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A study on the adsorption property of Cu(II) ions by chitosan immobilized on quartz sand

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Abstract : This paper is aimed at presenting a renovated approach to preparing a more cost-effective adsorbent based on chitosan immobilized on quartz with 1 g chitosan mixed with 20 g quartz sand. Dissolved with 25 mL 2.4 mol/L acetic acid, then neutralized with 20 mL 5 mol/L NaOH solution drop by drop until chitosan-coated sands were formed by precipitating the chitosan from the solution on the sand surface. The suggested method is expected to be highly useful and meaningful because large areas of land and vast water bodies have now been polluted by harmful metallic contaminants all over the world, which has brought serious dangers to the environment and human health. Furthermore, the so-far available metal-contaminant removal technologies are very limited for their expensive cost and negative side-effects. For example, chitosan is a kind of well-known and effective metal chelator, but its practical use is limited due to its relatively high costs. In spite of this, with our new adsorbent, it is easy to get purified after filtering, washing, drying and sieving. It can thus be used for copper(II) ions adsorption removal from aqueous solution. By using the new adsorbent, we have carried out adsorption experiments to explore the influence of adsorption time, pH value, adsorbent dosage, initial mass concentration of copper(II) ions, and grain diameter of quartz sand on the adsorption rate. The results of our experiments indicate that the removing power of chitosan immobilized on quartz sands can reach 91.57 % of copper ions(II) in solution under the optimum conditions performed with pH = 6.0, with adsorbing time about 30 min, and adsorbent dosage of 20 g/L. We have also explored the kinetics of the adsorption process by using Freundlich model and Langmuir model. The result of our investigation proves that the adsorption of copper(II) ions on chitosan immobilized on quartz sand can better be described by the Freundlich equation, whose linear correlation coefficient tends to be bigger than 0.98. Adsorption process accorded with first-order kinetic reaction, of which linear correlation coefficient is greater than 0.98. Thus, it can be concluded that chitosan immobilized on quartz sands can be effectively used to reconstruct the copper ions in the contaminated sewage.

Key words : water pollution prevention and treatment; chitosan; quartz sand; copper(II) ion; adsorption; kinetics

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基于ASM1的中药废水处理 数学模型研究 ——进水质组分(COD和氮)估计 *

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摘要:采用中药废水(两相厌氧消化系统出水)作为膜生物反应器(MBR)的进水,以活性污泥1号(ASM1)数学模型为基础,对中药废水的COD和氮(N)组分进行估计。结果表明,中药废水的COD和N组分不同于传统生活污水。中药废水COD组分中的 S_S 为141.2 mg/L, X_S 为2113.2 mg/L, X_1 为85.3 mg/L, S_1 为53.8 mg/L; N组分中的 S_{NH} 为20.93 mg/L, S_{NO} 为0.5 mg/L, S_{ND} 为17.6 mg/L, X_{ND} 为263.4 mg/L。组分估计是ASM1模型的输入项,它的正确性同模型的模拟结果直接相关,是模型参数的重中之重,同时对研究类似废水水质组分估计也有指导和借鉴作用。

关键词:水污染防治工程;膜生物反应器;中药废水;ASM1;
组分估计

中图分类号:X703.5

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0 引言

膜生物反应器(MBR)是一种新型污水处理技术,主要工艺特点是用膜分离单元代替传统活性污泥工艺中的二沉池,达到泥水分离的目的^[1-3]。目前有关MBR的大部分研究集中在生活污水和工业废水的运行效果、工程参数控制和活性污泥特性对膜污染的影响上^[4-6]。有关MBR废水处理数学模型的研究较少,如基于活性污泥1号(ASM1)模型的研究^[7,8]。数学模型研究在MBR工艺优化、故障检修、辅助设计和寻求最佳运行条件等方面都有十分重要的价值^[9,10]。

传统观点认为ASM1模型只适用于生活污水,不适用于工业废水^[7,8]。本文结合MBR运行动态试验,以哈尔滨中药二厂中药废水(两相厌氧消化系统出水)为研究对象,建立基于ASM1的中药废水处理数学模型。为了使ASM1模型能应用到MBR废水处理系统的设计和运行中,必须能够估计中药废水的参数值,还要能估计进水中各重要组分的质量浓度。本文的目的就是对基于ASM1的中药废水COD和氮(N)组分进行估计,并与传统生活污水水质(COD和N)进行对比。

1 装置与方法

1.1 试验用水的来源与水质

哈尔滨市中药二厂所排放的中药废水主要来自各车间生产过程中的洗药、煎煮、瓶罐清洗等工序,另有一部分来自管道及地面冲洗水、蒸汽冷凝水和处理离子交换树脂酸碱液的

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中和水等。该废水是一种污染物种类繁多、成分复杂的高浓度难降解有机废水,且间歇排放,虽无毒但有害,具有 COD 高、可生化性差、水质水量变化大、色度高等特点,处理难度极大。哈尔滨中药二厂采用以“CSTR 产酸发酵反应罐—UASB/AF 复合厌氧反应池—交叉流好氧反应池”为主体的工艺对中药废水进行处理。污水处理厂原水经过格栅、初沉池、调节池、换热罐、产酸反应器、产甲烷反应器和好氧反应池,最后经由二沉池出水。

本文以一体式膜生物反应器工艺(MBR)取代交叉流好氧反应池和二沉池,进行 MBR 处理两相厌氧消化系统出水的试验研究。两相厌氧消化系统出水水质见表 1。中药废水两相厌氧消化系统出水有机物含量高、悬浮物浓度高、pH 低,水质特性不同于传统生活污水。为建立基于 ASMI 的 MBR 处理中药废水的数学模型,非常有必要对中药废水两相厌氧消化系统出水水质组分进行估计。

1.2 试验装置

试验 MBR 工艺装置为自行设计,设在哈尔滨市中药二厂污水处理厂内。试验装置见图 1。哈尔滨中药二厂产甲烷反应器出水经污水泵(1)、水表(2)进入高位水箱(6),然后经闸阀(3)进入生物反应器(13),废水中大部分有机物经生物反应器内微生物自身分解代谢作用得到降解。含有大量未去除 SS 的混合液在真空抽水系统的作用下经过中空纤维膜组件(11)过滤出水。反应器的液位由液位自动控制系统控制。空气由空压机经压力缓冲罐和气体流量计后,由球冠状微孔曝气装置进入反应器,曝气装置的曝气量控制在 10~20 m³/h。反应器容积为 3.2 m³。试验设备用膜为天津膜天技术公司生产的聚偏氟乙烯中空纤维微滤帘式膜组件,膜孔径为 0.22 μm,中空纤维膜的内外径分别为 0.5 mm 和 0.8 mm。

1.3 试验方法

试验共持续 452 d,根据试验条件的不同,分为 3 个阶段,

各阶段试验条件列于表 2。表 2 中,污泥龄通过曝气池直接连续排放污泥来控制。试验过程中未对温度和 pH 值加以控制。COD、MLVSS、异养菌产率 Y_H 、耗氧速率(OUR)、氨氮 S_{NH} 、硝酸盐及亚硝酸盐氮 S_{NO} 、易生物降解有机氮 S_{ND} 等的测定均依照标准水质分析方法进行^[11]。

2 结果与讨论

2.1 中药废水中惰性有机物质 S_I 和 X_I 的估计

根据 ASMI,中药废水的底物可分为易生物降解底物和慢速生物降解底物两个部分。进水中总 COD 的组成为

$$COD_t = S_S + X_S + X_I + S_I \quad (1)$$

式中 S_S 为易生物降解底物,mg/L; X_S 为慢速可生物降解底物,mg/L; X_I 为惰性悬浮物有机物质,mg/L; S_I 为惰性可溶性有机物质,mg/L。

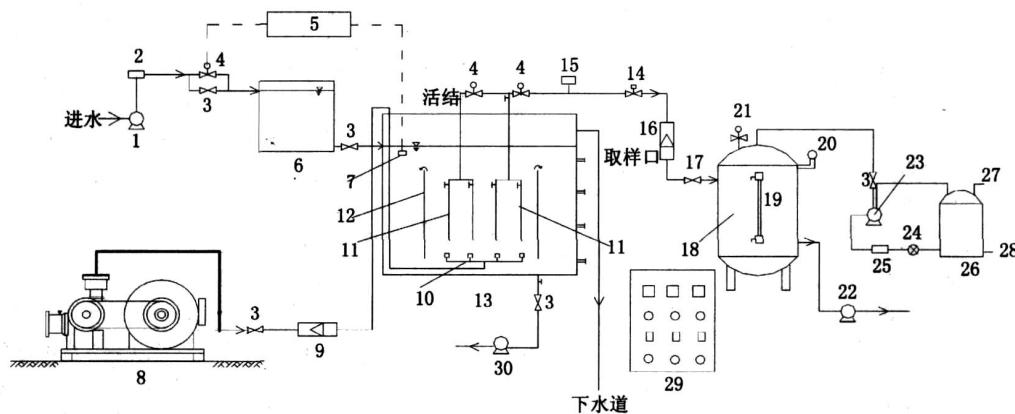
中药废水中溶解性有机物部分通过 0.45 μm 滤纸过滤后的过滤液测定。惰性溶解性有机物 S_I 的质量浓度通过滤纸过滤液进行试验,由 ASMI 模型确定。从完全混合式膜生物反应器(泥龄为 100 d)中取出部分混合液,稀释到悬浮固体质量浓度为 2 000~3 000 mg/L,在间歇试验中曝气;定期分析其可溶性 COD,其浓度可能恒定,也可能随时间减少。如果反应器中易生物降解 COD 小到可以忽略不计,将发生前一情况,否则将出现后面的情况。最终残留的溶解 COD 就是惰性物质,其值相当于进水中的 S_I 。由图 2 可见, S_I 为 53.8 mg/L。中药废水中的 S_I 值(53.8 mg/L)高于生活污水中的 S_I 值(25~40 mg/L),对比情况见表 3^[7,8,12]。

采用超声波预处理混合液,氧化分解易生物降解底物 S_S 和慢速可生物降解底物 X_S ,剩下的物质为惰性悬浮物有机物质 X_I 和惰性可溶性有机物质 S_I 。采用与测定惰性溶解性有

表 1 试验废水水质

Table 1 Characteristics of traditional Chinese medicine waste water in the tests

| COD/(mg L ⁻¹) | BOD ₅ /(mg L ⁻¹) | TN/(mg L ⁻¹) | TP/(mg L ⁻¹) | SS/(mg L ⁻¹) | pH |
|---------------------------|---|--------------------------|--------------------------|--------------------------|---------|
| 259.1~12 776.5 | 129.6~7 665.9 | 5~30 | 0.5~12 | 1 000~1 600 | 6.0~7.0 |



1—污水泵; 2—水表; 3—闸阀; 4—电磁阀; 5—液位控制器; 6—高位水箱; 7—液位传感器; 8—空压机; 9—气体流量计; 10—空气扩散装置; 11—膜组件; 12—隔板; 13—生物反应器; 14—稳压阀; 15—压力计; 16—液体流量计; 17—进水阀; 18—真空罐; 19—液位计; 20—真空表; 21—放气阀; 22—水泵; 23—水环真空泵; 24—球阀; 25—过滤器; 26—气水分离器; 27—排气口; 28—放水口; 29—电控柜; 30—排泥泵

图 1 试验装置流程示意图

Fig. 1 Schematic of MBR plant system

机物质 S_1 同样的方法测定,测定值减去溶解性惰性有机物部分就是颗粒性有机物 X_1 。由图3可见, X_1 为85.3 mg/L。中药废水中的 X_1 值(85.3 mg/L)高于瑞士、匈牙利和中国的生活污水中的 X_1 值(25 mg/L、70 mg/L、40 mg/L),低于丹麦生活污水中的 X_1 值(100 mg/L)。对比情况见表3^[7,8,12]。

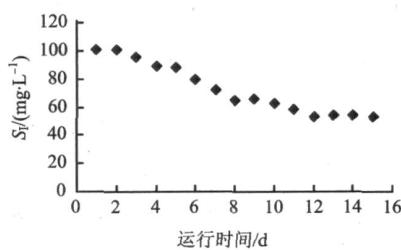
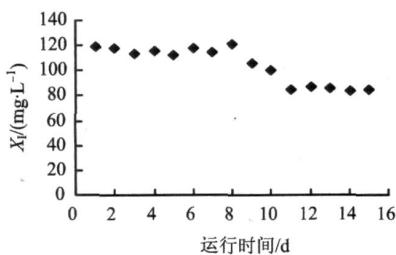
2.2 中药废水中可生物降解物质 S_S 和 X_S 的估计

在获得易生物降解物质 S_S 的质量浓度之前,必须知道异养菌产率 Y_H 。 Y_H 可以通过观察可溶底物去除过程中细胞物质的生成量来估计。废水首先经沉淀并滤去颗粒物,滤液中

表2 试验操作条件

Table 2 Operating conditions of the membrane bioreactor

| 项目 | 第I阶段 | 第II阶段 | 第III阶段 |
|--|-------|---------|---------|
| 运行时间/d | 1~155 | 160~307 | 310~452 |
| 污泥龄/d | 50 | 100 | 100 |
| 水力停留时间/h | 8 | 5 | 3.2 |
| 溶解氧/(mg L ⁻¹) | 2~4 | 2~4 | 2~4 |
| 膜通量/(L m ⁻² h ⁻¹) | 8.0 | 12.8 | 20.0 |
| 膜出水流量/(L h ⁻¹) | 400 | 640 | 1 000 |

图2 进水中 S_1 的测定结果Fig. 2 Results of S_1 in the influent图3 进水中 X_1 的测定结果Fig. 3 Results of X_1 in the influent

只含有可溶基质。从完全混合膜生物反应器中定期取出混合液,测量溶解性COD和总COD。异养菌的产率计算式为

$$\text{COD(细胞)} = \text{COD(总)} - \text{COD(可溶性)} \quad (2)$$

$$Y_H = \frac{\text{COD(细胞)}}{\text{COD(可溶性)}} \quad (3)$$

重复几次,就可得到大致的 Y_H 值(图4)。在估计过程中,任何误差都能在确定其他参数或进水质量浓度时得到补偿。从图4可以看出,3个阶段污泥产率都维持在比较低的水平。整个运行过程中污泥产率的平均值为0.56 kg/kg COD。第I阶段污泥产率的平均值为0.67 kg/kg COD;第II阶段污泥产率的平均值为0.47 kg/kg COD;第III阶段污泥产率的平均值为0.51 kg/kg COD。第I阶段污泥停留时间(SRT)为50 d,第II阶段和第III阶段SRT为100 d,而第III阶段污泥产率的平均值大于第II阶段和第III阶段污泥产率的平均值,这说明污泥产率随污泥浓度的增大而降低。

知道了 Y_H 值,在单个完全混合反应器中,SRT约为2 d的情况下,日循环脉冲式进水(进水12 h,停止进水12 h),可以通过测定其耗氧速率(OUR)估计进水中易生物降解底物质量浓度 S_S (该方法由Ekama等于1986年提出)。如图5所示,进水结束后,耗氧速率曲线立即快速下降。这是因为积累的易生物降解物质被迅速利用。然而OUR不会降至零,因为积累的慢速生物降解物质在一段时间内继续以相同的速率被利用。因此,OUR的立即下降只与易生物降解物质有关,可被用来计算其浓度。

$$S_S = \frac{\text{OUR} \cdot V}{Q(1 - Y_H)} \quad (4)$$

式中 OUR 为停止进水后 OUR 的变化,mg O₂/(L · h); V 为反应器的容积,m³; Q 为进水流量,m³/d。

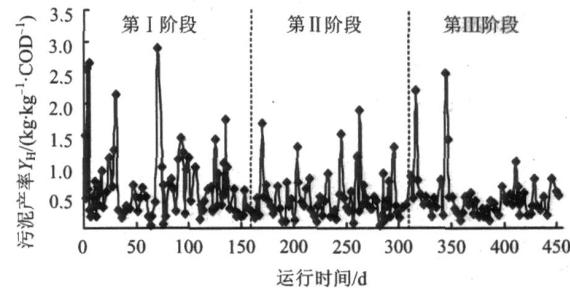
图4 污泥产率 Y_H 随运行时间的变化Fig. 4 Changes of the sludge yield (Y_H) in the SMBR during stages 1-3

表3 中药废水与生活污水特性对比

Table 3 Comparison of traditional Chinese medicine wastewater and domestic sewage

| 组分 | 意义 | 中药废水 | 生活污水 | | | |
|---------------------------|---|--------|------|-----|-----|------|
| | | | 丹麦 | 瑞士 | 匈牙利 | 中国 |
| COD/(mg L ⁻¹) | S_S 易生物降解基质 | 141.2 | 125 | 70 | 100 | 64 |
| | S_I 可溶性惰性有机物质 | 53.8 | 40 | 25 | 30 | 40 |
| | X_S 慢速可生物降解基质 | 2113.2 | 250 | 100 | 150 | 160 |
| | X_I 颗粒性惰性有机物质 | 85.3 | 100 | 25 | 70 | 40 |
| N/(mg L ⁻¹) | S_{NO} 硝酸盐与亚硝酸盐氮 | 0.5 | 0.5 | 1 | 1 | 0.5 |
| | S_{NH} NH ₄ ⁺ + NH ₃ 氮 | 20.93 | 30 | 10 | 30 | 12.5 |
| | S_{ND} 溶解性可生物降解有机氮 | 17.6 | 8 | 5 | 10 | 10.1 |
| | X_{ND} 颗粒性可生物降解有机氮 | 263.4 | 10 | 10 | 15 | 18.3 |

又已知 $V/Q = 12 \text{ h}$, 由图 5 可知, $S_S = \frac{\text{OUR} \cdot V}{Q(1 - Y_H)} = 141.2 \text{ mg/L}$ 。中药废水中的 S_S 值 (141.2 mg/L) 高于生活污水中的 S_S 值 ($64 \sim 125 \text{ mg/L}$), 对比情况见表 3^[7,8,12]。

确定了废水中的总 COD、易生物降解 COD、惰性溶解 COD 和惰性颗粒 COD, 就可以根据式 (1) 计算慢速生物降解 COD 值。图 6 为第一阶段中药废水中总 COD 随运行时间的变化情况。从图 6 可以看出, 进水平均 COD 为 2393.5 mg/L , 而膜出水平均 COD 为 33.7 mg/L , 去除率达 98.1% 。因此, $X_S = \text{COD}_t - S_S - X_I - S_I = 2393.5 - 141.2 - 85.3 - 53.8 = 2113.2 \text{ mg/L}$ 。中药废水中的 X_S 值 (2113.2 mg/L) 远高于生活污水中的 X_S 值 ($100 \sim 250 \text{ mg/L}$), 对比情况见表 3^[7,8,12]。

2.3 中药废水中 N 的估计

ASM1 模型的目的是预测单个污泥系统运行中碳氧化、同时硝化反硝化的情况, N 的描述很重要。氧化态的 N 有 4 种存在形式: 氨氮 S_{NH} 、硝酸盐及亚硝酸盐氮 S_{NO} 、易生物降解有机氮 S_{ND} 和慢速生物降解有机氮 X_{ND} 。中药废水中的 S_{NH} 、 S_{NO} 和 S_{ND} 质量浓度可通过过滤样品的适当分析来确定。中药废水中 N 的估计结果见图 7。

从图 7 可以看出, S_{NH} 平均值为 20.93 mg/L , S_{NO} 平均值为 0.5 mg/L , S_{ND} 平均值为 17.6 mg/L 。中药废水中的 S_{NH} 值 (20.93 mg/L) 低于丹麦和匈牙利生活污水中的 S_{NH} 值 (30 mg/L), 高于瑞士和中国生活污水中的 S_{NH} 值 (10 mg/L 和 12.5 mg/L)。 S_{NO} (0.5 mg/L) 低于生活污水的 S_{NO} ($0.5 \sim 1.0 \text{ mg/L}$), S_{ND} (17.6 mg/L) 高于生活污水的 S_{ND} ($5 \sim 10.1 \text{ mg/L}$)。对比情况见表 3^[7,8,12]。

如果进水中易生物降解和慢速生物降解有机氮之比类似于进水中易降解 COD 与慢速降解 COD 之比, 那么, 进水中慢速降解有机氮的质量浓度就可以通过易降解有机氮来求。

$$\frac{S_{ND}}{X_{ND} + S_{ND}} = \frac{S_S}{X_S + S_S} \quad (5)$$

因此, $X_{ND} = 263.4 \text{ mg/L}$, 远高于生活污水中的 X_{ND} 值 ($10 \sim 18.3 \text{ mg/L}$), 对比情况见表 3^[7,8,12]。

3 结论

1) 中药废水 (两相厌氧消化系统出水) 中的 S_S (141.2 mg/L) 和 X_S 值 (2113.2 mg/L) 要远高于生活污水中的 S_S 值 ($70 \sim 125 \text{ mg/L}$) 和 X_S 值 ($100 \sim 250 \text{ mg/L}$)。中药废水中的 X_I 值 (85.3 mg/L) 高于瑞士、匈牙利和中国生活污水中的 X_I 值 (25 mg/L 、 70 mg/L 和 40 mg/L), 低于丹麦生活污水中的 X_I 值 (100 mg/L)。中药废水中的 S_I 值 (53.8 mg/L) 高于生活污水中的 S_I 值 ($25 \sim 40 \text{ mg/L}$)。

2) 中药废水中的 S_{NH} 值 (20.93 mg/L) 低于丹麦和匈牙利生活污水中的 S_{NH} 值 (30 mg/L), 高于瑞士和中国生活污水中的 S_{NH} 值 (10 mg/L 和 12.5 mg/L)。 S_{NO} (0.5 mg/L) 低于生活污水 S_{NO} 的 ($0.5 \sim 1.0 \text{ mg/L}$), S_{ND} (17.6 mg/L) 和 X_{ND} (263.4 mg/L) 远高于生活污水的 S_{ND} ($5 \sim 10.1 \text{ mg/L}$) 和 X_{ND} 值 ($10 \sim 18.3 \text{ mg/L}$)。

3) 中药废水 (两相厌氧消化系统出水) 中 COD (S_I 、 X_I 、 S_S 、 X_S) 和 N (S_{NO} 、 S_{NH} 、 S_{ND} 、 X_{ND}) 组分不同于生活污水, 但也可用于 ASM1 建模。

4) 结果表明, 该模型可以很好的模拟 MBR 处理中药废水两相厌氧消化系统出水的动态过程。

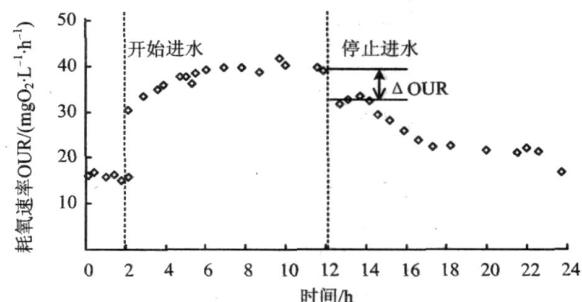


图 5 耗氧速率(OUR)随运行时间的变化

Fig. 5 Changes of the oxygen uptake rate with time

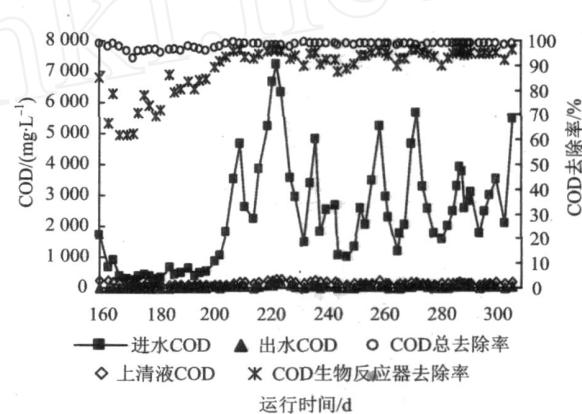


图 6 第阶段 SMBR 对 COD 的去除效果

Fig. 6 COD removal in the submerged membrane bioreactor at stage 2

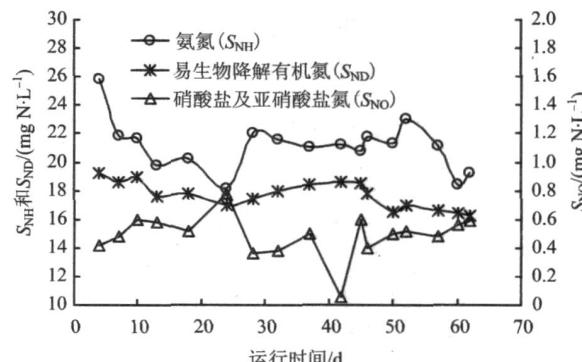


图 7 中药废水中 S_{NH} 、 S_{NO} 和 S_{ND} 的试验结果

Fig. 7 S_{NH} , S_{NO} and S_{ND} in traditional Chinese medicine wastewater

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On the evaluation of the sewage-contaminated COD and N in traditional Chinese medicine based on the activated sludge model No. 1

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Abstract: This present paper is focused on the study of the evaluation of the effluent chemical oxygen demand (COD) and nitrogen (N) based on the activated sludge model No. 1 (ASM1) through comparing the results with the modeling of traditional activated sludge processes. As is known, membrane bioreactors (MBRs) are attracting the whole global interest in water and sewage treatment because of its advantage of producing highly qualified effluent that can meet the commonly needed water quality regulations. However, due to the intrinsic complexity and instability of MBR processes, basic models are needed to provide a holistic understanding of the technology at a fun-

damental level and then compare the results of the treatment with the modeling of traditional activated sludge processes. The experimental results of our evaluation demonstrate that the influent COD and N in traditional Chinese medicine sewage proves quite different from that of domestic sewage. When compared with the experimental research and development, due to the commercialization of the technology, modeling studies for system design analysis and performance prediction are at a relatively rudimentary state. However, mathematical modeling of the biological performance of MBRs remains very limited in use, which has made us conduct a test in a membrane bioreactor (MBR) to treat high-strength traditional Chinese medicine (TCM) wastewater from two-phase anaerobic digest effluent. The experimental results we have gained demonstrate that the influent COD and N of traditional Chinese medicine wastewater were different to that of domestic sewage. The experimental results also demonstrate that readily biodegradable substrate (S_S) proves 141.2 mg/L, with its slowly biodegradable substrate (X_S) being 2113.2 mg/L, particulate (X_1) material being 85.3 mg/L, and soluble (S_1) material 53.8 mg/L. In addition, ammonia nitrogen (S_{NH}) in the medicine was 20.93 mg/L, nitrate nitrogen (S_{NO}) was 0.5 mg/L, soluble biodegradable organic nitrogen (S_{ND}) was 17.6 mg/L, particulate biodegradable organic nitrogen (X_{ND}) was 263.4 mg/L. COD and nitrogen (N) evaluation was input of ASM1 and its validity of evaluation was correlative to simulation results of ASM1. Thus, COD and N evaluation tends to be very important to the ASM1 model. Moreover, evaluation of COD and N may have the oriented and referential function for the similar sewage. And, therefore, it proves a real need to develop a model to predict and simulate the effluent COD quality and the process of membrane blocking.

Key words: water pollution prevention and treatment; membrane bioreactor; traditional Chinese medicine wastewater; activated sludge model No. 1 (ASM1); component evaluation

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