCs which is considered as a negative aspect of industrial activity, exerting several adverse effects on the environment and human health [2-6]. Volatile organic compounds are one of the most widely used materials in the production of refrigerants, plastics, adhesives, paints, and petroleum products [3, 5, 7-12]. Benzene and toluene are regarded

as the most hazardous materials in the volatile organic compounds family and regarding their importance, a considerable amount of literature has been published on the issue of benzene and toluene potential adverse effects on health [13-16]. According to the international agency for research on cancer [17], benzene is categorized as group 1 (carcinogenic to humans) and Toluene as group 2B (quietly carcinogenic to humans). What we know about the adverse effects of benzene and Toluene is largely based on previous studies which have proved that the common adverse effects of these organic compounds is neurotoxicity including drowsiness, headache, tremor, coma and dizziness [16, 18-20]. Benzene exposure has been reported to increase the risk of various cancers including leukemia and hematopoietic cancers

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[15, 21]. It has been also recognized that benzene and Toluene affect skin, eyes and respiratory tract by inducing irritation [22-25]. Increased threatening impacts of VOCs increase health and environment concern and therefore, there is a crucial need to develop effective strategies to remove them. Several attempts have been made to eliminate or recover VOCs from wastewater; for example, distillation is commonly used, regardless of azeotropes formation or high energy consumption [6, 7, 10, 12, 26-29]. Hitherto, a number of approaches including adsorption [30-32], condensation [33], incineration [34, 35] and thermal oxidation [36] have been established to eliminate VOCs from the environment. Treatment methods that have been established for VOCs removal are as follows: air stripping, adsorption, advance oxidation, distillation, anaerobic/aerobic biological treatment and the technology of membrane [27, 29, 37-42]. Many researchers have also applied pervaporation with nonporous membranes such as silicon rubber-coated PP to remove aromatic compounds from water sources [43-45]. Moreover, a large body of studies has taken the advantage of membrane-based air stripping (MAS) process by the means of microporous hollow fiber contactors which is very effective in the treatment of aqueous effluents containing VOCs [46-56]. In the context of catalytic oxidation (one of the used techniques for VOCs removal), many researchers have focused on catalysts including noble metals (e.g. Al_2O_3 , TiO_2 , CeO_2 , MnOx), nonmetal oxides (e.g. SiO_2), zeolites (e.g. ZSM, MCM, NaY) [57-62] and carbon derivatives [63, 64]. However, metal oxides that are in charge of VOCs elimination are mostly derivatives of elements distributed in groups III-B through II-B of the periodic table such as Ti, Cu, Mn, Al, Ce, Co, Fe and so on [65-67]. Despite extensive attempts toward VOCs removal, conducted technologies have, to some extent, shortcomings and limitations. This paper aims to review techniques used for VOCs removal and discuss their advantages/disadvantages and finally, focus on introduced solutions to improve

the process of VOCs elimination.

2. Data sources

In this review, we used information such as journal articles, statistical data and conferences/seminars papers as our data source. Surfing in scientific websites and databases including Google Scholar and Web of Science was a major way of accessing valuable information and related articles. Therefore, it was important to search at least one of related key words which are included in either titles or abstracts of papers and are as follows: "VOCs removal", "Catalytic oxidation", "Adsorbents" and "Nanomaterials". It was also crucial for papers to contain one or more of the aforementioned keywords to be embodied in this review.

3. Experimental procedures and methods

3.1. Sorbent Methods

The analytical methods for sorbents based on extraction and adsorption (chemical and physical) were used for VOCs removal from water and wastewater samples. Recently, the phenyl sulfonic acid (PhSA) modified carbon nanotubes (CNTs) were presented for benzene removal (BR) from waters. For separation process, the PhSA@CNTs based on the dispersive micro solid phase extraction method (D- μ SPE) was used for BR from water. The main mechanism was achieved by the polar- π or π - π electron donor-acceptor interactions between the benzene and $\text{SO}_3\text{H}/\text{C}_6\text{H}_5$ group of CNTs surface (Fig. 1). According to the procedure, 10 mg of CNTs@PhSA nanostructures was added to 5 mL of water samples with different benzene standard solution (0.1--10 mg L^{-1}) in GC vial. After shaking and centrifuging (3500rpm), the CNTs@PhSA sorbent separated from water samples and finally, its concentration was determined by static head space gas chromatography mass spectrometry (SHS-GC-MS) [68]. The mechanism based on CNTs@PhSO₃H was obtained with π - π stacking between aromatic chain and S=O bond of CNTs@PhSO₃H (Fig. 2).

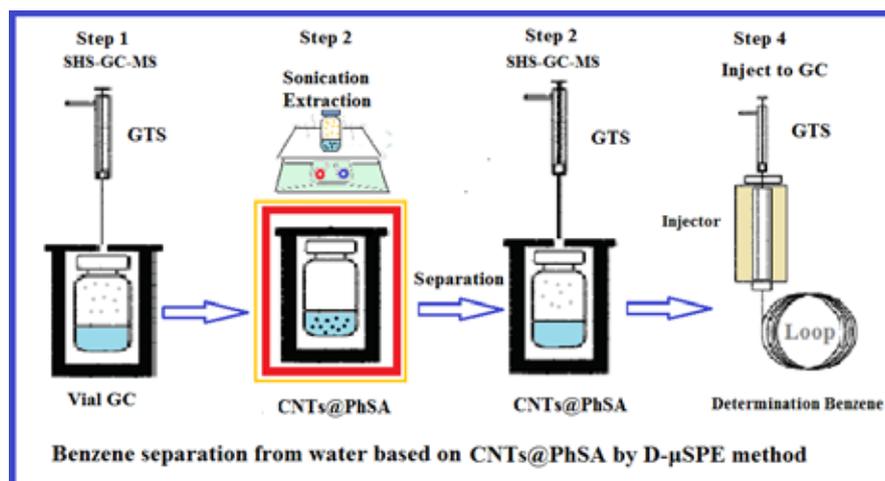


Fig. 1. Benzene extraction from waters based on CNTs@PhSA by D- μ -SPE method

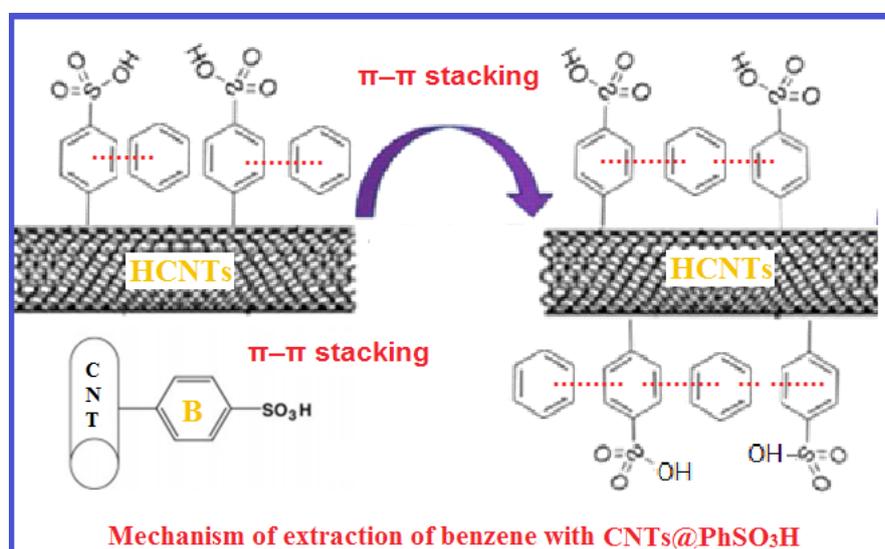


Fig. 2. Mechanism of extraction of benzene with CNTs@PhSO₃H

The graphene based materials (GBMs) have been also used as a new technology in different fields of science specially in the environmental chemistry. The GBMs adsorbent was used for the removal of VOCs with high adsorption capacity and cost-effectivity through various functionalization processes on the surface. The intermolecular forces of GBMs with the gaseous pollutants caused gas adsorption. The strength of the interaction of GBMs with VOCs depended on surface area/properties, pore volume/size of the GBMs. The GBMs showed the excellent adsorption for the removal of VOCs. Among the different graphene structures, GO and rGO have mostly used for the VOC removal from waters [69]. The various models

of the GO structures are shown in Figure 3 which was used for VOCs removal in different matrices. Hofmann and Holst first introduced graphite oxide and then Ruess et al designed a graphite oxide structure based on a wrinkled carbon sheet. Scholz and Boehm showed that the carbon sheet replaced by carbonyl and hydroxyl. Nakajima and Matsuo used two carbon layers linked to each other by sp_3 CeC bonds with carbonyl and hydroxyl groups. Lerf et al suggested a graphite oxide based on the unoxidized rings of benzene and that a wrinkled region of alicyclic 6-membered ring ethers is distributed randomly in a flat aromatic region. Szabó et al., showed a carbon network structure [70-73].

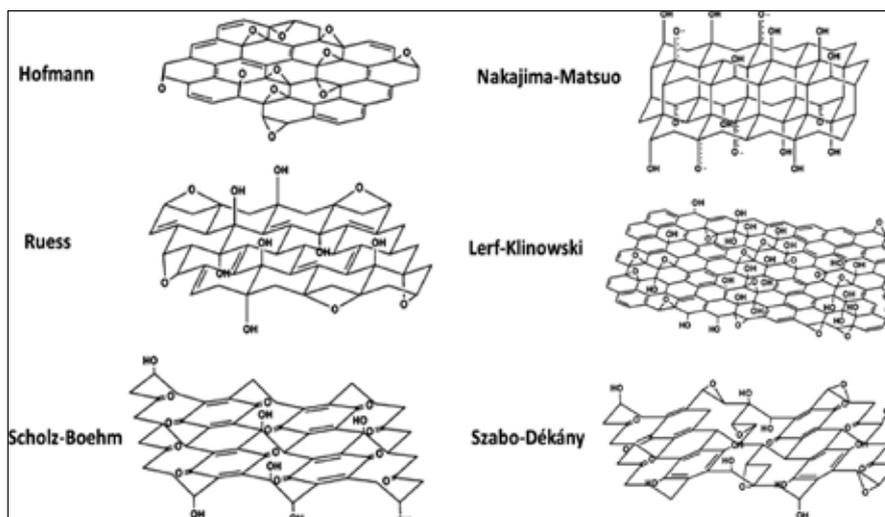


Fig. 3. Various models of the GO structure [70].

benzene, toluene, and xylene (BTX) are the major members of VOCs pollutions. These VOCs are preferably adsorbed on hydrophobic surfaces as compared to hydrophilic surfaces [74-75]. The previously results showed, the GO may exhibit less adsorption capacity for aromatic VOCs. Yu et al showed for 50 ppm benzene, the adsorption capacities for GO and rGO were obtained 216.2 and 276.4 mg g⁻¹, respectively. The rGO sorbent has a hydrophobic property with enhanced tendency (π - π bonds), increasing the adsorption capacity of VOCs relative to GO sorbents. In addition, the surface areas for rGO and GO were achieved 292.6 m² g⁻¹ and 236.4 m² g⁻¹, respectively and due to high surface area of rGO, the adsorption capacity of VOCs was increased [69, 74, 75]. Szczeńniak et al showed that the high surface area of GBMs and GO caused to improve the benzene adsorption capacities for OMC. The incorporation of GO with OMC / KOH increased the surface area from 740 m² g⁻¹ to 1370 m² g⁻¹. Also, the pore volume of OMC from 0.61 increased to 1.06 cm³ g⁻¹ after the formation of GO/OMC. Finally, adsorption capacities for benzene were obtained 633 mg g⁻¹ and 750 mg g⁻¹ for OMC and GO/OMC, respectively [76-77]. GO - MOF-5 was used for the removal of benzene vapor from air with capacity of 251 mg g⁻¹ [78]. Due to high porosity of MOFs, they cannot retain

small molecules under ambient conditions. So, the GO/MOF-5 composite was prepared using varying proportions of GO, such as 1.75 wt%, 3.5 wt%, 5.25 wt%, and 7 wt% (Fig 4).

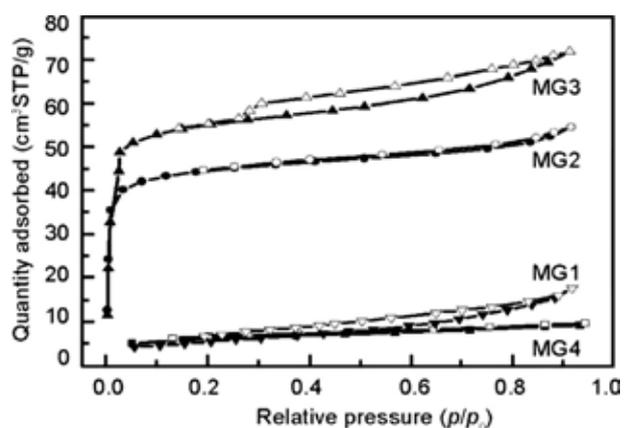


Fig. 4. Adsorption-desorption isotherms for graphene oxide/metal organic framework-5 (GO/MOF-5) for benzene [78]. MG (1–4) represents GO/MOF-5 composite with 1.75%, 3.5%, 5.25%, and 7% of GO in MOF-5.

The GO and rGO was used for the removal of toluene by Kim et al. The π - π bonds, hydrophobic and electrostatic interaction with toluene led to the absorption of toluene on GO / rGO surfaces (Fig. 5). Different types of GBMs such as graphene platelets (GP), rGOMW, and KOH activated rGOMW (rGOMWKOH) were analyzed for the toluene adsorption [79].

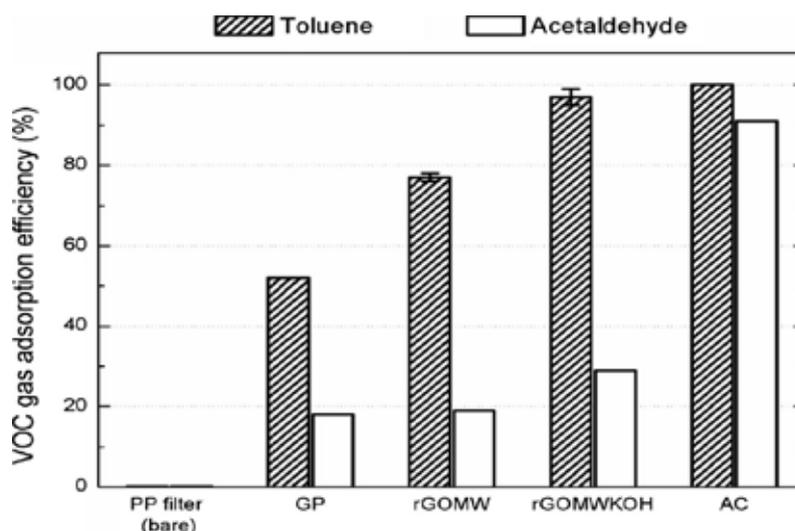


Fig. 5. Toluene and acetaldehyde removal capacity by rGOMWKOH compared to other adsorbents [81].

Aldehyde and ketone compounds are the most carbonyl VOCs which can be considered as sources of the environment contamination. GBMs can efficiently remove the carbonyl compounds from environment. For indoor formaldehyde removal, amino functionalized graphene sponge (G/S) or G/S decorated with graphene nanodots (G-GND/S) were used by Wu et al. G-GND/S with high amine groups on surface as compared to G/S caused a high interaction with formaldehyde molecules (Fig 6). The results showed that

the adsorption capacity of GGND/S and G/S were achieved 22.8 mg/g^{-1} and 7.5 mg/g^{-1} for formaldehyde, respectively [80].

Lim et al. prepared mesoporous-structured graphene powder through the method of thermal expansion (Fig. 7) and used them as adsorbents for removing VOCs. According to Figure 8 the characteristics and morphology of the prepared adsorbent was defined through different methods including scanning electron microscopy (SEM), X-ray photoelectron spectroscopy and N_2 isotherms. Adsorption

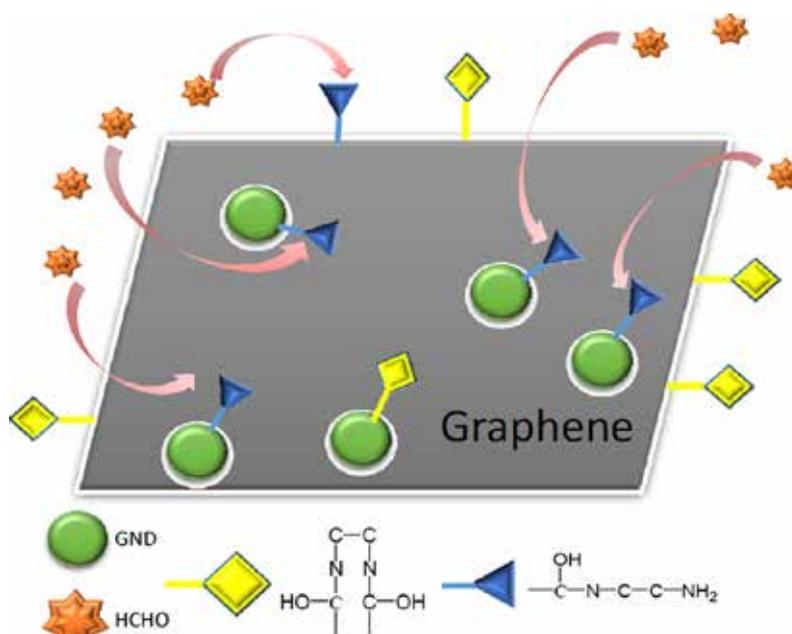


Fig. 6. Interaction of amino graphene nanodots decorated functionalized graphene sponge with formaldehyde molecules

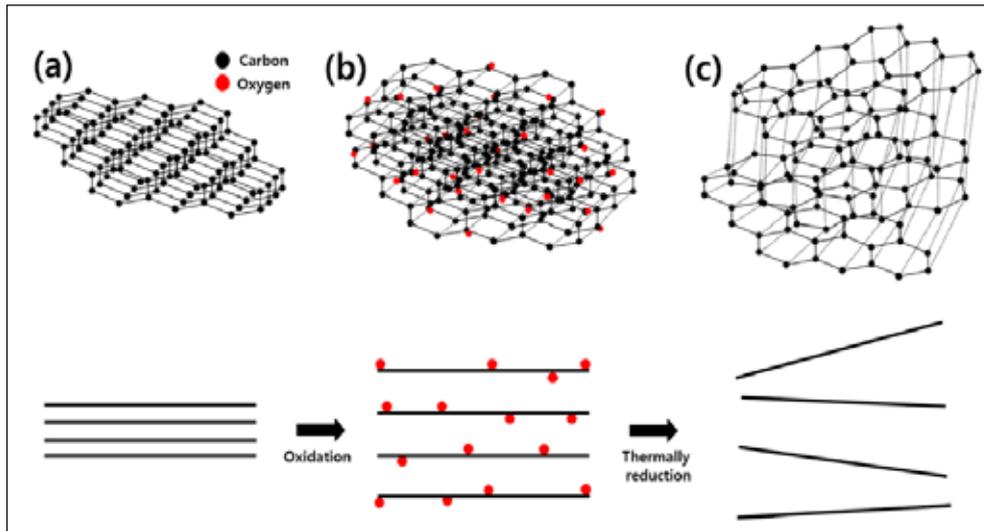


Fig. 7. The scheme of thermal expansion mechanism; a: graphite powder b: GO powder c: TEGP [82].

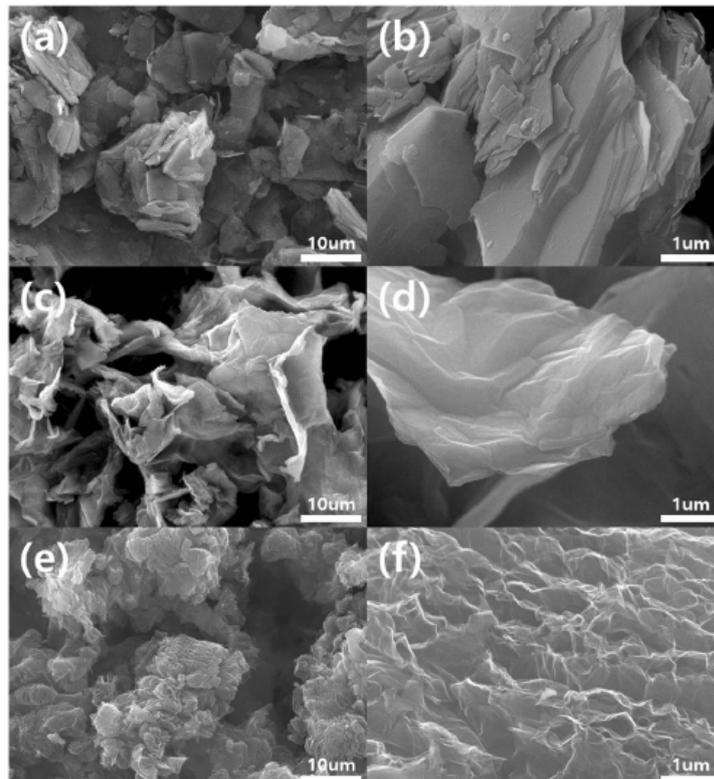


Fig. 8. SEM images of graphite powder (a,b), graphite oxide (GO) (c,d) and TEGP (e,f) [82].

capacity of graphene powder was examined using propylene filter, at a concentration range of VOCs (30, 50, 100 ppm). The results of the study indicated that thermal expanded graphene powder (TEGP) is an effective material for VOCs removal which acts in a proper chemical oxidation using heat energy. It was also reported that TEGP is of economical

materials, due to its reusability [82].

3.2. Sole Gel Method

The sole-gel process is a method for producing solid materials from small molecules. As a chemical-wet technique, recently, it has been widely used in the fields of material sciences and ceramic engineering. This kind of methods are employed

primarily for the synthesis of materials (mostly metal oxides) starting with a chemical solution that acts as a precursor for an integrated nexus of both segregated particle and network polymer [83].

Metal alkoxides and metal chlorides, which are considered as frequent precursors, undergo different forms of hydrolysis and poly-condensation reactions. Binding of metal centers with either oxo (M-O-M) or hydroxo (M-OH-M) bridges leads to the formation of metal oxides and generates metal-oxo or metal-hydroxo polymers in solution. Therefore, the sol involves in the formation of a gel-like diphasic system containing both solid and liquid with segregated particles to a continuous polymer structure morphologies [83].

The sole-gel method has been increasingly applied for the development of various materials including material for catalysis [84, 85], chemical sensors [86, 87], optical gain media [88], solid state electrochemical devices [88], photochromic and non-linear applications [90-92], membranes [93] and fibers [94]. One of the intriguing applications of the sole-gel technology is photo catalyst preparation. Photo catalysts have been widely used to degenerate VOCs. Common photo catalysts are

semiconductors like ZnO, GaP, TiO₂, SiC, CdS and Fe₂O₃ [95]. Among these photo catalysts, TiO₂ is the most applicable photo catalyst in the context of eliminating environmental pollutants because of its chemical stability, high oxidizing potential, low cost, non-toxicity and environmentally friendly properties [96-99]. Parvizi et al. in their study about perovskite nano-catalysts, synthesized a series of La_{1-x}A_xMnO₃ (A: Co, Zn, Mg, Ba) through sol-gel method (Fig. 9) and then evaluated the performance of these catalysts in the elimination process of BTX compounds. After conducting the research, the FTIR results showed that all characteristics related to efficient catalyst was present in synthesized catalyst, indicating acceptable outcomes of sol-gel method [100]. By applying nonhydrolytic sol-gel method, Debecker et al. synthesized V₂O₅-TiO₂ and added Mo and W oxides to promote the catalysis performance of catalyst (Fig. 10). The results of the research indicated a significantly better oxidation performance (93% oxidation) and a highly efficient action for CO₂ selectivity for the purpose of VOCs removal [101]. These results imply the importance of sol-gel method in the process of efficient catalyst synthesis.

Sarafray Yazdi et al. developed a novel fiber

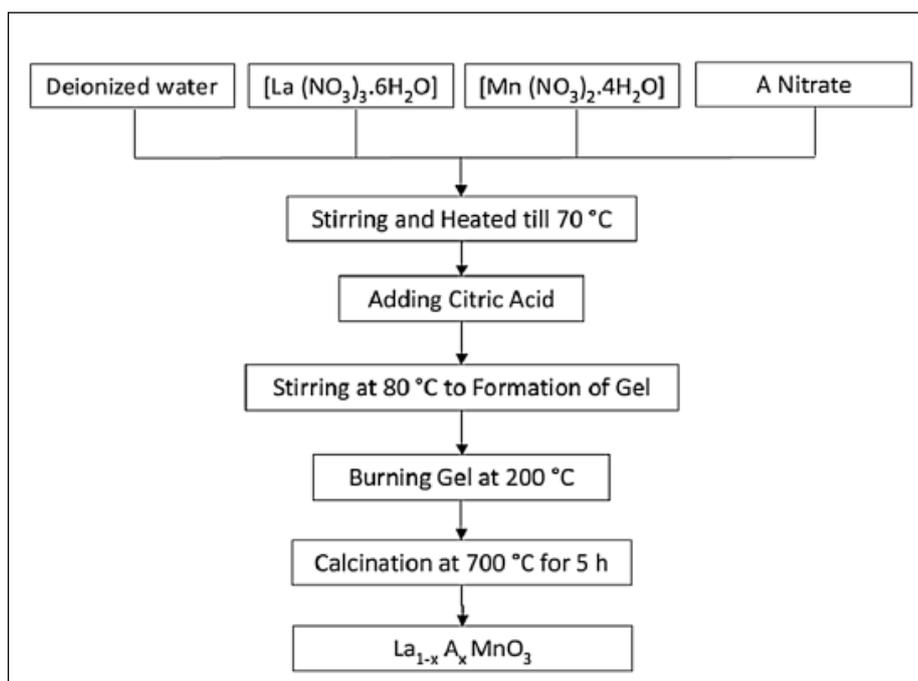


Fig 9. Schematic procedure of La_{1-x}A_xMnO₃ (A: Co, Zn, Mg, Ba) perovskite nano-catalyst preparation by sol-gel method [100].

to improve the elimination of trace amounts of BTEX. At first, poly ethylene glycol (PEG) grafted on multi-walled carbon nanotubes (PEG-g-MWCNTs) undergone a chemical bonding with sol-gel to produce the unique fiber, as shown in Figure 11. The results showed that the porous structure, thermal stability, potent selectivity and durability of mentioned fiber lead to a remarkably better performance in the route of removing BTEX. Also, due to porous structure of sol-gel coating, the surface area of fiber, extraction velocity, steps of desorption and capacity of sample loading increase significantly [102].

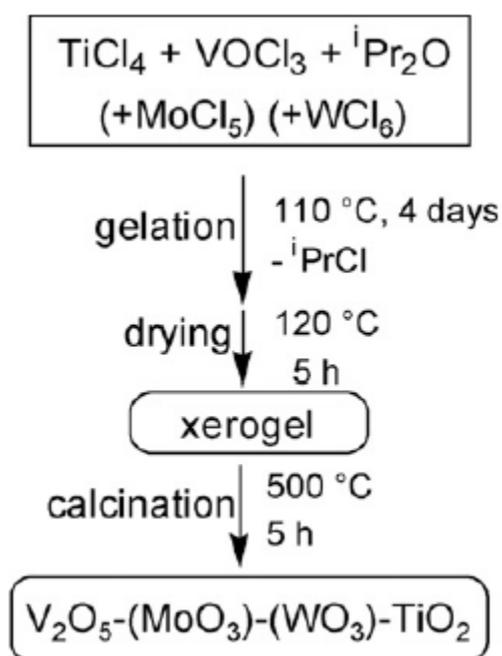


Fig. 10. Nonhydrolytic sol-gel preparation method [101]

3.3. Pervaporation

Membrane-based pervaporation (PV) technology serves as an economical and alternative technique in the organic-organic separation processes. A number of researchers have reported the elimination of VOCs from water sources which has been achieved through various polymeric membranes, using pervaporation technology [103-111]. Uragami et.al prepared a PVC membrane (hydrophobic polymeric membrane) and used an ionic liquid (1-allyl-3-butylimidazolium bis (trifluoromethane sulfonyl) imid ([ABIM]TFIS)) with a remarkable and low affinity for VOCs and water, respectively. Through the process of PV, Uragami and coworkers evaluated the performance of prepared [ABIM] [TFIS] /PVC aqueous solutions of dilute benzene and reported that the combination of PVC membranes with [ABIM] [TFIS] ionic liquid represents higher permeability and benzene/water selectivity in a concentration dependent manner. As shown in Figure 12, it was also revealed that incorporation of PDMS component decreases the density of membrane and induces benzene permeability [112]. Kujawa et al. functionalized and increased the hydrophobicity of two types of ceramic membranes with molecular sizes of 5 kDa and 300 kDa (Fig. 13). They reported that membranes with increased hydrophobicity can efficiently remove VOCs from binary aqueous solutions through vacuum membrane distillation procedure [113].

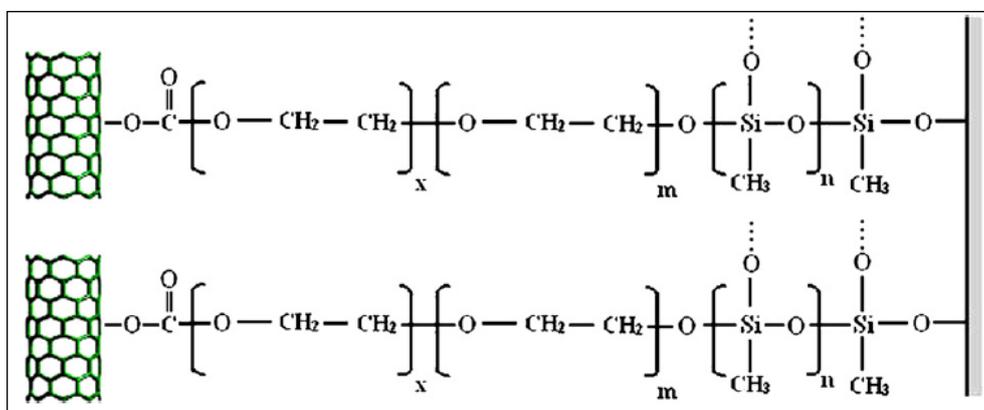


Fig. 11. Sol-gel PEG-g-MWCNTs coating [102]

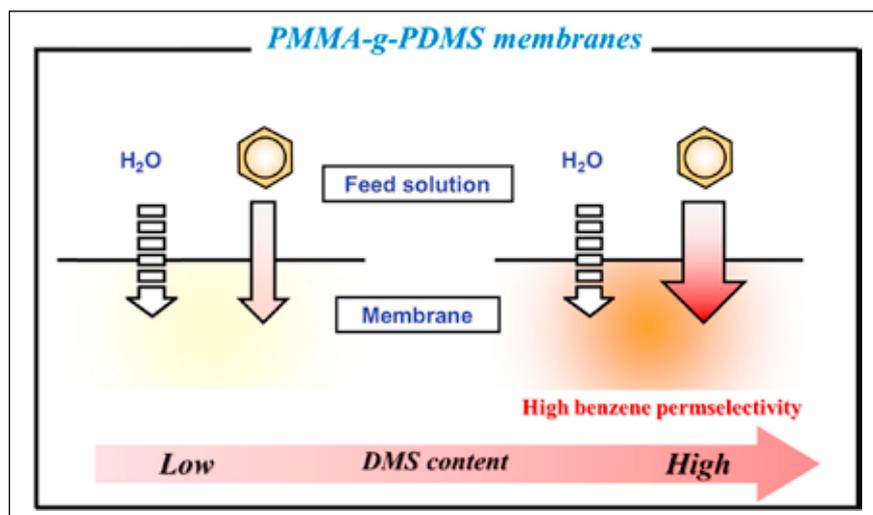


Fig. 12. The illustration of benzene permselectivity and benzene permeability under the effect of PMMA—PDMS membranes [112].

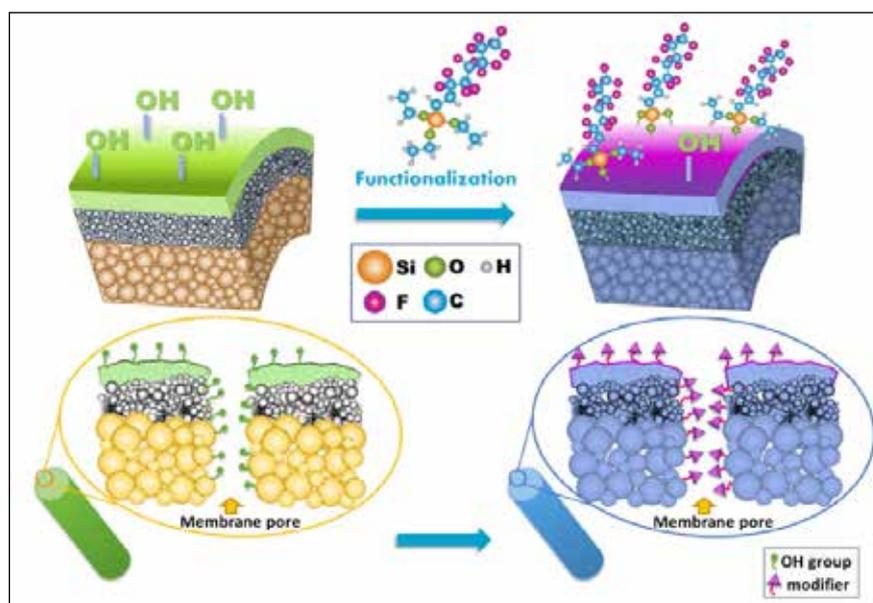


Fig. 13. Ceramic membrane's functionalization by perfluoroalkylsilanes [113].

3.4. Catalytic oxidation

3.4.1. Regenerative catalytic oxidation (RCO)

In parallel with the regenerative thermal oxidation (RTO), RCO is one of the most energy-saving techniques, being as relatively similar working mechanism as RTO. These techniques contain two or more beds with random or structured ceramic packs which are of high specific heat materials ($800\text{--}1000\text{ J Kg}^{-1}\text{ K}^{-1}$) and perform as heat transfer media. As shown in Figure 14, frequently used two bed RCO usually contains ceramic layer, catalyst layer, natural gas burner or electrical heater which plays a

role as heat storage, reaction media and heat supply, respectively. Due to its relatively lower price, natural gas is preferred to use rather than electrical heating. The mechanism of VOCs removal in this technique involves passage of VOCs flow from ceramics cabinet A which preheats VOCs, followed by a temperature increase up to $200\text{--}300\text{ }^{\circ}\text{K}$. When heater keeps the flow temperature of catalyst (i.e., TChamber) higher than the light-off temperature (TC), effective degradation of VOCs occurs. Simultaneously, the releasing heat from VOCs oxidation contributes to TChamber and even can

serve as a usable heat. In the next step, reacted flow goes down and its heat reserves as a high specific heat, preheating the inlet VOCs flow in the next cabinet B to A cycle. Thermal Recovery Efficiency (η) is a factor evaluating energy-efficiency related properties of oxidizing equipment. Although the η of RTO is up to 90%, the η of RCO can reach 95% or higher [114]. Liu et al. designed a formula of Ru-5M/TiO₂ (M: Mn, Co, Ce, Cu, Fe) for ruthenium-based bimetallic catalyst and examined its effect on benzene oxidation efficiency. After deep examination of different bimetallic species, it was proved that the combination of Ru-5Co/ TiO₂ can be the most effective species for the process of benzene removal. Figure 15 illustrates the represented mechanism of benzene removal, using combined Ru-5Co/ TiO₂ catalyst [115]. In the study of Zhang et al., nano-crystalline copper-manganese oxides were prepared using sol-gel method. The relativity between Cu and Mn was defined as Cu_{3x}-Mn_x (x can be equal to 0, 1, 1.5, 2, 2.5, 3 and is a representation of molar ratio of Cu and Mn); the optimal ratio was also found to be 2. Results showed that CuMn₂ with the spinel structure of CuMn₂O₄ exhibits a larger surface area, smaller pore size and network oxygen species, leading to enhanced catalytic activity of CuMn₂ which can be the result of stabilized CuMn₂O₄ active sites and synergistic effect between Cu and Mn (Fig. 16) [116].

3.4.2. Recuperative catalytic oxidation (CO)

Recuperative catalytic oxidation (CO), known as a simplified version of RCO, is a technique consisting of tubular or plate heat exchanger instead of regenerative thermal ceramic layers in RCO. In this technique, at first, a heat exchanger preheats VOCs flow, causing a temperature increase by about 50-200°K. Then, the next heater further heats the flow up to the light-off temperature of catalyst (usually above

573 K). Finally, reinforcement of VOCs oxidation occurs to produce CO₂ and H₂O with a significant amount of heat release. The η of a normal CO is generally lower than 70%, indicating that the customer should cost more than usual to obtain required energy for keeping the equipment to work

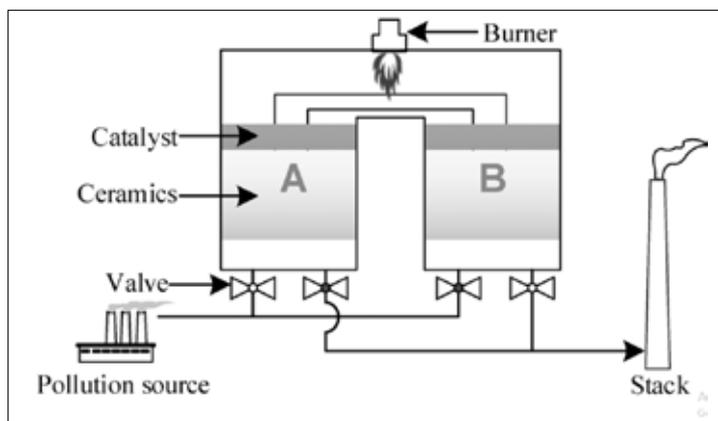


Fig. 14. The schematic diagram of regenerative catalytic oxidizer [114].

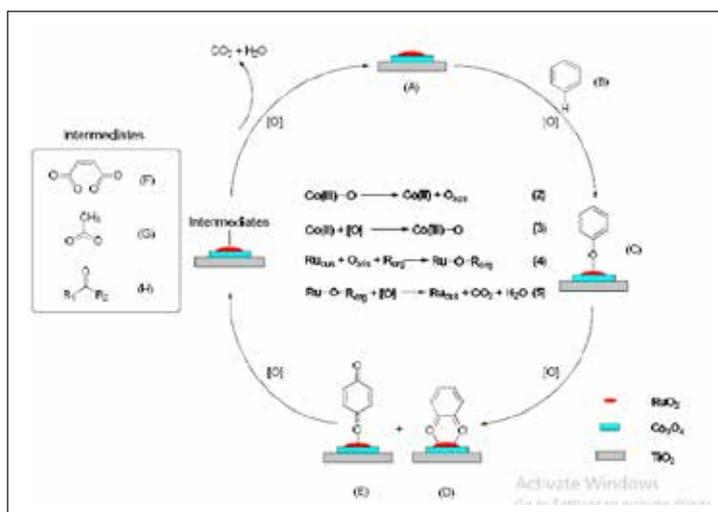


Fig. 15. Presented mechanism of the effect of Ru-5Co/ TiO₂ on the benzene oxidation [115].

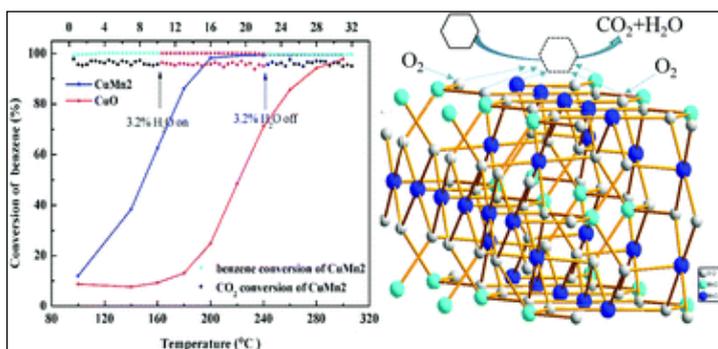


Fig. 16. The sample of CuMn₂ and relative spinel structure [116].

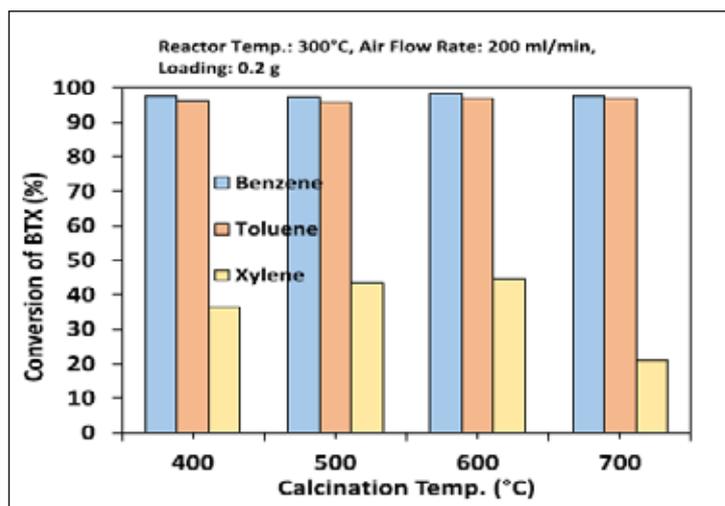


Fig. 17. The effect of manganese oxide calcination temperature on the thermal oxidation of BTX [118].

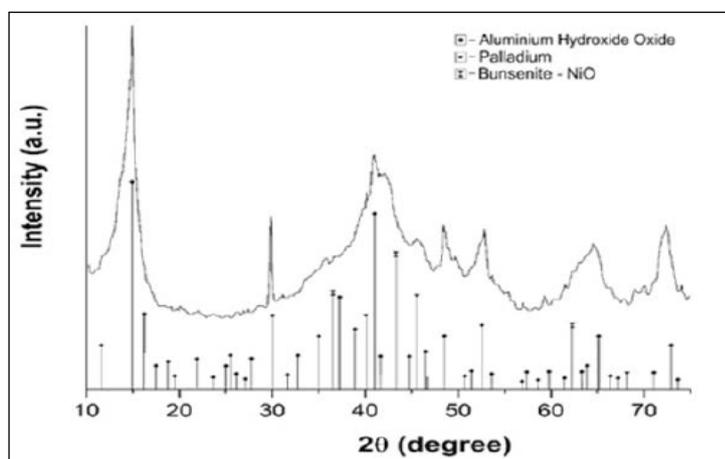


Fig. 18. The XRD pattern of the Ni/Pd/AlOOH [119].

on VOCs elimination. In fact, CO can't be effective for large-scale and low concentrations of VOCs pollutant removal [117]. However, considering low initial investment and high flexibility, the technique of CO can be effective for samples with small flowrate ($<5000 \text{ m}^3 \text{ h}^{-1}$) of VOCs pollutant [114]. Intriguingly, it is important to note that in the case of large-volume and low concentration VOCs emissions, there is an advanced technology named adsorptive concentration-catalytic oxidation which makes it possible to remove such VOCs samples. Hoseini and coworkers aimed to synthesize manganese oxide and impregnate it into different loadings of alumina. They utilized the resulted material in the procedure of BTX

oxidation followed by performance analysis. Findings of the study represented the most effective morphology and higher surface area at 10% of alumina loading. Further evaluation highlighted that the best condition for oxidation is 10 kV, 0.2 g Mn10Al in the 200 mL min^{-1} flowrate of pollutant with resulting oxidation of 97, 99 and 74% benzene, toluene and xylene, respectively. The catalytic activity of catalyst was investigated by catalyst calcination at four different temperatures (400, 500, 600, 700 °C). As Figure 17 it can be seen that the highest level of BTX conversion was found to be in Mn600 catalyst [118]. Georgiev in 2019 investigated ozone-assisted catalytic oxidation of benzene through alumina, silica and boehmite-supported Ni/Pd catalysts in 353 K. Three bimetallic Ni/Pd samples in a nano scale were synthesized with loadings of 4.7% Ni, 0.17% Pd supported on SiO_2 , AlOOH and Al_2O_3 and by the means of extractive-pyrolytic method. According to the results of the study, the highest steady-state activity of catalysts was attributed to Ni/Pd/AlOOH catalyst (Fig. 18). Georgiev reported that this activity is dependent on the amount of ozone decomposition capacity of catalysts which leads to oxidative species production; a sample with a high ozone decomposition ability (related to surface area of support) is capable of benzene oxidation in a high extent [119].

3.4.3. Photocatalytic oxidation (PCO)

As a different and distinguishable technology, photocatalytic oxidation has received considerable attention, due to its mild reaction condition and non-selectivity. By using UV or visible light in the environment temperature, photocatalytic oxidation works different from thermal catalysis; therefore, compared to RCO and CO, the configuration of PCO is simpler (Fig. 19). Photocatalysis has a wide variety of activities regarding various VOCs at the environment temperature, however, due

to low quantum efficiency and long residence time requirement, it has a limited oxidizing power and load adaptability [114]. Zhang et al. [120] introduced a new modified photocatalyst named $\text{TiO}_2\text{-UiO-66-NH}_2$ (constituting from the combination of TiO_2 and UiO-66-NH_2) and reported that the new photocatalyst can significantly improve photocatalytic performance for VOCs oxidation (Fig. 20). According to this study, the $\text{TiO}_2\text{-UN}$ photocatalytic system, represented good CO_2 selectivity and high photocatalytic activity with 72.70 % of toluene decomposition during 240 min of reaction, which was even higher than single TiO_2 (44.22 %) and UiO-66-NH_2 (7.48 %). In the study of benzene removal by the means of

PCO technology, Ji J et al proposed that VUV-PCO technique (Fig. 21) is significantly effective in comparison with the ordinary UV-PCO [121]. In contrast to UV-PCO in which benzene degradation is only attributed to photocatalytic oxidation, VUV-PCO technique consists of several decomposition pathways alongside VUV photolysis and PCO. Benzene degradation hardly occurs under the effect of UV irradiation [122], however, VUV irradiation have a benzene removal efficiency of about 48% radiation potency in the process of benzene degradation is related to the formation of hydroxyl radicals ($\cdot\text{OH}$) and oxygen species such as $\text{O}(\text{1D})$, $\text{O}(\text{3P})$ and O_3 [123].

3.5. Ozonation-catalytic oxidation

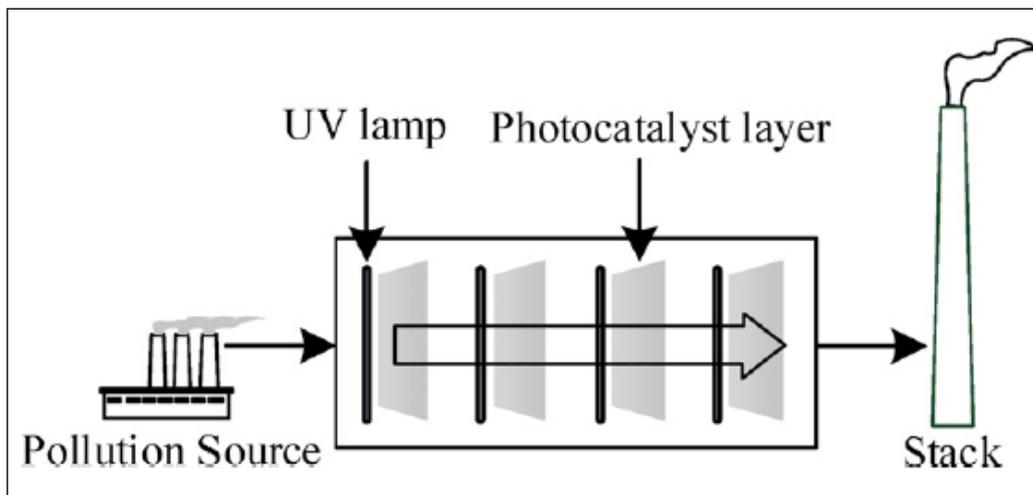


Fig. 19. Schematic configuration of PCO [114].

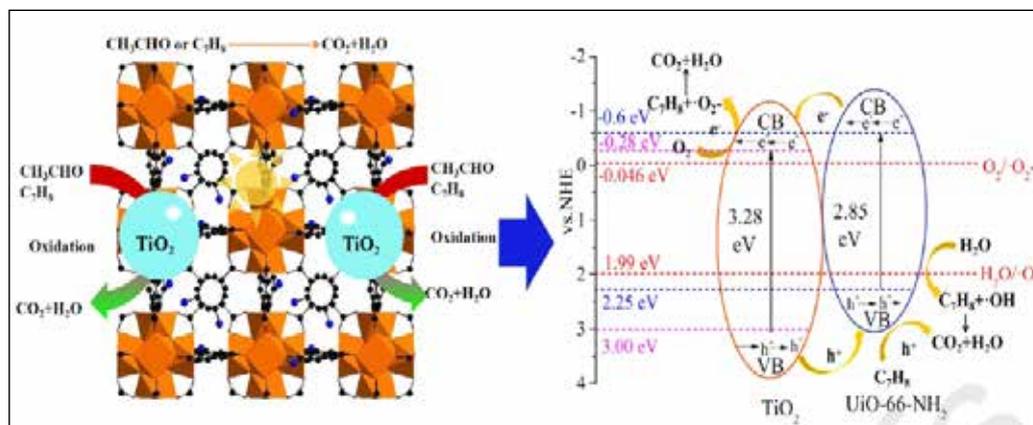


Fig. 20. Hypothesized pathways for photocatalytic oxidation of VOCs by $\text{TiO}_2\text{-UiO-66-NH}_2$ [120].

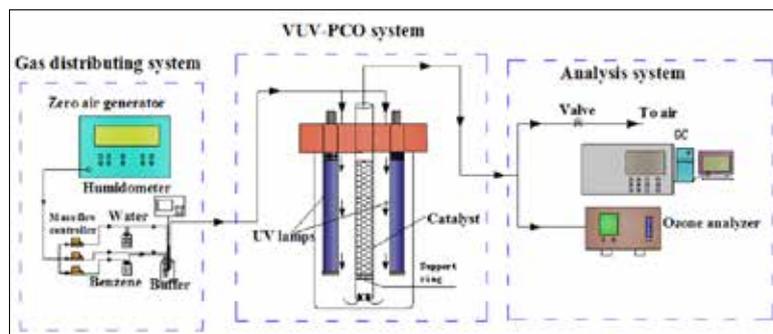


Fig. 21. The schematic diagram of VUV-PCO system [121].

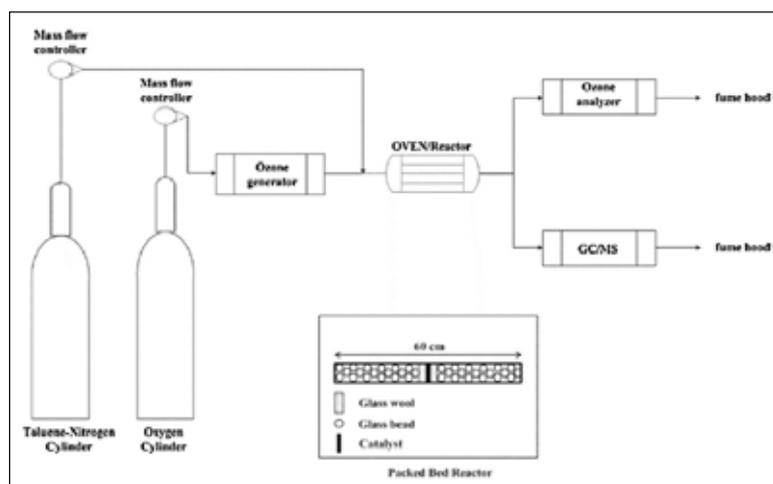


Fig. 22. The schem of experimental procedure [126].

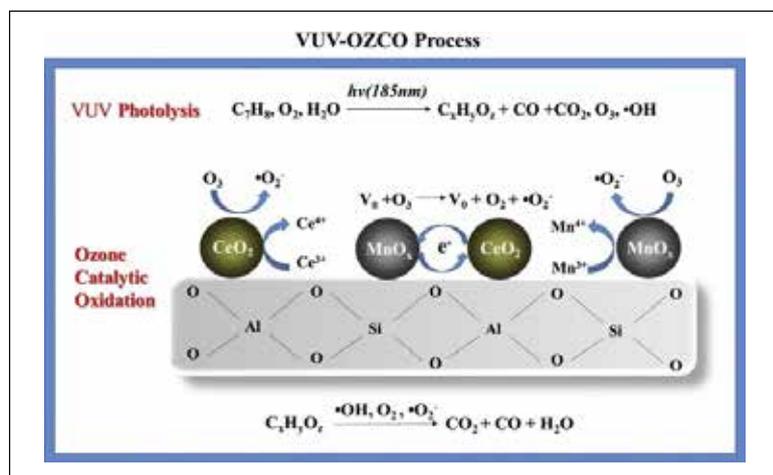


Fig. 23. VUV-OZCO process [125].

In the technology of ozonation-catalytic oxidation, Ozone (O_3), a strong oxidant with a standard redox potential at 2.07 eV, is used. Water sterilization and wastewater treatment procedures have extensively taken the advantages of ozone oxidation. Ozone

oxidation has been extensively applied in water sterilization and wastewater treatment procedures. Since ozone isn't very stable in gas environment, as a single technique, it can't be so effective in the oxidation of VOCs to CO_2 and H_2O . However, ozone oxidation has the capability of being used as a pre-treatment step before common catalytic techniques (e.g. thermal catalysis and photocatalysis) and promotes a synergistic effect alongside these technologies. Rezaei et al. in their 2013 study indicated that transition metal oxide-based catalysts propose efficient VOCs removal by catalytic ozonation, obviating the need for costly noble metals which are frequently used in the VOCs catalytic combustion with oxygen. The mentioned study investigated different loadings of Mn in a temperature range and reported that increased temperature leads to better activity of catalyst and lower loadings of Mn resulting in surged and efficient toluene oxidation (analyzed by GC-MASS) and ensuing ozone decomposition (measured by ozone analyzer) (Fig. 22) [124]. Shu et al. designed a novel process which was a combination of VUV photolysis and O_3 catalytic oxidation (VUV-OZCO). In this system, VOCs are firstly destructed by VUV and then are oxidized through VUV-generated O_3 in the presence of catalyst. O_3 by-product is also eliminated simultaneously. In the study of Shu et al. it was revealed that the novel Mn-xCe-ZSM-5 catalyst

along with VUV-OZCO system (Fig. 23) can have the capacity to simultaneously decompose O_3 by-product and improve toluene removal efficiency [125].

3.6. Non-thermal plasma-catalytic oxidation

Non-thermal plasma (NTP) which has been introduced as green technology for elimination of VOCs from indoor and industrial gas streams, is a superior source of chemically active species (OH and O^2 radicals, ions, excited species, etc). This property leads non-thermal plasma to provide a highly reactive environment (caused by electron's acceleration, dissociation and ionization); without any energy consumption on heating the entire gas stream, in which reactive species oxidize various VOCs molecules and consequently degrade them. Besides low energy efficiency and inferior CO_2 selectivity, a major difficulty with non-thermal plasma is production of by-products including NO_x , O_3 and other intermediates relating to the fact that electrons do not have enough energy to mineralize BTX molecules [127]. Thus, non-thermal plasma oxidation can't be considered as a single technique to deal with VOCs pollution, because products of uncompleted reactions in this technique can act as secondary pollutions on their own.

A great solution for the aforementioned problem is combining the non-thermal plasma oxidation with catalysis (Fig. 24). This combination has been extensively investigated during the last decade, indicating that the system of non-thermal plasma-catalysis is obviously capable of improving energy efficiency and suppressing unwanted by-products in the process of VOCs degradation [128]. Non-thermal plasma catalysis system consists of two configurations: in-plasma catalysis (IPC) and post-plasma catalysis (PPC). Guo et al. investigated the efficiency of NTP+ MnO_x catalyst in the elimination of benzene and showed

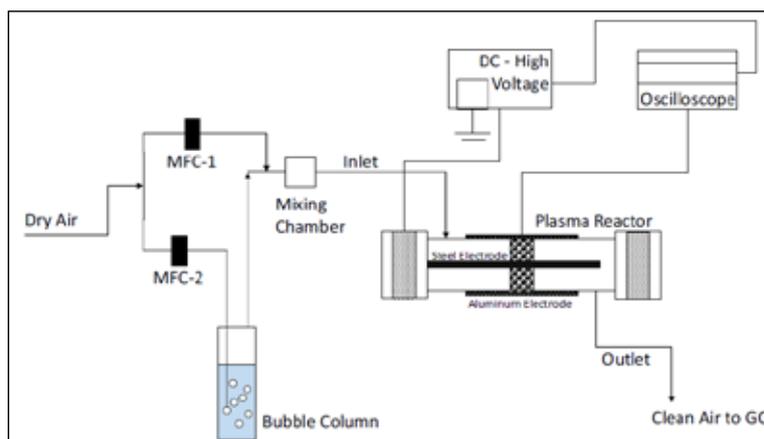


Fig. 24. Experimental setup of hybrid plasma-catalytic system for oxidation of VOCs [115].

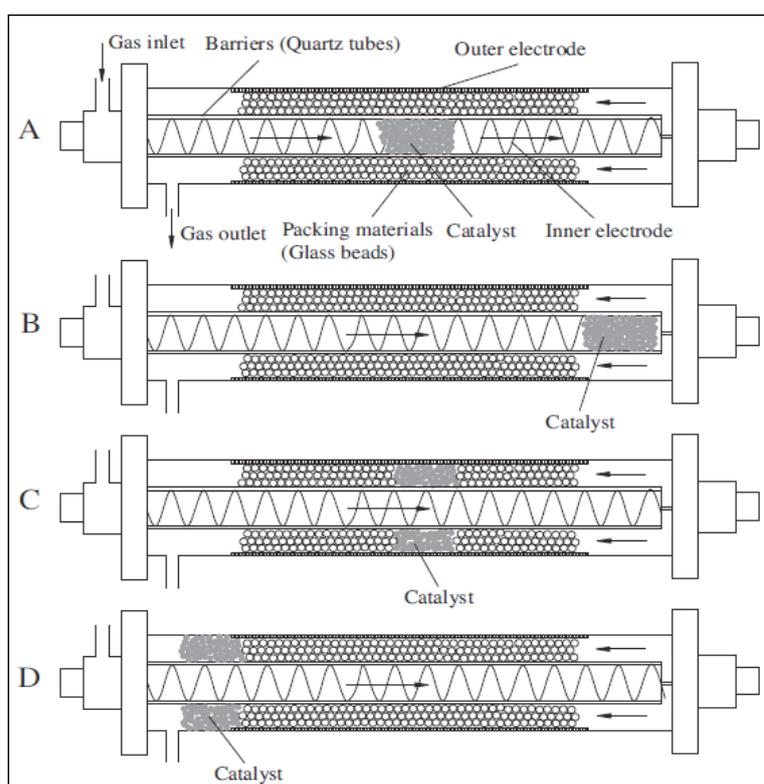


Fig. 25. Different plasma-catalysis systems in IPC and PPC configurations: (a) catalysts in the region I, (b) catalysts in the downstream of region I, (c) catalysts in the region II, (d) catalysts in the downstream of region II [130].

that this combination can significantly improve benzene removal efficiency and promote CO_2 formation, with a simultaneously suppression of CO [129].

In the study of Jiang et al. both IPC and PPC

configurations of NTP (Fig. 25) constituted from a hybrid surface/packed bed discharge (HSPBD) with different catalysts including $\text{Ag}_x \text{Ce}_{1-x/\gamma} \text{-Al}_2\text{O}_3$ was applied. From the result of the study, it was revealed that through the plasma-catalysis system and present of $\text{Ag}_x \text{Ce}_{1-x/\gamma} \text{-Al}_2\text{O}_3$ catalyst significantly enhance benzene degradation and improve CO_x selectivity. The study of Jiang also showed that the PPC process has a better effect on the decomposition of O_3 and benzene [130].

3.7. Nanotechnology

Nanotechnology is an increasingly important area of the recent technology, playing a cardinal role in a bunch of fields. Among the various subtypes of this technology, carbon nanotubes (CNTs) has attracted a great deal of interest in the context of industrial applications and implementations. As shown in Figure 26 and based on the number of the structure layers, CNTs are classified as single walled carbon nanotubes (SWCNTs) and multi walled carbon nanotubes (MWCNTs).

Pourfayaz et al. evaluated the adsorption capacity of two types of multi walled carbon nanotubes

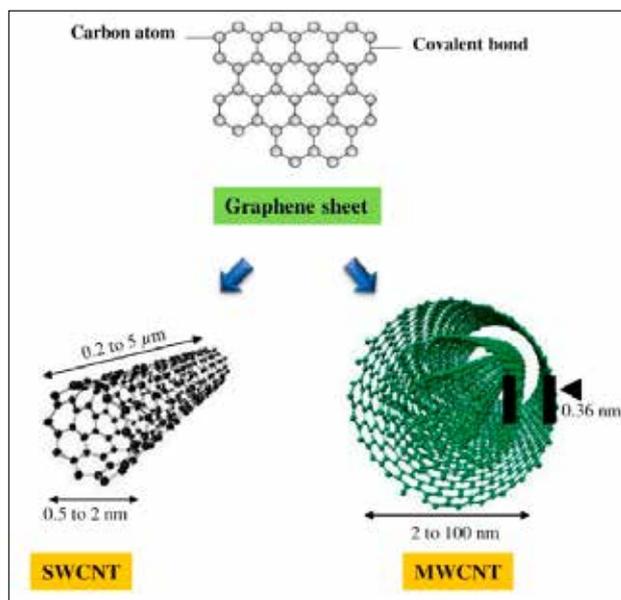


Fig. 26. The conceptual scheme representing general dimensions of the length and width of single walled carbon nanotubes (SWCNTs) and multi walled carbon nanotubes (MWCNTs) [131].

(MWCNTs) (Fig. 27) with different functional groups and analyzed them using gas chromatography (GC). Confirmation of functionalization was performed through fourier transform infrared (FTIR). The observed findings demonstrated that the MWCNTs with a larger surface area and higher crystallinity have a significant adsorption capacity for both benzene and toluene [132].

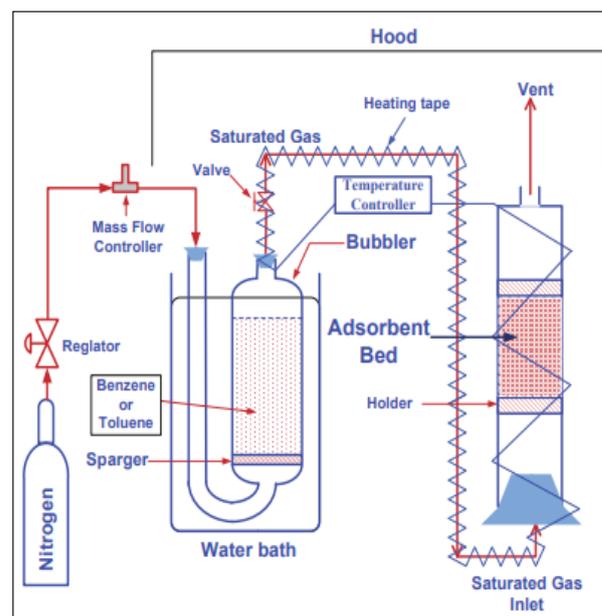


Fig. 27. Schematic diagram of experiment setup toward measurement of adsorption capacity of MWCNTs [132].

4. Comparing of diriment sorbent for removal VOCs from waters/gas

The removal procedure for VOCs from waters was compared by different analyzer such as GC-MS, GC-FID, HS-GC-MS, GC-FID/ HPLC-UV, SWV and HPLC which was shown in Table 1.

Due to Table 1, the different sorbents and techniques compared as detection limit (LOD), recovery(R), relative standard deviation(RSD%), adsorption/ desorption, temperature(T) and absorption capacity(AC) in water samples.

Table 1. Determination and separation VOCs from water/gas by different sorbents and methods

VOCs	Method	Analyzer	Analytical Features	Matrix	Ref.
BTEX	ASTM	GC-MS	LOD: 0.11–0.48 Recovery: 94–107%	Water	[133]
BTEX	DAI	GC-FID	LOD: 0.61–1.11 Recovery: 95–99%	Water	[134]
BTEX	Sorbent	HS-GC-MS	LOD: 0.001–0.05 RSD: <4.2%	Water	[135]
BTX	LV-LLE	GC-FID HPLC-UV	RSD: 2.4–11.9 R: 0.8452–0.9999 Permenkes: 0.01–0.7 Deviation: 2.13–10.96 LOD: 0.1–0.3	Water	[136]
BTEX	CBD-DE	SWV	LOD B: 3.0×10^{-7} mol L ⁻¹ LODT: 8.0×10^{-7} mol L ⁻¹ LOD X: 9.1×10^{-7} mol L ⁻¹ Recovery: 98.9–99.4	Water	[137]
ROS	SE-UOOG	GC	LOD: 0.023 mg/g RSD: <2% Recovery: 95.4–102%	Water	[138]
p-xylene	SLS-MOF/ zeolites	SOM	Recovery: more than 95% Selectivity values of 24.0, 10.4 and 6.2 vs. <i>oX</i> , <i>eB</i> and <i>mX</i>	Water / Gas	[139]
Benzene	D – μ -SPE	SHS-GC-MS	AC: CNTs@PhSA, 157.34 mg g ⁻¹ AC: CNTs, 157.34 mg g ⁻¹ Recovery: 96.8–102	Water	[140]
Toluene	G-PhAPTMS,- SGEP	GC-FID GC-MS	Removal efficiency: > 95% Adsorption: Chemical and physical Flow rate : 200 ml min ⁻¹ Temperature: 40°C	Air	[140]
benzene toluene	copper oxide nanoparticles (CuO-NPs)	HS-GC	AC for benzene: 100.24 mg g ⁻¹ AC for toluene: 111.31 mg g ⁻¹ Adsorption efficiency : 98.7% for benzene Adsorption efficiency : 92.5% for toluene	Water	[142]
VOCs	SPME-sol-gel SWCNT / silica	GC-MS	LOD: 0.09–0.2 ng mL ⁻¹ Adsorption: 15 min for 25°C Desorption: 3 min for 280 °C	air	[143]
Formaldehyde	SPME PDMS-DVB	GC-MS	LOD: 0.002–0.032 μ g m ⁻³ Adsorption: 15 min for 25°C Desorption: 4 min for 250 °C	Air water	[144]
phenol	HS-SPME CW-DVB	GC-MS	LOD: 1.13–4.60 ngmL ⁻¹	water	[145]
Alkyl PAH	HS-SPME- PDMS	GC-MS	LOD: 0.002–0.6 ng mL ⁻¹	water	[146]
Volatile sulfur	HS-SPME-PDMS-CAR	GC-FPD	LOD: 1.6–93.5 ng L ⁻¹	water lakes	[147]
PBDE	DI-SPME MWCNT	GCECD	LOD: 3.6–8.6 ng L ⁻¹ Recovery: 90–119%	river water	[148]
OCP	HS-SPME	GC μ ECD	LOD: 0.16–0.84 ng L ⁻¹ Recovery: 63–127%	Sea water	[149]
VOCs	3D-SPE-CB-PLA)	HPLC	r ² =0.96 Infill print densities: 15 - 50%. Ambient temperatures : 19.0 \pm 0.5 °C	water	[150]

AC: Absorption Capacity
 GC-MS: Gas chromatography mass spectrometry
 GC-FID: Gas chromatography equipped with flame ionization detector
 SWV: Square wave voltammetry
 SHS-GC-MS: Head space gas chromatography mass spectrometry
 GC: Gas Chromatography
 SOM: Sized Organic Molecules
 ROS: Residual organic solvent (ethanol, tetrahydrofuran, cyclohexane, n-heptane)
 D- μ -SPE: Dispersive micro solid phase extraction method
 SLS: Solid liquid separation
 SE-UOOG: Solvent extraction-Unconventional oil ore Gangues
 CBD-DE: Cathodically pretreated boron doped diamond electrode
 LV-LLE: Low volume liquid-liquid extraction
 DAI: Direct aqueous injection
 ASTM: ASTM D-5790 Purge and trap
 G-PhAPTMS- SGEP: Functionalizing graphene with N-Phenyl-3-aminopropyl trimethoxy -sorbent gas extraction
 HS-GC: Headspace gas chromatograph
 3D-SPE-CB-PLA: 3D printed solid-phase extraction carbon black modified polylactic acid (PLA)
 OCP: Organo chlorine pesticides
 PBDE: Polybrominated diphenyl ethers ACN: acetonitrile
 BTEX: benzene, toluene, ethyl benzene, ortho-xylene and meta- and para-xylene
 BTX: benzene, toluene and xylene
 DAD: diode array detector
 DI: direct immersion
 ECD: electron capture detector
 FID: flame ionization detector
 FPD: flame photometric detector
 FD: fluorescence detection
 LOD: limit of detection ($\mu\text{g L}^{-1}$)
 LOQ: limit of quantification ($\mu\text{g L}^{-1}$)
 MA: microwave assisted
 MAE: microwave assisted extraction
 MW: multiwalled; PAHs: polycyclic aromatic hydrocarbons
 PDMSAC: PDMS mixed with activated C
 TSD: thermoionic specific detection

5. Conclusions

Through entering from different sources such as water, air and foods, the VOCs cause several diseases in humans. So, the environmental samples such as water, wastewater and air must be controlled and determined by applied

methodologies. The human life depends on water future and elimination of pollutions such as VOCs in waters. By growing economy and increasing population, the main theme is water supplies without any contaminations. Quantity and quality of water must be checked daily and main parameters of waters should be controlled. So, the water, especially drinking water conservation is extremely important, and contaminations such as BTEX, VOCs and other organic pollutions in waters should be removed by new technologies. The technologies based on sorbents depend on water characteristics, affordability, acceptability and level of application. Every methodology for VOCs removal from waters have many advantages and disadvantages for water treatment. Therefore, the important parameters for any methodology such as speed, simplicity and selectivity must be studied. The vary methodology such as, solid phase extraction, liquid phase extraction, the adsorption/desorption, the sole gel technology, RCO, CO and PCO have been used for VOCs removal from waters. In this review the recent technologies based on sorbents or catalysts are introduced for VOCs removal from water samples.

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