自来水厂除锰滤砂的催化活性分析*

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摘要 研究来自我国东北 2 个自来水厂的除锰滤砂对 Mn^{2+} 的催化活性, 以确定微生物在除锰过程中的作用. 利用 PYCM 培养基可在每 g 湿砂上计到 10^5– 10^6 个细菌, 其中有 40%– 50% 具有 Mn^{2+} 氧化能力. 对滤砂表面的微生物进行原位培养, 灭菌和 $HgCl_2$ 抑制, 表明微生物群落的存在与稳定对于活性表面的存在与稳定是至关重要的. 当生物活性被抑制后, 锰砂残余活性约为原活性的 20%. 这部分活性可能来源于化学催化. 微生物是除锰滤砂活性的主要来源.

关键词 地下水, Mn^{2+}, 微生物, 化学因素, 滤砂.

国内外普遍采用接触过滤法(快过滤法)去除地下水中的 Mn^{2+} [1, 2]. 对于该法除锰机理的认识, 化学法除锰和生物法除锰已争论多年[1, 3]. 对机理的认识不同, 直接影响工艺参数的控制, 因而影响除锰效果、生产周期和生产稳定性. 为了尽快形成较为完善的工艺设计规范以取得良好的社会效益和经济效益, 有必要对滤砂催化氧化的机理进行深入的研究和认识.

本文对鞍山大赵台自来水厂和抚顺经济开发区自来水厂的除锰滤砂进行了研究, 对微生物及可能的化学因素在除锰过程中的作用进行了初步的研究和探讨.

1 材料和方法

1.1 材料

滤砂: 未使用过的锰砂(简称生料); 鞍山大赵台水厂与抚顺开发区水厂除锰滤池中经驯化的成熟锰砂(简称熟料); 抚顺水厂中试试验柱中驯化成熟的石英砂.

试剂: N, N, N′, N′-四甲基对苯二胺(简称 TMPD), Aldrich 出品, AR; 其它均为国产分析纯或生物纯试剂.

1.2 细菌计数与分离

PYCM 培养基: 蛋白胨 0.8g, 酵母浸膏 0.2g, $MnSO_4 \cdot H_2O$ 0.2g, K_2HPO_4 0.1g, $MgSO_4 \cdot 7H_2O$ 0.2g, $NaNO_3$ 0.2g, $CaCl_2$ 0.1g, $(NH_4)_2CO_3$ 0.1g, 加水 1000ml, 调 pH6.8– 7.2. 固体培养基则加琼脂 1.5%. 湿热灭菌后使用.

取适量熟料, 加无菌水充分振荡. 将振荡后得到的悬浊液梯度稀释, 用 PYCM 培养基进行混合平板培养, 25℃, 15d.

1.3 细菌活性的判定

刮取少量菌落, 分别用过硫酸法[4]和 TMPD 法[5]测定其中的锰. 过硫酸法结果呈红色且 TMPD 法结果呈蓝色的菌落是具有 Mn^{2+} 氧化能力的, 其它情况均表明菌落无活性.

从上述检验有活性的菌落中随机选取 5 个, 用 TMPD 培养基进行摇瓶培养, 25℃, 100r/min, 7– 10d. 100×g 离去培养液中的沉淀, 上清液 3000×g 离心 10min, 将沉淀用 10mmol/L, pH= 7.0 的 Tris-HCl 缓冲液悬浮, 再离心, 重复 2 次. 离心机, 缓冲液均预冷至 4℃. 菌体再次用同上缓冲液悬浮, 调节菌浓度至 OD_{600}= 1.0. 取上述菌悬液 10ml, 加 $MnSO_4$ 至 20mg/L, 静置 12h, 用 TMPD 法测定其中的高价锰. TMPD 法具体操作见文献[5]. 改进之处在于, 测定吸光度之前将菌体离心除去.

* 国家建设部“八五”科技攻关项目

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收稿日期: 1996-06-25

1.4　微生物群落与熟料表面结构稳定性关系

(1) 细菌增殖的影响　取适量熟料, 填装成内径 2.5cm, 高 60cm 的滤柱, 用 1/10 浓度的 PYCM 培养基淋洗. 用原子吸收法检测流入和流出液中的 Mn^{2+} 浓度.

(2) 灭菌的影响　取适量熟料和成熟石英砂灭菌, 填装成内径 2.5cm, 高 60cm 的滤柱, 用含锰约 1.40mg/L 的地下水淋洗, 测定进水和出水的 Mn^{2+} 浓度.

1.5　滤砂活性分析

(1) 滤砂的处理　取适量成熟锰砂, 湿热灭菌. 另取适量成熟锰砂用 1% $HgCl_2$ 溶液浸泡 72h. 经上述处理的滤砂, 从其上分离细菌以检验微生物被抑制的程度.

(2) 将经不同处理的滤砂分别填装成内径 2.5cm, 高 60cm 的滤柱. 过滤用水为含 Mn^{2+} 约 1.40mg/L 的地下水. 用原子吸收法分别测定各滤柱进水和出水的 Mn^{2+} 浓度, 计算 Mn^{2+} 去除率.

2　结果和讨论

2.1　细菌计数与活性检定

表 1　熟料表面的细菌数[1)]

滤料来源	总菌数/个·g^{-1}湿砂	具有 Mn^{2+} 氧化能力的细菌数/个·g^{-1}湿砂
鞍山大赵台	6.2×10^5	2.5×10^5
抚顺开发区	5.5×10^6	2.8×10^6

1) 指能在 PYCM 培养基上生长的细菌

从表 1 中可以看出, 在成熟滤料表面存在着不少于 10^5– 10^6 的细菌, 其中至少有相同数量级的细菌具有 Mn^{2+} 氧化能力. 由于有些细菌在滤砂表面吸附得较牢固[6], 而且有些细菌未必适于在 PYCM 培养基上生长, 滤砂表面上细菌(包括有 Mn^{2+} 氧能力的细菌)的数量应该比表 1 中所示的数量大.

判断细菌是否有氧化能力时发现, 棕色的菌落无一例外都具有 Mn^{2+} 氧化能力, 而其它颜色的菌落(白色、黄色、红色等) 都不具有 Mn^{2+} 氧化能力. 这种棕色物质是锰的高价氧化物[7].

摇瓶培养得到的细菌的活性测定结果见表 2.

表 2　棕色菌落菌悬液活性

菌株编号	1	2	3	4	5
被氧化的 Mn^{2+} /nmol·d^{-1}	140	120	80	65	70

表 2 所示的结果, 进一步证实了形成棕色菌落的细菌具有催化 Mn^{2+} 氧化的能力.

细菌的进一步纯化和活性定位正在研究中.

2.2　微生物群落与熟料表面结构稳定性关系

(1) 细菌增殖的影响　结果见图 1.

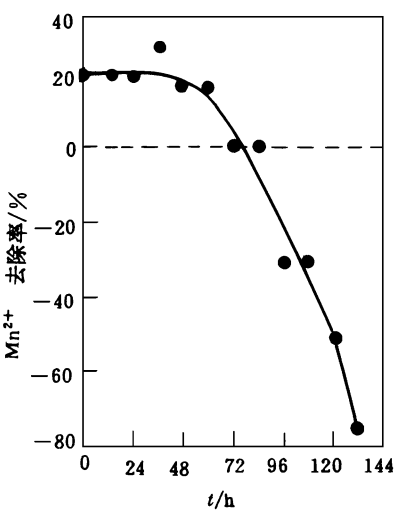

图 1　锰砂滤柱对 1/10-PYCM 培养基中 Mn^{2+} 的去除

图 1 曲线大致分为 2 部分. 其一, 72h 以前, 除锰率稳定在 20% 左右, 滤柱对培养基中的 Mn^{2+} 有一定的去除能力; 其二, 72h 以后, 除锰能力急剧下降, 96h 后不但不能除锰, 反而发生了 "漏锰" 现象(出水锰含量高于进水锰含量). 这说明锰砂表面沉积的锰又脱落下来了. 由于培养基灭菌后的溶氧度远低于曝气后的地下水, 该淋洗条件可能更适于厌氧或兼性厌氧菌的生长, 从而破坏了滤砂表面的微生态. 滤料表面上某些能够在厌氧或兼性厌氧条件下还原 Mn(Ⅲ, Ⅳ) 的细菌大量繁殖后, 就会导致已经

沉积在滤砂表面的 Mn(Ⅲ, Ⅳ) 被还原成可溶的 Mn^{2+}, 从而又脱落下来.

(2) 灭菌的影响　结果见图 2.

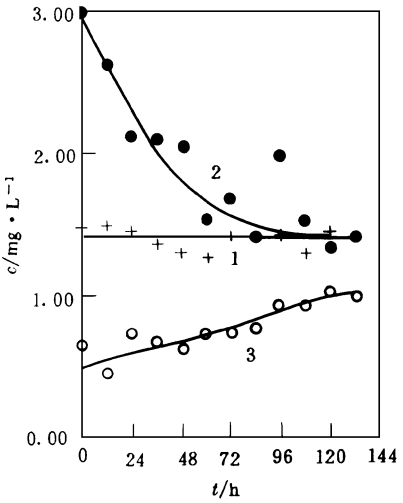

图 2　灭菌石英砂(熟砂) 和灭菌锰砂(熟料) 对地下水中 Mn^{2+} 的去除

1. 滤柱进水锰含量　2. 灭菌石英砂滤柱出水锰含量　3. 灭菌锰砂滤柱出水锰含量

从图 2 发现, 灭菌石英砂发生了严重的 '漏锰 '现象(曲线 2), 出水锰含量最初高达 2.98mg/L, 是进水锰含量的 2 倍. 随着时间的推移, 出水锰含量逐渐降低, 最后稳定在与进水相当的程度. 在光学显微镜下可以观察到, 随着石英砂的成熟, 其表面逐渐形成了一层黑色的膜. 灭菌后, 这层黑膜大量脱落, 重新暴露出石英砂表面. 已经验证黑膜中存在一种含锰的化合物[8], 它是石英砂催化 Mn^{2+} 氧化的产物. 同时, 黑膜也是其催化 Mn^{2+} 氧化的活性表面. 当细菌被杀灭后, 其活性表面也随之崩解. 脱落下来的黑膜使得其出水锰浓度高于进水锰浓度. 由于石英砂对 Mn^{2+} 的吸附能力非常弱[1], 所以灭菌石英砂在失去了其活性表面后, 对地下水中的 Mn^{2+} 几乎没有去除能力, 因而该滤柱的出水锰含量最终与进水相当.

从图 2 中的曲线 3 可以发现, 锰砂熟料灭菌后并未发生 '漏锰 '的现象, 这说明其表面物质并未脱落. 这可能是由于沉积的锰氧化物在形成过程中, 已与锰砂(化学成分是锰的氧化物) 原有的化学结构形成了化学键, 因而结构较稳定. 但是也发现, 此时锰砂除锰能力已降低. 这表明其表面活性结构也受到了一定的破坏.

综上笔者认为, 滤砂表面的微生物群落与其催化形成的含锰沉积物共同组成了滤砂的活性表面. 微生物群落的存在与稳定对于活性表面的存在与稳定是至关重要的. 微生物群落受到某种破坏, 能够导致表面结构的破坏, 甚至完全崩解.

2.3　滤砂活性分析

从灭菌和经 $HgCl_2$ 处理的熟料上未能分离到细菌. 说明其表面的细菌已被杀灭, 生物催化活性已不存在.

处理后的滤砂活性见图 3.

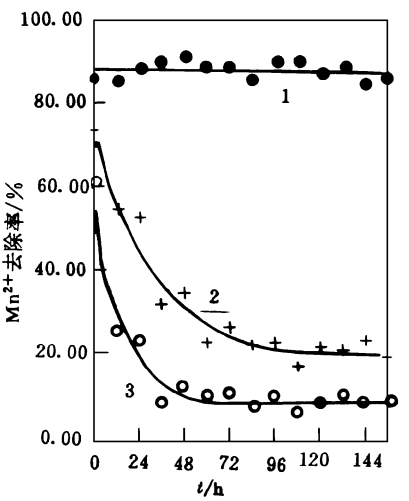

图 3　锰砂熟料在灭菌和 $HgCl_2$ 处理后对 Mn^{2+} 的去除能力

1. 锰砂熟料的除锰率　2. 湿热灭菌的锰砂熟料的除锰率　3. 经 1% $HgCl_2$ 溶液处理的锰砂熟料的除锰率

从图 3 可以看到, 当熟料上的细菌被杀灭后, 滤料对 Mn^{2+} 的去除能力有所下降(从 90% 降到 60% – 70%). 熟料除锰能力是比较稳定的(曲线 1); 而处理后的滤料的除锰能力是可饱和的, 说明此时滤料是通过吸附作用来除锰的. 未处理前, 吸附的 Mn^{2+} 被迅速氧化而形成新的可吸附表面, 因而活性是稳定的; 处理后, 生物催化活性被抑制, 吸附的 Mn^{2+} 不能迅速氧化, 所以可吸附表面逐渐被饱和.

从图 3 中还可以看到, 处理后的滤料的活

性在下降到一定程度后保持平稳. 这表明, 此时滤料表面的 Mn^{2+} 氧化速度与吸附速度达到了新的平衡. 由于细菌已被杀灭, 生物催化活性已不存在, 此时的催化活性应归因于化学催化. 笔者发现熟料表面存在着一种含锰化合物, 其结构与六方晶系的 $Al_5Mn_{18}\cdot 8H_2O$ 相似, 含 Ca∶Mn∶Fe≈10∶70∶15(原子)[8]. 很可能这种物质就具备催化 Mn^{2+} 氧化的能力.

通过曲线 2、3 的比较发现, 经 $HgCl_2$ 处理的熟料的活性比灭菌熟料的活性要低. 这可能是由于 Hg^{2+} 与 Mn^{2+} 有竞争性吸附的原因。因而, 灭菌熟料(曲线 2) 更接近于熟料原来具有的表面化学状态. 从曲线 2 可以推知, 化学活性约占总活性的 20%. 不过这个比例未必具有普遍意义, 因为 Mn^{2+} 催化氧化的产物的结构, 也就是滤料的表面化学结构取决于反应所处的热力学环境[9], 不同结构的产物未必具有相同或相近的催化氧化能力.

3 结 语

在熟料表面存在着一个复杂的微生物群落, 其中有大量的具有 Mn^{2+} 氧化能力的细菌. 这个复杂的微生物群落的存在与稳定对于滤料活性表面的存在与稳定是至关重要的. 来自于 Mn^{2+} 氧化细菌的活性, 是滤料除锰活性的主要部分. 化学催化活性不仅只占了一小部分, 而且它本身也是生物催化氧化的产物. 因此, 微生物(Mn^{2+} 氧化细菌及其它组成微生物群落所必需的微生物) 在除锰过程中扮演了重要的角色. 在进行工艺设计时应当充分考虑这一点, 为微生物群落的生长和稳定提供条件.

致谢 感谢吉林大学分子生物学系王宗睦、刘兰英、朴真三等教授在实验过程中给予的帮助. 感谢吉林大学化学系门瑞芝、邹明珠、白求恩医科大学侯芳玉等教授对实验工作所惠予的宝贵意见. 石玉华同志协助整理本文, 在此也向她表示感谢.

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dexes controlled in producing.

Key words: dye intermediates, J-acid, wasteliquor, extraction, resource recovery.

Study On Full-scale Test of Biological Contact Oxidation Pretreatment in Drinking Water Treatment from Huaihe River Source Water (Bengbu Reach). Liu Wenjiun, He Beiping et al. (Dept. of Environ. Eng., Tsinghua University, Beijing 100084), Lu jianhong et al. (Bengbu Water Company, Bengbu, 233000): *Chin. J. Environ. Sci.*, **18**(1), 1997, pp. 20- 22

In this study, the test of full-scale biological contact oxidation pretreatment in drinking water treatment was discussed, which is first in domestic. The results demonstrated that biological pretreat process can remove organic compounds and ammonia of source water by 13. 6%- 20. 5% and 70%- 90% respectively when the ratio of water to air is 1 ∶ 1; the key factor to affect the biological pretreat process performance is dissolved oxygen and temperature.

Key words: drinking water, biological pretreatment, pilot scale test, Huaihe River.

Degradation of Black Liquor Lignin Produced from Kraft Pulping Process of Pine by White-Rot Fungi. Lin Lu, Yang Gao et al. (State Key Laboratory of Pulp and Paper Engineering, South China University of Technology, Guangzhou, 510641): *Chin. J. Environ. Sci.*, **18**(1), 1997, pp. 23- 25

Black liquor lignin is the main pollutant in the black liquor produced from kraft pulping process of paper-making raw materials. In this paper, effect of white-rot fungi on degradation of black liquor lignin produced from pine kraft cook was studied. Results showed that white-rot fungus could degrade more than 74. 5% of black liquor lignin in the medium after 10 days of culture, the main part of black liquor lignin degraded was in the range of 1500- 3000kD of molecular weight. Culture factors such as cabon and nitrogen source, pH value in the medium and temperature exerted during the culture had an important role respectively on the effect of degrading black liquor lignin by white-rot fungus.

Key words: white-rot fungus, black liquor from kraft pulping process, sulfonate lignin, biodegradation.

Pilot Scale Petrochemical Wastewater Treatment Using Inner Loop Fluidized Bed Bioreactor. Zou Ping, Wang Chengwen and Qie Yi (Dept. of Environ. Eng., Tsinghua University, Beijing 100084): *Chin. J. Environ. Sci.*, **18**(1), 1997, pp. 26- 29

A Pilot scale experiment on petrochemical wastewater treatment using inner loop fluidised bed bioreactor and floatation process was conducted. The effluent COD from the process is about 200 and 100 mg/L when influent COD is 800 and 500mg/L, respectively. The loading rate of the bioreactor can be achieved above 15kgCOD/(m^3.d).

Key words: inner loop fluidised bed bioreactor, petrochemical wastewater, floatation process.

Investigation of the Landfill Gas Composition and Its Yield in South China. L. Y. Chan and S. C. Lee (Dept. of Civil and Structural Engineering, The Hong Kong Polytechnic University, Hong Kong), Y. Qin (Institute of Environmental Science, Zhongshan University, Guangzhou, 510275): *Chin. J. Environ. Sci.*, **18**(1), 1997, pp. 30- 34

Five landfill gas monitoring wells were installed and the composition of landfill gases were monitored in Wufengshan landfill in Foshan, south China. For the wells located in the late landfilled region, CH_4 and CO_2concentrations of landfill gases are high and stable. For the wells located in the early landfilled region, CH_4 and CO_2 concentrations of landfill gases are low and variable. In the last field measurement, the gases in the well located in early landfilled region has lost the characters or landfill gas. It's implication is that the biological decomposition process of the refuse underground has completed or the anaerobic environment has been destroyed. It just lasted for about 4 years and is much shorter than the expected time of 10- 20 years. The differences of landfill gas between Foshan Wufengshan landfill and Hong Kong Shuen Wan landfill were compared and discussed. The yield of landfill gas in Wufengshan landfill was estimated according to the original carbon component of the refuse.

Key words: landfill, waste gas, biological decomposition, monitoring well, CH_4, CO_2, gas yield, Foshan.

Photolysis of α-Naphthaleneacetic Acid in Aqueous Solution. Zufei Zhou, Weichuan Jiang and Weiping Liu (Dept. of Chemistry, Zhejiang University, Hangzhou 310027): *Chin. J. Environ. Sci.*, **18**(1), 1997, pp. 35- 37

Photolysis of α-naphthaleneacetic acid(NAA) has been investigated at 25℃ in aqueous solutions by irradiation at different wavelengths. The shorter wavelength of 254nm is considerably more effective in promoting degradation than wavelength of 365nm. The primary degradation of NAA follows a pseudo-first-order kinetics. The photolysis half-life and rate constant were determined to be 60min and $1.15\times10^{-2}min^{-1}$ respectively. The optimum photolysis rate has been observed using TiO_2 powder as photocatalyst. Several reaction intermediates were identified using GC/MS technique. The photolysis of NAA involves decarboxylation and oxidation on aromatic ring. On the basis of the analytical data, a mechanism of the process has been proposed.

Key words: photolysis, α-naphthaleneacetic acid, ultraviolet light.

Mn^{2+}-Oxidizing Bacteria and the Mn^{2+}-Removing Activity of the Filter Sand Used in Water Plants. Bao Zhirong et al (Dept. Molecular Biology, Jilin Univ,

Changchun 130023), Xu Aijun et al (Chinese Northeast Institute of Municipal Engineering Design, Changchun 130021): *Chin. J. Environ. Sci.*, **18**(1), 1997, pp. 38-41

The Mn^{2+} removing activity of the filter sand from 2 Chinese Water Plants was analyzed to determine the role of the bacteria in the Mn^{2+} removing Procession. Enumeration of bacteria on PYCM medium showed that there were 10^5-10^6 bacteria per g wet sand and about 40%-50% of the colonies had the ability to oxidize Mn^{2+}. By the in situ enrichment of the bacteria, sterilization and the $HgCl_2$ inhibition of the mature sand, it was found that bacteria were indispensable to the maintenance of the activity of the sand. When the bacterial activity was inhibited, the activity of the sand was reduced to20% of the original one. The remaining activity might be due to the chemical catalysis. Bacteria were the major source of the Mn^{2+} removing activity of the filters.

Key words: groundwater, Mn^{2+}, bacteria, chemical factors, filter sand.

A Study on the Characteristics of the Activated Sludge for Anaerobic Attached Microbial Film Expanded Bed Process. Zhang Jianli and Li Lijian (Dept. of Food Science, Laiyang Agricultural College, Laiyang 265200), Feng Xiaoshan (Dept. of Environ Science, Zhejiang Agricultural University, Hangzhou 310029): *Chin. J. Environ. Sci.*, **18**(1), 1997, pp. 42-44

The characteristics of activated sludge in the anaerobic attached microbial film expanded bed (AAFEB) reactor were studied. The results showed that there were three consecutive phases in the course of biofilm formation and development, namely, adsorbtion phase, partly coating phase and fully coating phase. In this process, the predominant microorganisms were changed gradually from coccus to filamentous organisms, which caused anaerobic sludge activity increasing. Under the acidification condition, there were a lot of streptococcus and extracellular polymer on the surface of activated sludge, and the sludge activity was low.

Key words: anaerobic attached microbial film expanded bed reactor, anaerobic activated sludge, biofilm.

Catalytic Properties of Two Kind of Catalysts in Toluene Combustion Reaction. Li Shiyao, Li Shulian et al. (Dalian Institute of Chemical Physics, Chinese Academy of Sciences, Dalian 116023): *Chin. J. Environ. Sci.*, **18**(1), 1997, pp. 45-47

Toluene organic exhausts were regarded as index reaction in evaluating the catalytic performance of honeycomb ceramic monolith catalysts consisting of noble metals and non-noble metals respectively by means of a continuous system with a fixed bed of catalysts. The effect of toluene concentration and oxygen content in the exhaust, space velocity, linear velocity on toluene reaction, and the thermal stability of catalysts were investigated. It is found that the activity of noble metal catalyst is superior than that of non-noble metal catalyst under different conditions. The light-off temperature of toluene exhaust on non-noble metal catalyst is by 50℃ higher than that on noble metal catalyst after catalysts calcine at 600℃ for 3h. Meanwhile, after catalysts calcine at 900℃ for 3h, the light-off temperature of toluene exhaust on noble metal catalyst increases only by 3℃ as compared with the calcination of 600℃ for 3h. But under same condition, the light-off temperature of toluene exhaust on non-noble metal catalyst increases by 87℃.

Key words: combustion reaction of toluene exhaust, honeycomb ceramic monolith catalyst, thermal stability, light-off temperature.

Photochemical Disinfection of Wastewater. Kong Lingren, Chen Xi et al. (Dept. of Environ. Sci. and Eng., Nanjing University, 210093): *Chin. J. Environ. Sci.*, **18**(1), 1997, pp. 48-50

A new method of photochemical disinfection for wastewater from Nanjng city was investigated. By aerating and using methylene blue(MB) as photosinsitizer, the wastewater samples were disinfected under sunlight and a medium pressure mercury lamp separately. The results were as follows: (1) The disinfection of wastewater were remarkably affected by the light sources, light intensity, irradiaded time, MB concentration and dissolved oxygen; (2) The bacteria which were disinfected by UV could be partially photoreactivated under sunlight; (3) The disinfection rate could reach 100% and the bacteria photoreactivation were not appeared when 1 liter of the sample containing 3.1×10^6 bacteria and 2 mg MB were irradiaded by a 300W medium pressure mercury lamp for 4 min; (4) The residual MB in the samples could be removed by bentonite clay. The disinfection mechanisms of UV and photosensitization, the bacteria photoreactivetion and the effects for affecting disinfection were discussed.

Key words: photochemistry, photosinsitization, UV irradiation, disinfection.

Summary of Studies on the Ecology of Lake Donghu. Liu Jiankang and Huang Xiangfei (Institute of Hydrobiology, Chinese Academy of sciences, Wuhan 430072): *Chin. J. Enwiron. Sci.*, **18**(1), 1997, pp. 51-53

Taking the Donghu (in Wuhan), a representative lake of the middle and lower basins of Chang Jiang River as a base, the present project has conducted stationary monitoring and systematic researches on the ecology of Lake Donghu for more than 30 years. Achievements of the studies include the estimation of the budgets for the main nutrients nitrogen and phosphorus of the lake, as well as their distribution and accumulation in