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REMOVAL OF AMMONIA FROM WASTEWATER WITH SPECIAL EMPHASIS ON COKE-OVEN EFFLUENTS

Removal of ammonia from the wastewater discharged from various types of plants, including those producing coke-oven by-products, is one of the major problem in environment protection.

This paper discusses the adverse affects of ammonia on environment and also reviews various available techniques for ammonia removal by recovery and destructive processes. The techniques opted for coke-oven effluents have been critically analysed.

1. INTRODUCTION

Ammonia, which is discharged by many industrial plants in high concentrations, can have an adverse impact on receiving fresh water streams. It is highly soluble in water and if it is not removed, it will exert deleterious effect on the receiving stream. Further, it increases nutrient concentration in surface water, which results in eutrophication due to the growth of algae and other aquatic plants. Ammonia is converted into nitrite and nitrate which requires large amount oxygen dissolved in receiving water, thus the process is responsible for depletion of dissolved oxygen. Ammonia is toxic to fish even if its concentration is very low (0.2–2.0 mg/dm³). It is also toxic to other aquatic animals. Ammonia in its nitrate form, if present in drinking water, causes a blood disease called methemoglobinemia or "blue baby" in infants. It reacts with chlorine to form chloramines (a less powerful disinfectant than chlorine), thus, increasing chlorine demand at both water and wastewater treatment plants. For this reasons ammonia in wastewater is reduced to very low level by different techniques.

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The available techniques for ammonia removal are based on the following processes:

(i) recovering ammonia by concentrating it (ion exchange, reverse osmosis, chemical precipitation),

(ii) stripping it as ammonia gas (air or steam stripping),

(iii) destroying it by converting into nitrogen (breakpoint chlorination or biological nitrification-denitrification).

These techniques and also existing techniques of ammonia removal from coke-oven plants are described below vis-á-vis their advantages and disadvantages.

2. ION EXCHANGE

Ion exchange is a well-known technique for purification of wastewater. Ion exchange resins may be synthetic polymeric materials or naturally occurring zeolites. The ion exchange capacity for ammonium removal has been studied using natural and synthetic zeolites [3], [4], [49], Italian phillipsite tuff [6], [7], Hungarian natural zeolite clinoptilolite [9], [21], Nenjian China's clinoptilolite [8] and clinoptilolite [4], [7], [34]. The clinoptilolite has been regenerated effectively using 3% NaCl solution [34] and solutions of NaOH or NaCl-Ca(OH)₂ mixture [61]. The regenerated solution [61] containing 1000 ppm of ammonia nitrogen was aerated at 50°C to remove 90% of the ammonia. LIBERTI et al. [33] have treated secondary domestic effluents using commercial ion exchangers, i.e., natural zeolite clinoptilolite and porous strongly basic anion resin. Ammonia and phosphate have been removed up to 90% and the regeneration of resin using 0.6 M sodium chloride allowed us to produce pure, sterile magnesium ammonium phosphate. KOROBCHAN-SKII et al. [31] have removed ammonium from coking wastewater treated by settling, sand filtration or coagulation with FeCl3 and by cation exchange on clinoptilolite or Kv-2 exchanger regenerated by H₂SO₄. This technique has also been used for ammonia removal from effluents produced in fertilizer plants [23]. POLTA et al. [42] have shown that application of clinoptilolite allowed 41-89% removal of ammonium from sewage. The removal efficiency and the exchange capacity of the zeolite were significantly affected by both the regenerant volume and pH. HASHIMOTO et al. [19] have removed ammonium from wastewater using such minerals as synthetic zeolite, clinoptilolite, montmorillonite, porous limestone, brown coal, activated carbon and activated alumina and found that zeolite and clinoptilolite are characterized by high ammonium adsorption capacity. WANG et al. [64] have treated wastewater containing 100-1000 mg of ammonium per dm³ using inorganic exchanger ZC-1 and obtained an effluent containing less than 15 mg of ammonium per dm³. VOKACOVA et al. [63] have shown that thermal treatment of clinoptilolite increases the attrition resistance and selective adsorption of NH_4^+ from wastewater. GRUENWALD et al. [15] have found that the capacity of clinoptilolite for sorption of NH_4^+ ions is equal or lower than 3.02 mg/g, thus it may be used to remove trace of NH_4^+ ions from potable water. Further it has been observed that the intraparticle diffusion [36], [57] is rate-limiting factor in removal of ammonium from wastewater by natural clinoptilolite zeolite. HORVARTHOVA et al. [22] have passed wastewater containing 860 mg of ammonia nitrogen per dm³ through a bed of ground clinoptilolite in Ca²⁺ form and the effluent obtained contained 44 mg of ammonia per dm³. Using 1 or 2 columns packed with weakly acidic cationic resins and 1 column with strongly acidic resin, HUBNER et al. [23] have removed free NH_3 and NH_4^+ from wastewater. The regeneration of resin is either direct (by blowing steam) or two-stage (by $NaOH-H_2SO_4$ process). A model assisting the design of NH_4^+ removal in wastewater treatment using natural zeolite clinoptilolite has been applied by SEMMENS et al. [48]. KLIEVE et al. [30] have pretreated samples of clinoptilolite, erionite, mordenite and phillipsite in various ways to improve the efficiency of ammonium removal from wastewater.

Ion exchange, as discussed above, is an efficient process for removal of ammonia from many types of wastewater, but the process is expensive mainly due to regeneration cost of ion exchangers.

3. REVERSE OSMOSIS

In reverse osmosis technique, a semipermeable membranes, whose pore sizes are quite small, are used. These membranes are permeable to water molecules, but impermeable to ammonia and other molecules. Under conditions of osmosis, a solvent (water) tends to flow spontaneously through the membrane into solution. By applying an external pressure greater than the osmotic pressure on the concentrated solution, the direction of water flow will be reversed, i.e., water will pass through the membrane from the concentrated solution to the diluted solution. Although the removal of ammonium by reverse osmosis can be quite efficient, this process is applied rather to concentrate ammonia than to remove it. Reverse osmosis is more suitable for pretreatment prior to other methods of ammonia removal.

4. CHEMICAL PRECIPITATION

Change in pH and the use of chemicals to form insoluble compounds of ammonia sometimes are used for treating wastewater containing ammonia in soluble form. Precipitation is accompanied by settling to remove the precipitate and use it as by-product. In many industries, wastewater is treated by chemicals in order to remove ammonia. GRUENWALD [16] has achieved over 80% degree of ammonia removal. Ammonia precipitated in the form of ammonium phosphate as a result of its reaction with either $MgSO_4-KH_2PO_4$ or $MgCl_2-H_3PO_4$ precipitants. DAMIECKI et al. [10] have removed ammonium from wastewater in two stages. The first one comprised air stripping, and second – conversion of ammonium into stable ammonium sulphate by its reaction with H_2SO_4 which did not affect negatively further activated sludge process.

The ammonia from fertilizer wastewater has been removed using lime and potassiummagnesium sulfate [2] and phosphate and magnesium [56]. Precipitate of struvite $(Mg(NH_4)-(PO_4)\cdot 6H_2O)$ formed have been used as fertilizer. The recovery product as fertilizer could significantly affect the ammonia treatment cost. Ammonia recovery by precipitation offers promising potential for application in fertilizer plant and other industrial wastewaters.

5. AIR STRIPPING

pH of wastewater containing ammonia was adjusted to the value equal to 11.0 or greater. Then the wastewater was passed through a stripping tower characterized by a high degree of agitation and mixing. Large amounts of air are circulated in counter-current fashion through the tower, promoting the transfer of ammonia from the water to the air phase. POWERS et al. [43] have investigated air stripping of ammonia from semiconductor manufacturing wastewater. KAWANISHI et al. [28] performed air stripping by taking air from the tower bottom and discharging from the tower top. They found that the process efficiency depended on pH. A wastewater of ammonia initial concentration amounting 15 000 mg/dm³ showed the removal efficiency greater than 100% at pH 11 and equal to 0% at pH 7.0.It has been proved that pH>10.5 is required to prevent NH₃ conversion into ammonium. During aeration pH declines and therefore it must be maintained artificially. The optimum ammonia removal (~99%) is achieved at 40°C, 1500:1 air-wastewater ratio at 10 hours. The removal efficiency improved as the wastewater temperature and air blowing time increased.

Air stripping is highly efficient in ammonia removal, but some precautions should be taken against scale formation and reduced efficiency at lower temperatures. Air stripping is significantly less efficient than steam stripping for ammonia removal. Both air and steam stripping of highly concentrated ammonia waste have been applied by TSAI et al. [59].

If high amounts of ammonia are stripped, the exhaust air may easily exceed allowable air standards and odour. Therefore ammonia removed by air stripping can be reabsorbed into acid absorption solution. If sulfuric acid is used as absorber, ammonium sulfate slurry formed can be potentially used in fertilizer production. When air temperatures approach 0° C, the air stripping process comes to an end. In the winter time, a plant processing nuclear fuels (USEPA, NEIC, COLORADO) in Tennessee changes over from air stripping to breakpoint chlorination to properly treat wastewater loaded with ammonia. SURI et al. [52], [53] have discussed disadvantages of the air stripping applied to ammonia removal from fertilizer containing wastewater.

6. STEAM STRIPPING

Steam stripping [29] of ammonia from wastewater is a reliable treatment process used in many industries. High capital costs of steam stripping are offset by the recovery and reuse of ammonia in the process. Ammonia in the overheads of the steam distillation column is collected as ammonium hydroxide of the concentration of 30% or higher. Caustic soda or lime is used for adjusting pH of the steam stripped. Scaling is minimized by means of caustic soda. GRUENWALD et al. [17] have adjusted pH of wastewater to 9.25 by caustic soda and recovered 99.8% ammonia in 3 hours by absorbing it in boric acid. TSAI et al. [60] have treated sour water from coal liquification plant by steam stripping and removed 99.8% ammonia at 9.25 pH. WICKRAMANAYAKE et al. [65] have investigated the effect of temperature, presence of SO_4^{2-} anions and the type of chemicals used for pH adjustment on pH of wastewater during steam stripping. TSAI et al. [59] have investigated both air and steam stripping for the treatment of highly concentrated chemical wastewater and found that steam stripping required less caustic soda for pH adjustment and permitted average ammonia recovery of 50%.

The major operating cost of steam stripping is steam production and consumption of lime and caustic soda for pH adjustment. Factors facilitating efficient ammonia removal comprise proper design of the steam stripper, control of hydraulic rates of flow, adequate steam properties, pH values of 11.0 and greater, temperature of 93°C or higher in the stripping column, sufficient deentrainment space and a compatible ammonia condensing system

7. BREAKPOINT CHLORINATION

Ammonia present in many industrial wastewaters can be removed due to its reaction with chlorine. Free chlorine reacts with ammonia to form mono-, di- and trichloramines which are further oxidized to give nitrous oxides, nitrates and finally gaseous nitrogen. ATKINS et al. [41] have removed ammonia from fertilizer wastewater by chlorination followed by dechlorination with granular activated carbon. OSANTOWSKI et al. [44] have removed ammonia from coke plant effluents by alkaline chlorination followed by dechlorination using either activated carbon or sodium metabisufate. HUANG et al. [24] have removed ammonia from wastewater by its partial chlorination followed by activated carbon adsorption. The process led to formation of some mono- and dichloramines soluble in aqueous solutions of different pH and different Cl_2/NH_3 ratio. The quantitative interactions between activated carbon and each of chloramine species were evaluated. WATANABE et al. [66] have applied this method, with 86.9–100% efficiency, to remove ammonia from wastewater using chlorine water and NaOCl. Reaction was completed in 5 minutes, independently of temperature, and optimum removal efficiency was achieved at pH ranging from 7 to 8.

Both breakpoint chlorination and partial chlorination associated with dechlorination by activated carbon are efficient processes for removing ammonia from wastewater. One gram of nitrogen for its complete oxidation needs 7–15 grams of chlorine. Breakpoint chlorination is the best method in removal of relatively low ammonia concentration. In this process, 90–95% of ammonia can be removed and there is no recovery of ammonia.

8. BIOLOGICAL NITRIFICATION-DENITRIFICATION

Ammonia, if present in high concentration in wastewaters, can be removed by biological nitrification-denitrification. In nitrification, ammonium nitrogen is first oxidized by *Nitrosomonas* type of bacteria to nitrites which are further oxidized to nitrates by *Nitrobacter* type of bacteria. In denitrification, nitrates are converted to gaseous nitrogen by the ni-

trate reducing bacteria which are facultative anaerobic heterotrophs. Several workers [5],[11]–[13], [35], [37], [38], [45], [55] have investigated usability of nitrification-denitrification processes for removal of ammonia from coke-oven wastewaters. In many industrial wastewaters, neutralization of toxic ammonia by nitrifying bacteria has been applied [1], [25], [32], [39], [46], [67]. Processes of biological nitrification-denitrification using trickling filters and biological disc contactors have been applied by ZANDER [68] and SFORZA et al. [50].

In many industrial wastewaters, particularly in coke plant wastewater, nitrificationdenitrification, nitrification and denitrification technologies are normally used for treating ammonia wastewater. These processes are very efficient in removing ammonia from wastewater.

9. DISCUSSION OF TECHNOLOGIES OPTED IN COKE PLANTS

Ammonia present in coke plant effluents is removed mainly by biological nitrification-denitrification or steam stripping processes. Biological nitrification-denitrification process for ammonia removal is a cost-effective approach for treating wastewater. But in this process the recovery of ammonia is not possible. Steam stripping process allows recovery of more than 90% of ammonia which can be used for many purposes. TAKADA et al. [58] have used steam stripping for removal of ammonia from coke-oven wastewater. The spent cooling water has been flash-evaporated at a reduced pressure and the resulting vapour has been pressurized to give steam for ammonia stripping. GANCHARCHYK et al. [14] have alkalized coke-oven wastewater in a contact chamber. Thereupon the wastewaters have been directed to a settler producing NH₃ liquor containing free ammonia which have been removed by steam stripping. GU YUGANG et al. [18] have proved that efficiency of ammonia recovery from coke-oven effluents by steam stripping at pH > 10amounted to 89-95%. KAMEDA et al. [27] have removed ammonia from coking wastewater by steam generated by such a technique which reduces by 75% the cost of steam production, compared to conventional method. At Heisaka Works Ltd [20] (Japan), ammonia and naphthalene are steam-stripped in the presence of hydrophobic organic solvent to prevent fouling problems. In Kako Co Ltd of Japan (1980), sulfur has been used to precipitate H₂S and HCN as H₂S₂O₃ and HCNS from coking wastewater, then steam stripping of ammonia has been done. This gives better results than the earlier one when the wastewater is not treated with sulfur. From wastewater produced by coking plant, JOSIS et al. [26] have removed phenol, ammonium compounds and volatile ammonium compounds in a fractionating column. They have passed steam from the bottom of the column using $Ca(OH)_2$ for decomposition of solid ammonia compounds.

Steam stripping process is an efficient technique for removal and recovery of ammonia from coke-oven effluents. The major cost of this technique is production of steam which requires high energy input.

Due to their selectivity and high ion-exchange capacity, zeolites can be applied to both removal and recovery of ammonia from industrial effluents. Work regarding the removal of ammonia from the coke-oven effluents by various techniques is in progress and a cost effective technique would be developed and demonstrated in *in situ* condition.

10. CONCLUSIONS

Many plants discharge effluents containing high concentrations of ammonia which gives rise to adverse effect on aquatic life of the fresh water stream. Many treatment techniques such as recovery and destructive processes may be adapted for removal of ammonia.

Biological nitrification-denitrification is a very cost-effective treatment process for removal of ammonia from coke-oven effluents, but it does not guarantee recovery of ammonia. Steam stripping is a very efficient method for recovery of ammonia from coke-oven effluents, but it requires high input of energy for steam production. Zeolites are characterized by very efficient ion-exchange capacity and selectivity for removal of ammonia from many industrial wastewaters. These zeolites can be regenerated many times using inexpensive brine solution. Moreover, zeolites are very cheap and easily available. Zeolites can also be used for selective removal of ammonia from coke-oven effluents.

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USUWANIE AMONIAKU ZE ŚCIEKÓW ZE SZCZEGÓLNYM UWZGLĘDNIENIEM ŚCIEKÓW KOKSOWNICZYCH

Jednym z problemów ochrony środowiska jest usuwanie amoniaku ze ścieków przemysłowych, m. in. ze ścieków koksowniczych. W artykule omówiono szkodliwy wpływ amoniaku na środowisko i dokonano przeglądu różnych metod jego usuwania. Amoniak ze ścieków można usunąć albo przez jego rozkład, albo odzyskanie. Szczególną uwagę zwrócono na metody mające zastosowanie do ścieków koksowniczych.

УДАЛЕНИЕ АММИАКА ИЗ СТОЧНЫХ ВОД С ОСОБЕННЫМ УЧЕТОМ КОКСОВЫХ СТОЧНЫХ ВОД

Одной из проблем охраны среды является удаление аммиака из промышленных сточных вод, м.др. из коксовых сточных вод. В статье обсуждено вредное влияние аммиака на среду и сделан обзор разных методов его удаления. Аммиак из сточных вод можно удалить или посредством его разложения, или восстановления. Особенное внимамие было обращено на методы, применяемые для коксовых сточных вод.