硫酸根对有机废水厌氧生物处理的影响

刘　燕

（华东化工学院，上海　200237）

摘要　利用连续流上流式厌氧污泥床在控制和不控制 H_2S 浓度两种情况下，考察 SO_4^{2-} 对有机物厌氧生物处理的影响．试验表明，硫酸根本身对厌氧处理没有毒害作用，它对厌氧处理过程的破坏主要是其还原产物 H_2S 造成的．在用 Fe^{2+} 控制 H_2S 的情况下，硫酸根的存在和浓度大小对出水 TOC、TOC 去除率、产气量、气化率无不利影响，但会使气体中甲烷含量及甲烷产率逐渐减小而二氧化碳含量及二氧化碳产率逐渐增大．

关键词　硫酸根，厌氧生物处理，甲烷，有机废水．

多种 SO_4^{2-} 含量很高的废水，如脂肪酸废水、糖蜜酒精废水、土霉素废水、麦迪霉素废水和味精废水等不易处理，许多人认为这是 SO_4^{2-} 起了抑制作用[1-3]．然而 SO_4^{2-} 对厌氧生物处理的影响至今还没有成熟的、公认的结论[4-7]，有必要进行深入细致的研究并寻找控制方法．本文通过 UASB 反应器的动态试验，在控制和不控制 H_2S 浓度两种条件下，研究了硫酸根对厌氧生物处理过程的影响，以找出含 SO_4^{2-} 的有机废水厌氧生物处理失败的原因、控制对策及 SO_4^{2-} 本身对有机废水厌氧生物处理的影响．

一、试验方法与材料

1. 试验装置

试验装置见图 1．其中 UASB 的反应体积为 5.69L，三相分离器部分 2.03L，反应器置于恒温箱内（35±1℃）．

2. 基质

为了排除不明因素的干扰，样水在实验室用自来水配制，碳源采用葡萄糖或可溶性淀粉，磷源采用 KH_2PO_4．在考察硫酸根综合影响时，氮源和硫酸根来源于硫酸铵．控制硫化氢浓度考察 SO_4^{2-} 本身的影响时，氮源采用谷氨酸钠．SO_4^{2-} 和用于控制 H_2S 浓度的 Fe^{2+} 来源于 $FeSO_4 \cdot 7H_2O$．

3. 分析项目和方法

TOC 用 TOC-500 型仪器，SO_4^{2-} 用重量法[8]，硫化物用碘量法[9]，有机酸用 103 型气相色谱仪；气体产量用 BSD-05 型湿式流量计；气体成分用 100 型气相色谱仪．

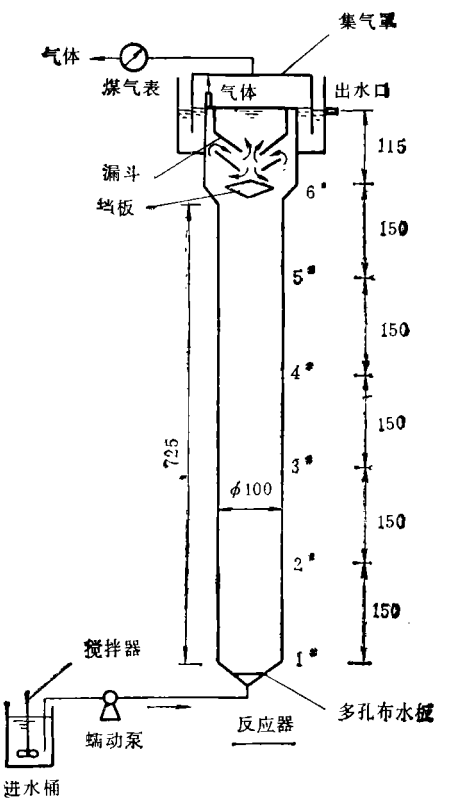

图 1　试验装置

二、试验结果

1. H_2S 是造成 SO_4^{2-} 对厌氧生物处理过程

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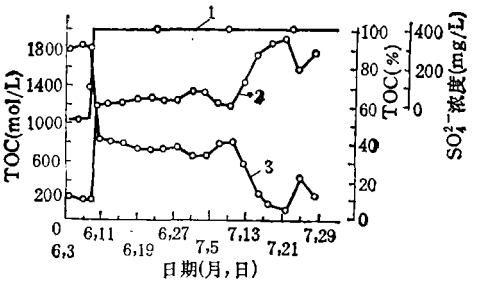

图 2　出水 TOC 浓度及 TOC 去除率的变化

1.进水 SO_4^{2-}　2.出水 TOC　3.TOC 去除率

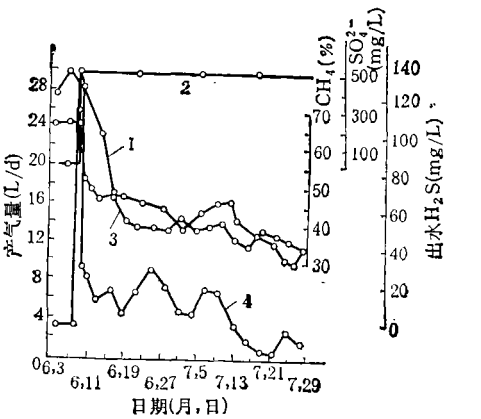

图 3　产气量和 CH_4% 的变化

1.出水 H_2S　2.进水 SO_4^{2-}　3.CH_4 含量(%)　4.产气量

破坏的主要原因.

图 2、3 表示当进水 TOC = 2000mg/L,流量为 600ml/h, 停留时间 9.48h 时, 一个正常运行的 UASB 反应器在不控制 H_2S 浓度情况下,500mg/L SO_4^{2-} 加入前后出水 TOC 浓度、TOC 去除率、出水 H_2S 浓度、产气量、CH_4(%) 含量等参数的变化情况. 样水加 $(NH_4)_2SO_4$ 后 24 小时, 有机物去除率由 90%陡然降至 40%, 产气量由 28L/d 降至小于 10.0L/d, CH_4 含量由 66%降至 50%. 同时, 出水 H_2S 从零上升到 135.3mg/L. 出水硫化氢浓度迅速上升表明硫酸还原菌的生长繁殖相当快, 能在较短时间内大量繁殖并利用发酵阶段的中间产物为电子供体还原 SO_4^{2-} 为 H_2S. 有机物去除率的下降表明反应器中起主要作用的微生物活性下降, 降解有机物的能力降低, 使稳定的处理过程受到一定程度的破坏. 产气量和气体中 CH_4 成分的减少表明甲烷菌已受到严重影响. 从产气量下降 2/3 而有机物去除率减少仅约 55% 可知, 甲烷菌受到的影响较大. 出水 pH 值从 7.0 下降至 5.0 和有机酸及挥发酸浓度的上升 (见表 1) 同样表明, 甲烷菌特别是利用乙酸的甲烷菌首先受到抑制, 但此时反应器尚有一定的降解有机物和产甲烷能力. 随着试验的进行, 处理效率继续下降, 有机物去除率降至 33%, 产气量下降到 4.1 L/d, 甲烷含量降至 37%. 即使加入 NaOH 和 Na_2CO_3 使进水 pH 值升至 9, 有机物去除率及产气量也未上升. 在样水中加入 SO_4^{2-} 500mg/L 后 6 周, TOC 去除率下降至 5%以下, 产气率下降到 0.3L/d/L, 可以说反应器中的生物作用基本上已停顿, 且难以恢复. 而用 Fe^{2+} 控制硫化氢的浓度考察 SO_4^{2-} 对厌氧反应的影响时, 未采取其它控制措施, 硫酸根浓度高达 1000mg/L, 厌氧反应器仍正常进行, 有机物去除率和产气量未受影响 (图 5、图 6). 由此可见, SO_4^{2-} 的还原产物 H_2S 是破坏厌氧反应正常运行的主要原因.

表 1　SO_4^{2-} 对有机酸和挥发酸的影响

SO_4^{2-} (mg/L)	挥发酸 (mg/L)	有机酸 (mg/L)	乙酸 (mg/L)	丙酸 (mg/L)	丁酸 (mg/L)	戊酸 (mg/L)	出水 pH 值
0	142	84.4	33.1	24.3	17.2	9.8	7.0
500	1628	603.1	435	87.8	80.3		5.0

2. 控制 H_2S 浓度时 SO_4^{2-} 对厌氧反应的影响

为了控制 H_2S 的浓度, 在样水中投加 Fe^{2+}, 其摩尔浓度大于或等于生成的 H_2S, 这一阶段试验中出水 H_2S 浓度均小于 1mg/L. 图 4—7 表示 H_2S 浓度控制试验结果, 试验条件为进水 TOC = 1100mg/L, 停留时间 11.4h. 从图 4—7 可以看出, 出水 TOC 浓度随样水中 SO_4^{2-} 浓度的增大而减小, 相应的 TOC 去除率随样水中 SO_4^{2-} 浓度的增大而略有增大 (线性相关系数分别为 0.9494, 0.9498, 出水 TOC 浓度低于 105mg/L, TOC 去除率高于 90%). 产气量和气化率与样水中 SO_4^{2-} 浓度大小无关 (产气量维持 13.0L/d, 气化率维持在 1.04 L/g 左右). 气体中 CH_4 含量逐渐降低而 CO_2 含量逐

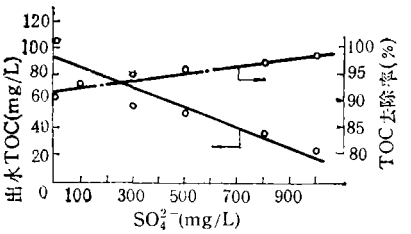

图 4　SO_4^{2-} 浓度与出水 TOC 及去除率的关系

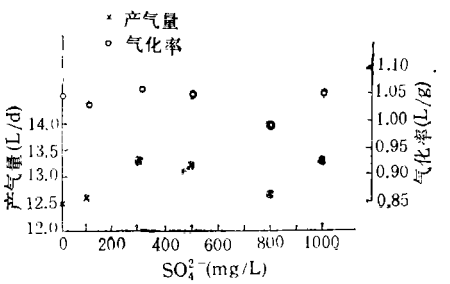

图 5　SO_4^{2-} 浓度与产气量和气化率的关系

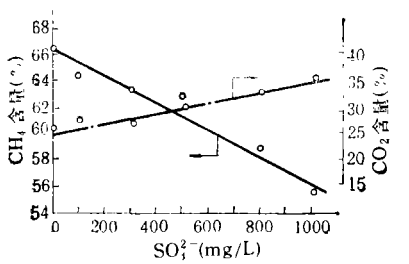

图 6　SO_4^{2-} 与气体中 CH_4% 和 CO_2% 的关系

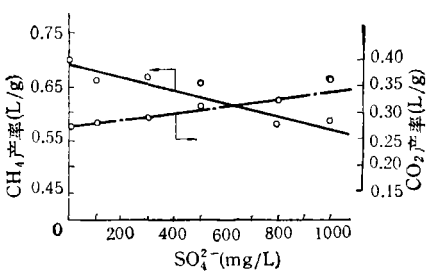

图 7　SO_4^{2-} 浓度与 CH_4 产率和 CO_2 产率的关系

渐升高(线性相关系数分别为 0.9736,0.96301). CH_4 产率逐渐下降而 CO_2 产率逐渐上升(线性相关系数分别为 0.9303, 0.9601).

三、分析与讨论

进水中 SO_4^{2-} 浓度大小对 TOC 的去除率没有不利影响，对产气量和气化率也没有不利影响，却影响气体中 CH_4 和 CO_2 含量. 即随着样水中 SO_4^{2-} 浓度的增高，气体中 CH_4 含量及 CH_4 产率呈线性减小，而 CO_2 含量及 CO_2 产率呈线性增加. 这是由有机物的厌氧生物降解过程、SO_4^{2-} 的氧化还原特性及硫酸还原菌在厌氧生物处理过程中的作用造成的. 试以葡萄糖经 EMP 途径酵解的最重要中间产物——丙酮酸为例讨论之.

无 SO_4^{2-} 存在时，丙酮酸的产氢产乙酸过程为:

$$CH_3COCOOH + H_2O \rightleftharpoons CH_3COOH + CO_2 + H_2 \quad (1)$$

$$\Delta G_f = -81.7\text{kj/反应}$$

有 SO_4^{2-} 存在时，丙酮酸的产氢产乙酸过程为:

$$CH_3COCOOH + \frac{1}{4}SO_4^{2-} \rightleftharpoons CH_3COOH + CO_2 + \frac{1}{4}S^{2-} \quad (2)$$

$$\Delta G_f = -111.2\text{kj/反应}$$

因此当有 SO_4^{2-} 和硫酸还原菌存在时，反应沿式(2)途径进行，但并不影响碳的转化形式，生成相同数量的乙酸和 CO_2. 而多数硫酸还原菌不能分解乙酸[10]，它们在处理系统中只是一种产氢产乙酸菌，不能参与产甲烷阶段的反应，不存在直接竞争有机底物从而抑制甲烷菌生长的问题. 本试验中，还原 SO_4^{2-} 的脱硫弧菌与甲烷八叠球菌和巴氏甲烷球菌在 UASB 反应器内大量存在，即是证据. 所以 SO_4^{2-} 对出水 TOC 浓度、TOC 去除率没有不利影响. 但从(2)式可以看出，当 SO_4^{2-} 存在时，无 H_2 产生，从而甲烷菌不能将 CO_2 经 H_2 还原生成 CH_4，使气体中 CH_4 含量和产率降低而 CO_2 含量和产率逐渐增加，但对总的产气量、产气率无影响.

四、结　论

1. SO_4^{2-} 本身对厌氧生物处理没有毒害作用，它对厌氧处理过程的破坏主要是其还原产物 H_2S 造成的.

2. 硫酸还原菌属于一种产氢产乙酸菌，它

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表7　处理系统中分离各类细菌对4种染料脱色的株数

细菌名称	分离出菌株数	阳离子红 2GL	活性红棕 KB_3R	酸性红 B 2GL	阳离子兰 RED
芽孢杆菌属	42	12	23	41	39
欧文氏菌属	14	9	*	12	13
假单胞菌属	36	28	31	33	36
黄单胞菌属	19	15	11	18	18
葡萄球菌属	24	21	12	22	16
气单胞菌属	30	6	24	24	24
邻单胞菌属	14	12	6	12	11
微杆菌属	11	7	7	11	7
志贺氏菌属	18	13	6	10	10

琼脂平皿上的菌落和电镜的细菌形态观察表明，分离菌和接入菌的鉴定结果及形态也是非常一致的。例如，接入菌 T-36 和分离菌E同属邻单胞菌属，菌落均为半透明，圆形肉红色凸起状。接入菌 S-62 与分离菌G也同属金黄葡萄球菌属。

由此可见，处理系统经近半年的运行，投入的脱色菌仍存在于反应器中，并占有优势。

三、小　结

1. 应用高效脱色菌和 PVA 降解菌接入厌氧-好氧处理系统，处理印染废水挂膜快、去除 PVA、CODcr、BOD_5、色度的活性高，并能保持处理效果的稳定性，可作为印染废水处理的种子。

2. 从运行期间细菌的种群看，高效菌接种的处理系统，染料脱色菌、PVA 降解菌以及厌氧菌和好氧菌总量等主要功能细菌数量多于活性污泥接种的处理系统。生物膜中的生物量高于沉泥或悬浮物。反应器中分离出的细菌种属，大部分与接入的高效菌类同，并占有优势，对染料的脱色效果亦相同。

致谢　在试验中得到北京第二印染厂、纺织部设计院和天津大学的大力支持和帮助，在此表示真诚的感谢。

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与利用乙酸的甲烷菌是互生关系而不是竞争关系。

3. 能形成难溶硫化物的金属盐有抑制 SO_4^{2-} 影响的作用。在用 Fe^{2+} 控制 H_2S 条件下，SO_4^{2-} 不影响有机物中碳的转化，对 TOC 去除率无不利影响；SO_4^{2-} 对产气量、气化率无影响，但它却造成无氧呼吸取代部分发酵，影响一部分 CH_4 的生成。

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dyeing wastewater, microbial distribution.

Preliminary Studies on the Buffering Effects of Forest Litter on Soil Acidification. Wang Silong, Chen Chuying (Institute of Applied Ecology, Academia Sinica, Shenyang 110015): *Chin. J. Environ. Sci.*, 1992, **13**(5), pp. 25—30

The ash element content and decomposition rate of the litter of *S. Superba* were found to be much higher than those of the litter of *C. lanceolata*. Subsequently, it is inferred that the former possesses a higher potential to buffer acidification of the soil compared with the latter. When the soil was covered with litter of *S. superba*, the base cation content and pH-value of leachate down from the litter and soil were evidently higher than the control; the basesaturation percentage and pH-value of the soil itself were also markedly higher, and the acidity and Al/Ca of the soil were far lower than the control. That is, the buffering ability of litter of *S. superba* to soil acidification is very strong, while the litter of *C. lanceolata*, is not.

Key words: *Cunninghamia lanceolata* (Lamb.) hook, *Schima superba* gardn. et champ, litter, buffering ability, soil acidificaton.

Studies on the Effects of Heavy Metals on the Contents of Nucleic Acids and Activities of Nucleases in the Root Tips of *Vicia faba*. Duan Changqua, Wang Huanxiao, Qu Zhongxiang (Biology Department, Yu nnan University. Kunming 650091): *Chin. J. Environ. Sci.*, 1992, **13**(5), pp. 31—35

Under the treatment of Pb^{2+}, Cd^{2+}, Zn^{2+} ions at low concentrations the contents of DNA (treated with the concentration less than 10.0, 0.50 and 50.0 ppm), and RNA (treated with the concentration less than 10.0, 50.0 and 50.0 ppm) and the activities of DNase (all less than 10.0 ppm) and RNase (less than 20.0, 10.0 and 50.0 ppm) in the root tips of *Vicia faba* L. increased with the increase of the concentrations of metal ions; however, the contents of DNA and RNA and the activities of DNase and RNase decreased when the concentrations of the metal ions were higher than the concentrations stated above. Hg^{2+} did not exert regular effects except that the activities of RNase increased with the treatment concentration of Hg^{2+} below 0.05 ppm, and decreased when the concentration of Hg^{2+} was above 0.05 ppm. The results demonstrate that the effects of heavy metals on nucleic acids metabolism depend on the interaction between the extents of the adaptive responses of plant tissues and the toxic action of heavy metal ions. Metal ion characteristics and the tolerant inducible system of different metabolism that might exist in plants, and the effects of protein metabolism on the adaptive actions of plants were also discussed.

Key words: *vicia faba* L., heavy metals, nucleic acids, nucleases, adaptive responses.

Research on Integer Programming Model of Urban Waste Water Reuse System. Wu Limei et al. (Beijing Municipal Research Institute of Environmental Protection, Beijing 100037): *Chin. J. Environ. Sci.*, 1992. **13**(5), pp. 36—41

A 0—1 integer programming model for the planning of urban wastewater reuse system were established. The planning of the wastewater collection system, location of the treatment plant and the efficiency of the treatment were formulated. Emphases were put on discussions of how to determine the user of the renovated water, the quantity of the reused water and the scale of additional advanced treatment so as to establish renovated water allocation system. The model was validated to be rational and applicable by examining a wastewater land treatment system.

Key words: urban wastewater reuse, integer programming, water allocation system, wastewater treatment.

A Study on the Characteristics of Phosphate Metabolism of *Pseudomonas* sp. Zhou Yuexi, Qian Yi, Gu Xiasheng (Department of Environmental Engineering, Tsinghua University, Beijing 100084): *Chin. J. Environ. Sci.*, **13**(5), 1992, pp. 42—45

The characteristics of the phosphate metabolism of *Pseudomonas* sp., the predominate genus in the activited sludge mixed liquor from the sequencing biological batch system which effectively removes nitrogen, phosphorus and organic matter, were investigated. Experimental results domonstrate: (1) Under aerobic conditions and with acetates as carbon source, the addition of sulphuric acid causes *Pseudomonas* sp. to release phosphates, and the addition of NaOH and $NaHCO_3$ causes pseudomonas sp. to excessively take phosphate. (2) The phosphate uptake capacity of *Pseudomonas* sp. is nearly the same in the aerobic system as in the anaerobic-aerobic system with acetate as carbon source.

Key words: waste water treatment, *pseudomonas* sp., phosphorus.

Study on Comprehensive Thectment of Waste Water from Furfural Production. Chen Yulian et al. (Department of Chemical Engineering, Taiyuan University of Technology, Taiyuan 030024): *Chin. J. Environ. Sci.*, 1992, **13**(5), pp. 46—49

During the process of furfural production, waste liquor containing 1—2.5 % acetic acid is discharged. Not onlv is acetic acid wasted but also the Surrounding environment is polluted. In the operation described in this paper, the waste water passes through a 400×1600 mm electrodialyzer equipped with 90—120 pairs of membrane modules, recovering acetic acid with concentrations of 18—20%. The treated waste water contains only 0.02% of acetic acid, and could be safely discharged or reused. Furthermore, an industrial graded acetic acid with purity of 99% could be produced by means of extraction-distillation of the recovered acetic acid solution. Industrialization of the comprehensive technology of electrodialysis-extraction-distillation for the treatment of the waste water from furfural production mentioned here has been achieved.

Key words: waste watert reatment, membrane separation, electrodialysis, acetic acid, recovery.

Effect of Sulfate Ion on Anaerobic Biological Treatment of Waste water. Liu Yan (Department of

Abstracts

Chinese Journal of Environmental Science

Environmental Engineering East China University of Chemical Technology, Shanhai 200237): *Chin. J. Environ. Sci.*, 1992, **13**(5), pp. 50—52

This paper describes the study on the effects of sulfate ion on the anaerobic biological treatment of wastewater under controlled or uncontrolled sulfide concentrations. A continually operated upflow anaerobic sludge bed (UASB) experimental reactor was used. It was found that sulfate ion itself is non toxic to anaerobic biological process. However, sulfate could be reduced to hydrogen sulfide which is toxic to the bacteria. When ferrous ion was used to control the concentration of hydrogen sulfide, sulfate did not exert any harmful effect on the removal efficiency of organic matter from the waste water and the gas producing rate, but it caused the gradual decrease of CH_4 content in the produced gas and methane yield and the increase of the yield of carbon dioxide as increasing the concentration of sulfate ion.

Key words: sulfate, anaerobic biological treatment, methane, wastewater.

A Study on the Strategy of the Utilization of Coal Fly-Ash Resources in Nanjing Area. Zuo Wensheng (Communication Planning and Designing Office of Zhenjiang, Nanjing 210003), Ye Liansheng (Department of civil Engineering, South-East University, Nanjing): *Chin. J. Environ. Sci.*, 1992, **13**(5), pp.53—57

This paper analyses the strategy, restrictive factors and policies of the utilization of coal fly-ash resources in Nanjing area by using systems engineering and decision-analysis method of operation research. A series of measures was put forward. This may lay a scientific foundation for the municipality of Nanjing to formulate the policy in the management of coal fly-ash in this area.

Key words: coal fly-ash strategy, hierarchcal analysis.

Changes of Ecological Environment in the Shiyanghe River Basin and Measures Proposed for Rational Utilization of Water Resources. Hou Yinwei, Li Shengcai, Wang Changming, Zheng Yi (Changchun college of Geology Science): *Chin. J. Environ. Sci.*, **13**(5) 1992, pp. 58—66

The authors present a new viewpoint that deterioration of ecological environment in the arid region the Shiyanghe River basin of northwest China, has originated from three causes: (1) people put undue emphasis on surface water controlling, and neglected the regulative function of natural ground water reservior; (2) water for farmland use was stressed but water for oasis use was neglected; (3) the backward way of irrigation leads so far to serious waste of water resources. In order to solve the problem, a proposed multi-goal decision-making model has been built for managing rational use of water resources in the Shiyanghe River basin. In this model, as the basin is divided into 16 sub-districts, the quantity of water for both oasis-farmland ecosystem and vegetation ecosystem necessities is considered as a main variable, and integrates with economic development. the investment of water resource development, amelioration of ecological environment etc 9 goals to be regarded. The result of the model operation shaws that deterioration of ecological environment in the basin will be in effective control.

Key Words: the Shiysnghe River basin, management of water resources, arid area, decision-making model

Sulfur Dioxide Removal from Flue Gas by Means of Active Carbon Catalysis——Dynamic Adsorption of SO_2 on Catalyst. Zhao Xiusong et al.(Dalian Institute of Chemical Physics, Chinese Academy of Sciences, Dalian 116023): *Chin. J. Environ. Sci.*, 1992, **13**(5), pp. 67—70

Influence of the main component (O_2 and water steam) in flue gas on the amount of adsorption and the dynamics of adsorption of SO_2 on active carbon catalysts were studied by flow method. It was found that when the content of O_2 is about 5% (V%) and the content of water steam is about 8%, adsorption amount of SO_2 on the catalyst reached the maximum value. Adsorption dynamics of SO_2 almost exactly fit the law of Elovich. The apparent adsorption activation energy (E_a^0), Elovich parameters and the relation between adsorption activation energy and temperature were hence obtained.

Key words: active carbon, flue gas, SO_2 adsorption activation energy.

A Study on the Adsorbability of a New Separation Adsorbent for Heavy Metallic Ions. Zhu Guoyi, Zhou Yanxiu, Wang Bingwu(Changchun Institute of Applied Chemistry, Chinese Academy of Sciences, Changchun, 130022): *Chin. J. Environ, Sci.*, 1992, **13** (5), pp. 70—73

The adsorbability of a new separation adsorbent containing sulfhydryl-activated carbon compounds for heavy metallic ions was studied. Experimental results indicate that in static state the equilibrium time for the adsorption is 1 hour for Cd^{2+} and Zn^{2+} and 1.5 hours for Pb^{2+} and Cu^{2+}. The sequence of adsorption velocity is $Pb^{2+}>Cu^{2+}>Zn^{2+}>Cd^{2+}$. When $[Mg^{2+}]<200$mg/L, $[Ca^{2+}]<400$mg/L and $[K^+]<350$ mg/L, the three kinds of ions do not interfere with the adsorption of Cu^{2+}. In dynamic state, the adsorbent could adsorb Hg^{2+} and Cu^{2+} ions completely under the following conditions: the amount of the adsorbent was 0.1g for Hg^{2+} or 0.15g for Cu^{2+}, pH was 0.3—8 for Hg^{2+} or 3—8 for Cu^{2+} and the velocity of flow < 7 mL/min for Hg^{2+} or <10mL/min for Cu^{2+}. In addition the saturated capacity of adsorption of the new separation adsorbent is about 3—20 times higher than that of sulfhydryl cotton fiber. The adsorbent has high adsorbability and good stability.

Key words: adsorption, heavy metallic ions, new separation adsorbent.

Clean-up of Soil Sample by GEL Permeation Chromatography. Sun Sien, Liu Xiufen (Research Center for Eco-Environmental Sciences, Academia Sinica, Beijing 100085): *Chin. J. Environ. Sci.*, 1992, **13** (5), pp. 74—78