REMOVAL OF FORMALDEHYDE FROM WASTEWATER AND AIR: REVIWE

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Abstract: Formaldehyde is one of the most widely used and widespread aldehydes, and it has been classified as a toxic chemical and may be carcinogenic. Millions of tons of it are released annually into the environment, whether the environment is internal or external, as it is included in many industries in which it and its products are in direct contact with humans. It became necessary to remove formaldehyde, whether from air or water contaminated with it. The main purpose of preparing this paper is to review previous research in removing formaldehyde from waste water and the air by various methods and techniques, including chemical, biological and physical.

Keywords: *Removing formaldehyde, formaldehyde, polluted water, polluted air, removal efficiency.*

1. Introduction

The most prevalent aldehyde in the environment is formaldehyde (HCHO). As an environmental pollutant, it is abundantly found in the aquatic and air environments [1]. Formaldehyde is a colourless gas that has a strong ability to react as well as water-soluble. The solubility of formaldehyde in water is known as formalin [2].

Its aqueous solution, which contains 37 percent - 40 percent formaldehyde by volume or by weight is named formalin, it is also hazardous and poisonous substance. Formaldehyde is naturally produced by the oxidation of hydrocarbons in the earth's atmosphere (troposphere). In its natural state, it occurs as an intermediate compound at low concentrations in the methane cycle in the

gaseous state. It is also produced in soil during the early stages of decomposition of plant residues [3]. Car emissions from engines and household combustion, such as smoke of tobacco, cooking and heating are the main man-made of formaldehyde sources [3]. Formaldehyde in water, is hydrated which is normally taken the form of methylene glycol as well as its oligosaccharides [4]. In 1859, Alexander Mikhailovich Butlerov first synthesized formaldehyde (HCHO) and again described in 1868 via A.W. Hofmann. It is an industrial chemical that is commonly employed. Formaldehyde is considered as one of the most important industrial chemicals for manufacturing synthetic resins, explosives, fertilizers, and more. It is used in many industrial processes [5-7]. Formaldehyde was considered one of the most important air pollutants due to its toxic effect [8,9]. There are 45 toxic organic compounds that have effects on the environment, and formaldehyde is one of them [10]. Recently, the International Agency for Research on Cancer and the National Toxicology Program have designated formaldehyde as a human carcinogen, which causes both pharyngeal cancer and myeloid leukemia [11]. If the air contains 10 parts per million of this highly toxic substance, it

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causes lung and respiratory cancer, skin damage, as well as serious damage to the eyes [12]. In Germany and specifically in Berlin during the Second World War, the first commercial wooden board was produced. Since 1950, particleboard has replaced solid wood in the manufacture of furniture, as well as wood panels have been used in the construction of houses. In the mid-1960 it was observed that the effect of formaldehyde in prefabricated dwellings was evident, causing irritation to the eyes and upper airways. Formaldehyde emissions from particulate matter adherent to urea-formaldehyde resin were identified as a cause of complaints. That is why in 1977 the German Federal Health Agency proposed a twig value of 0.1 ppm in order to reduce human exposure in homes, and in Germany and Denmark in 1981, standards were developed to determine and regulate formaldehyde emissions. Formaldehyde was discussed as a cancer-causing substance in the 1980, when carcinogenic cases were reported to mice and rats after they inhaled it for long periods [13,14]. Given the lack of confidence in the presence of formaldehyde in the environment, the Scientific Committee on Health and Environmental Risks decided to consider its effect similar to the effects (carbon monoxide, nitrogen dioxide, benzene, naphthalene, environmental tobacco smoke (ETS), radon, lead, and organophosphate pesticides) which are considered to be exciting substances. For fears due to its negative impacts on the environment [15].

2. Remove Formaldehyde From Water

Many researchers and scientists have done research on the removal of formaldehyde from water and in different removal methods. Below, the removal methods will be presented:

Zvulunov et al. [16] Found it possible in a one-step process, a multi-functional, self-

regenerating material was designed to adsorb and made biodegradation to formaldehyde from water. The material is made up of montmorillonite clay, polyethyleneimine, and *Pseudomonas putida*, which degrades formaldehyde. The positive surface charges and high concentration of functional amine groups in a polyethyleneimine-clay composite facilitated formaldehyde bacterial adhesion and adsorption. The formaldehyde binding was pH dependent and the results are consistent with the model of Langmuir and had a composite q_{max} of 62 mg. g⁻¹.

Memon et al. [1] Were investigated on adsorption effectiveness Amberlite IRA-910. pH, adsorbent dosage, adsorbate concentration, shaking speed, volume, and shaking time have all been extensively investigated. The formaldehyde content in the water was determined using ultraviolet-visible spectroscopy. The Langmuir, Freundlich, and D-R isotherms were used in the isothermal study. Amberlite IRA-910 has a greater adsorption efficiency for the removal of formaldehyde at high and low concentrations, according to the results.

Wang et al. [17] Studied monitoring pollution of water and improving human safety, ultrasensitive and ultraprecise detection of toxic target molecules is highly desirable. Using an ethylenediamine (EDA)-functionalized poly (ionic liquid)/polyacrylonitrile nanofibrous membrane (PIL/PAN NFM), a novel ionic microchannel responsive for formaldehyde (CH₂O) was fabricated successfully. When reacting with more HCHO the electron affinity increase and zeta potential decrease of the microchannels, so the resulting ionic current output will switch from low to high when using CH₂O with EDA immobilized on the prepared ionic nanofibrous membrane. Through an established linear correlation between responsive ionic current and HCHO concentrations, the

ionic microchannels showed high sensitivity for the determination of HCHO ranging from 360 ppm (3.6 102 mg/L) to 0.036 ppt (3.6 108 mg/L) ($R^2 = 0.93$). Moreover, because of the abundant amino grafted on the membrane, the ionic microchannels can remove a huge amount of HCHO molecules from an aqueous solution.

Gong et al. [18] Checked The feasibility of using water hyacinth to treat formaldehyde that found in wastewater .In a experiment of laboratory, water hyacinth had a great efficiency to removal for formaldehyde inputs of 100-300 parts per million. The formaldehyde eaten by the root of the water hyacinth was transferred from the root to the stem zone, then to the leaf, and eventually to an acidoid. In water hyacinth, the removal activity was boosted by increased activity of root and chlorophyll by adding of 0.5 mg/L extract of *Eupatorium odoratum* L. Meanwhile, growth of water hyacinth was effective regulated, with sprout emergence suppressed and a low rate growth (≤ 0.032 g.day⁻¹). In a pilot testing during the four recycle experiments, water hyacinth also demonstrated outstanding ability for formaldehyde removal, and the rate of growth was kept to a little value of about 0.023 g.day⁻¹.

Gutiérrez-Arzaluz et al. [19] Were presented the synthesis of cerium oxide, cobalt oxide, mixed cerium, and cobalt oxides, as well as a Ce–Co/Al₂O₃ membrane, as catalysts for the catalytic wet oxidation (CWO) reaction and the removing formaldehyde from wastewater of industries. The CWO reaction was happened in a slurry batch reactor and a membrane reactor using Ce- and Co-containing catalysts.

(Kulikauskaitė and Paliulis) [20] By using natural zeolite, studied its potential for use as a formaldehyde remover. Natural zeolite was used in the experiment, which had different contact times with different concentrations of formaldehyde solutions. The Photocolorimetric

Method was used to calculate the formaldehyde concentration. After 8 hours of contact time with the adsorbent, the average sorption efficiency was determined and it was greatest when concentration of formaldehyde was lowest, e.g. 2 mg/l (45.94 percent). When the time of contact was increased, sorption efficiency improved, but when the time of contact was extended to twelve hr, efficiency of sorption remained the same due to zeolite saturation.

Poyan Ghanbarnejad et al. [21] Investigated the possibility of using as an adsorbent, human hair and sheep wool were used to remove formaldehyde . In wastewater the concentration of formaldehyde was measured by using the method of chemical oxygen demand during the adsorption process in current study (COD). The effect of various parameters on human hair and sheep wool, like temperature, pH, hydraulic retention time, weight, formaldehyde concentration and were then assessed. Formaldehyde can be removed from wastewater in five minutes, and increasing the retention time from five to 25 minutes has no effect on formaldehyde adsorption performance. The findings revealed that the adsorption of formaldehyde effectiveness of colored hair with chemical or natural colors (e.g. Henna), white hair, and sheep wool is comparable to that of common human hair. It is possible to remove formaldehyde from industrial wastewater using human hair or sheep wool.

Taghizadeh [22] In this study the photo catalytic oxidation of formaldehyde with TiO₂ Nano powder, ultra violet radiation, and H₂O₂ was studied. Solution of formaldehyde [35 mg/l] was maked in an experimental laboratory scale unit and exposed to UV, n-TiO₂, hydrogen peroxide, and a mixture of them in different PH levels in separate stages. UV, TiO₂ Nanopowder, and H₂O₂ together have the highest effectiveness of formaldehyde removal, according to the

experiments. In the fundamental condition [pH=12], UV radiation degraded 92 percent of formaldehyde, according to this study. The results as well show that the number of UV lamps used, the concentration of n-TiO₂, and the amount of hydrogen peroxide used all have a significant impact on formaldehyde removal effectiveness.

Goli et al. [23] They look into the process of making from cypress tree leaves activated carbon, as well as its effectiveness in treating wastewater contaminated with formaldehyde. Various parameters effects such as pH, formaldehyde concentration, retention time and temperature were studied in addition to the adsorption ability of the adsorbent. The most effective formaldehyde removal condition by the adsorbent is a pH of 7, a retention time of 5 minutes, temperature of 10C, and which resulted in a 98 percent removal. The obtained activated carbon has proven effective in removing formaldehyde.

Agarwal et al. [24] Were studied the adsorption mechanism and the removal efficiency from formaldehyde on treated active carbon and active alumina for analysis effect of surface modified chemistry of absorbers. At STP, the capacities of adsorption for all adsorbents studied in a batch study with different retention times and initial formaldehyde concentrations. So if the initial concentration of adsorbate is reduced, commercially adopted treated activated alumina with an effective nucleophile showed substantial adsorption for formaldehyde with little transformation in capability of adsorption.

Afkhami et al. [25] Observed by grafting the external surface of -alumina (-Al₂O₃) nanoparticles with functional groups of 2,4-Dinitrophenylhydrazine (DNPH) which had a robust affinity to the formaldehyde, a firsthand and effective sorbent for remove of

formaldehyde in the samples of water was suggested. This adsorbent was made by a simple process of amination of the γ -Al₂O₃ surface. By reacting sodium dodecyl sulfate coated nano-alumina with DNPH solution, nitrogen-containing groups were grafted on the γ -Al₂O₃ nanoparticles. In real-world samples, the generated sorbent performed admirably in removing formaldehyde. pH, adsorbent dosage and contact time have all been analyzed and determined to be optimal experimental conditions. The experimental data was analyzed using Langmuir and Freundlich isotherm models.

Salman et al. [26] Investigated in batch mode, the formaldehyde adsorption on bentonite and kaolin. The effects of variables such as adsorbent dose, pH, contact time, and agitation speed were studied. The isotherms of Langmuir, Freundlich, and Temkin were used to describe adsorption equilibrium. According to the Langmuir isotherm, the maximum concentrations of formaldehyde adsorbed (q_{max}) were 3.41 and 5.03 milligrams per gram of kaolin and bentonite, respectively. The findings led to deduction that inorganic clays like kaolin and bentonite, which are widely obtainable, have respectable potential as adsorbents for removing toxic organic pollutants like formaldehyde from water.

Moussavi et al. [7] To fully treat concentrated formaldehyde wastewater, they combined the catalytic oxidation process (CAOP) of O₃/MgO/H₂O₂ with a sequencing batch reactor (SBR) system. For the O₃/MgO/H₂O₂ degradation of a 7000 ppm formaldehyde in wastewater, the impact of some variables operational was studied, including MgO powder dosage, pH, and H₂O₂ and O₃ concentrations. 5 g/L dose of powder of MgO, pH 8, 0.153 g/L min dose of O₃ and concentration 0.09 mole/L of H₂O₂ were found to be the best conditions. Under the optimum conditions, the concentrations of formaldehyde and COD in the

CAOP were reduced by 79 percent and 65.6 percent, respectively, after 120 minutes of reaction time. The remaining of COD and formaldehyde concentrations in the effluent were 3200 ppm and 1500 ppm, correspondingly. Formaldehyde degradation in CAOP was found to be a first-order reaction with a constant of 0.015/min, with radical oxidation as the most common mechanism of degradation. This effluent was handled for a total of 24 hours in an SBR system. The formaldehyde was entirely removed, and 98 percent of the COD was removed, lowering the concentration of COD to less than 60 mg/L.

Lotfy and Rashed [27] They investigated experiments of biodegradation to sample of wastewater having formaldehyde concentrations range from 31.5 to 125 ppm, as well as remaining formalin (a solution of formaldehyde gas in water) concentrations range from 40 percent to 85 percent, sequentially, when the run was end (16 days), demonstrating the inhibition effect of formalin, which increased as formalin concentration increased. They describes a method for converting formaldehyde to a biodegradable substance. Formalin biodegradation was markedly reduced at concentrations greater than 300 ppm. After treating the wastewater with calculated quantities of sulphite of sodium, biotransformation of phenol in a wastewater contain great levels of formaldehyde began. The reaction of sodium sulphite with formaldehyde is thought to produce sodium formaldehyde bisulphite, which is biodegradable and non-toxic to microorganisms. The researchers noted that the amount of dissolved oxygen in a wastewater holding formaldehyde is unaffected by addition of the calculated amount of sulphite of sodium, which is just enough to eat the amount of formaldehyde that measured in wastewater, based on DO

measurements taken before and after the addition of sulphite of sodium.

3. Remove Formaldehyde From Air

Removing formaldehyde from air has not received as much attention as it does in removing it from water, but there are many studies and research on removing formaldehyde from air. Below is a review of most of these research:

Pásztori et al. [28] They looked at how the *Populus × euramericana* cv. Pannónia bark reacts in a rich environment with formaldehyde. The bark samples were made like Prism-shaped and preserved for 1, 2, 5, 10, 18 and 36 days in a formaldehyde-contaminated environment. The adsorbed formaldehyde which is then desorbed was measured after the contamination. The uncontaminated poplar bark had a formaldehyde content of 0.0036 mg/g. As a function of time, the quantity of bound formaldehyde exhibited a saturation curve. The equilibrium value for formaldehyde adsorption was 0.9 mg HCHO/g bark. The formaldehyde emission from bark samples contaminated with it followed curve which is basic as a time function, and after the formaldehyde was released, some residual formaldehyde content was detected.

Wang. et al. [29] The biological component in this study was the root zone of *Canna fasciata* Salisb. They discovered that *C. fasciata*'s formaldehyde adsorption rate was initially low before rapidly rising. *C. fasciata* can metabolize HCHO, according to CNMR analysis, also the main metabolic product is organic acids and amino acids. To remove formaldehyde, they created a suspended growth bioreactor (SGB). An SGB put to the test under a variety of conditions, and the results revealed that zone of root weight, water volume, and rate of flow of gas all had a significant impact on formaldehyde removal efficiency. In one hour, more than 52% of the formaldehyde was removed, with a max

capacity elimination of 35.112 mg m⁻³ h⁻¹. There was not at all formaldehyde left in the SGB after 24 hours. The formaldehyde concentration in the lab reduced from 1.12 to 0.82 mg m⁻³, and the formaldehyde concentration in the SGB touched a small level after fifteen days of operation.

Talaiekhosani et al. [30] Were removed the formaldehyde from the air stream absorbed in a packed layer in an aqueous stream continuously. They are then removed via using a mixture of ultraviolet rays (UV) and ferrate (VI) as the high-strength oxidant in endless stirring tank. In fact, both batch and continuous methods of removing formaldehyde from water were studied. The main goal of their research was to comparison the effectiveness of utilizing UV and ferrate (VI) separately versus UV/ ferrate (VI) together to get rid of formaldehyde that found in both water and air. In addition, the effects of temperature on removal process of formaldehyde from water using the ferrate (VI) way, pH and ferrate (VI) concentration were investigated. The best initial ferrate (VI) concentration and pH for obtaining the highest formaldehyde removal were 1 and 2 mg/l, respectively, according to the results of the in batch condition. Also, increasing the temperature from 25 to 50 degrees' Celsius increases removal of formaldehyde from 69 to 97 percent; however, increasing the temperature more reduces removal efficiency. Within 35 minutes, a mixture of ferrate(VI) and UV boosts removal efficiency of formaldehyde to nearly 100%. Using a packed bed scrubber with a gas over liquid flow rate ratio of 1.28 m³ /m³ treatment of the continuous air stream, full removal formaldehyde of 94 percent was achieved.

Talaiekhosani et al. [31] They were investigated the possibility of using human hair to take away formaldehyde from the air by use a packed reactor with hair of human. At a primary

concentration of 8500 mg/L, formaldehyde-contaminated air was presented into a reactor have cylindrical shape packed with hair of human. Parameters that are effective at removing formaldehyde were measured, such as the amount of human hair and the temperature of the environment. According to the findings, each human hair gram will remove 0.13 to 0.49 g formaldehyde from the air. Under 20 degrees Celsius, 98 percent of formaldehyde adsorbed by human hair from synthetically polluted air. Increase in temperature had a reverse effect on removal formaldehyde, according to these findings.

Xu et al. [32] Mesoporous aluminum oxyhydroxides they made up of nanoflakes were made using salts of aluminum as precursor and hydroxide of ammonium as means of precipitating in a water-in-oil microemulsion-assisted hydrothermal process at 50 °C. Due to the abundant presence of hydroxyl aggregates on the surface, the specific surface area is large, and the pores are of an adequate size so that the nanostructures of oxyhydroxide aluminum examined are active absorbents for removing formaldehyde at room temperature. The materials' microstructure formation and adsorption efficiency of the products that resulted were also dependent on the type of aluminum precursor used. In the first run, the sample made of aluminum sulfate (Al-s) had a relatively great capability adsorption HCHO, while the samples made of chloride (Al-c) and aluminum nitrate (Al-n) had a great adsorption capability and a relatively steady recyclability.

Rezaee et al. [33] They test the adsorption capacity of modified bone char (BC) with acetic acid for the removal of formaldehyde from air. The modified BC was discovered to have a larger area on the surface than original BC. The amended BC had a maximum surface area of 118.58 m²/g. The modified BC had a higher

breakthrough, equilibrium time, and adsorption ability than the original BC. Furthermore, the times of equilibrium for BC were 85, 75, 65, and 45 minutes and modified BC 95, 85, 70, and 50 minutes, in turn, at primary concentrations of 20, 50, 100, and 200 mg/L. Modified BC's formaldehyde adsorption capacity appears to be influenced by both physical and chemical characteristics.

Xu et al. [34] They were created Ni(OH)₂/SiO₂ composites with hierarchical flake-like nanostructures, which they characterized using X-ray diffraction, thermogravimetry, transmission electron microscopy, Fourier transform infrared spectroscopy, scanning electron microscopy, X-ray photoelectron spectroscopy and nitrogen adsorption. The formaldehyde (HCHO) removal performance of the as-prepared hierarchical porous Ni(OH)₂/SiO₂ composites in air at ambient temperature is excellent. The surface area, pore structure, and HCHO adsorption were found to be significantly affected by the aging time. When compared to Ni(OH)₂, the Ni(OH)₂/SiO₂ composite aged for 4 hours in the presence of tetraethyl orthosilicate (TEOS) showed a relatively high capacity of adsorption for HCHO as well excellent recyclability, owing to a large surface area relatively, a suitable proportion of Ni(OH)₂ and SiO₂, an optimum pore size, and a synergistic effect between Ni(OH)₂ and SiO₂, and a synergistic effect between Ni(OH)₂ and SiO₂. The findings show that hierarchical porous Ni(OH)₂/SiO₂ composites can act as an effective adsorbent for HCHO in the air.

Xu et al. [35] They used X-ray diffraction, thermogravimetry, Fourier transform infrared spectroscopy, scanning electron microscopy, transmission electron microscopy, X-ray photoelectron spectroscopy and nitrogen-adsorption/desorption to characterize mesoporous ferrihydrite/SiO₂ composites

synthesized using a water-in-oil microemulsion technique. At room temperature, the porous ferrihydrite/SiO₂ composites demonstrated outstanding performance for adsorption formaldehyde (CH₂O) removal from indoor air. The pore structure, surface area, and HCHO adsorption of these materials were found to be significantly affected by the aging time during the synthesis. The ferrihydrite/SiO₂ composite aged for three hours in the tetraethyl orthosilicate (TEOS) presence had a relatively high capacity for adsorption HCHO and worthy recyclability, which was attributed to a relatively large BET surface area, optimum size for pore, a ferrihydrite/SiO₂ synergistic impact and a suitable Si/Fe atomic ratio.

Wen et al. [36] By using a self-adsorption designing and distillation system, they compare performances of the activated carbon adsorption obtained from sludge of sewage (ACSS) to three commercial activated carbons (CACs) for gaseous formaldehyde removal. Thermogravimetric (TG) analysis was used to investigate the desorption of formaldehyde by activated carbons for regeneration. The results indicate that ACSS have outstanding adsorption effectiveness, which is superior to CACs in general. According to adsorption theory, the ACSS outperforms the CACs according to structure of its porous and surface chemistry features that are conducive to formaldehyde adsorption. The best temperature for regenerating ACSS is 75 degrees Celsius.

Xu and Hou [37] They treated formaldehyde polluted air with a biodegradation system during the four-month experiment, air pressure dropped 12 mm water in the trickling biofilter. This biodegradation system had a degradation capacity of 4.0–40.0 mg h⁻¹ in the range of 20–300 mg m⁻³ formaldehyde influent, with a degradation efficiency of 100%–66.7 percent. The trickling biofilter degraded more

formaldehyde than the bioreactor of activated sludge under 200 mg m⁻³ influent formaldehyde gaseous, but less than the activated sludge bioreactor over 200 mg m⁻³ influent gaseous formaldehyde.

Johnson et al. [38] They looked at parallel flow membrane contactors counter-current to figure out how much formaldehyde is removed from the air during air humidification membrane and cooling evaporative. Shell side mass transfer coefficients dominated in determining removal efficiencies. When velocity of water inside the lumens fiber is little and water supplied to the modules approaches module evaporative damages, however, removals decrease. While the rates of removal were found to be sufficient for controlling emissions of formaldehyde in most space air-conditioning applications, big amounts of water were consumed.

Matsuo et al. [39] They were using graphite oxide silylated by 3-aminopropylmethyldiethoxysilane as a low-concentration adsorbent for formaldehyde from the vapor phase. Even when water molecules were preadsorbed, the amount of adsorbed formaldehyde on this adsorbent was considerably greater than on activated carbon. The amino groups appeared to be more useful in samples with lower organic component content between the graphite oxide layers and greater interlayer spacing's.

4. Conclusions

It is necessary to remove formaldehyde from the environment due to its toxic and dangerous effect on all living organisms and the environment, as well as to reduce its use in industries that come in direct contact with humans because it is one of the most used aldehydes, given the above-mentioned methods of treating water and air from formaldehyde, including chemical and physical treatment methods. Including the biology of

these methods, some of them are expensive, others are economical, others with simple techniques and some with high techniques, but there is no method that achieves a 100% formaldehyde removal efficiency.

Conflict of Interest

The authors confirm that the publication of this article cause no conflict of interest.

5. References

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