

# 烟煤掺烧污泥 HCl 的排放和脱除实验研究

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**摘 要:** 污泥与煤混烧发电是一种较有发展前景的污泥处理方法, 而混烧过程中产生的大量 HCl 气体会造成受热面腐蚀等危害。利用水平管式反应炉研究了升温速率、燃烧温度、污泥掺混比例以及反应气氛等因素对 HCl 释放的影响, 考察了 CaCO<sub>3</sub> 添加量和添加温度对 HCl 脱除效果, 并利用 GAS-MET DX4000 型红外气体分析仪实时测定 HCl 释放浓度。结果表明: 随着污泥掺烧比例增加, HCl 释放体积增加, 而 Cl<sup>-</sup>→HCl 转化率降低; 温度升高会促进 HCl 释放, 但 600 °C 后 Cl<sup>-</sup>→HCl 转化率变化不大; 载气中氧气比例升高使得部分 HCl 转化为 Cl<sub>2</sub>, 导致样品中 Cl<sup>-</sup>→HCl 转化率降低; 高升温速率导致 HCl 释放峰值变大; Ca/(S+0.5Cl) 摩尔比增大, 则 HCl 脱除率增加, 且 Ca/(S+0.5Cl) 摩尔比为 2 时, 700 °C 是 HCl 脱除最佳温度。

**关 键 词:** 污泥; 烟煤; HCl 释放; HCl 脱除

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## 引 言

根据预测 2015 年我国需处置干污泥量约 1.2 万 t/d<sup>[1]</sup>, 我国将面临巨大的污泥处理压力。而焚烧法处理污泥具有“剩余物少, 不需灭菌处理, 处理速度快, 不需长距离运输, 可回收能量用于发电和供热”的优点, 因此, 对污泥和煤混烧的研究正得到专家学者越来越多的关注。

但是, 污泥的 Cl 含量远高于煤<sup>[2]</sup>, 污泥和煤混烧时, 不仅会释放 SO<sub>x</sub>、NO<sub>x</sub> 等有害气体, HCl 气体也是主要的污染气体之一。而 HCl 的强腐蚀性会造成受热面的高温腐蚀损毁及尾部受热面和烟道的低温腐蚀, 还会促进毒性有机挥发物的生成, 因而, 对污泥和煤混烧时 HCl 释放及脱除行为的研究十分必要。

Brooke Shemwell 等人研究表明, 水蒸气会加速氯的析出反应和传递的速率, 促进医疗垃圾和生物质中氯的析出<sup>[3]</sup>; 李晓东和李诗媛等人研究表明, 五氯酚和有机废液焚烧时 Cl<sup>-</sup>→HCl 转化率随着温度

的升高而增加<sup>[4~5]</sup>; 李水清等人研究表明, 钙基吸收剂(Ca(OH)<sub>2</sub>、CaCO<sub>3</sub>、CA) 呈现出良好的 HCl 脱除性能<sup>[6]</sup>; Matsukata 的研究表明<sup>[7]</sup>, 利用石灰石脱除 HCl 的效率与 H<sub>2</sub>O 浓度密切相关, 而 SO<sub>2</sub> 对 HCl 的脱除效率影响不明显。

以往的研究大都通过测定吸收溶液中的 Cl<sup>-</sup> 离子来获得一段时间内的 Cl 释放总量, 而本研究将污泥和烟煤在水平管式反应炉中混烧, 并利用 GAS-MET DX4000 型红外气体分析仪对 HCl 释放情况进行实时检测, 研究内容包括: (1) 污泥和烟煤混烧时 HCl 的释放规律; (2) 钙基添加剂对 HCl 脱除行为的影响。

## 1 实验装置及方法

图 1 所示的实验系统主要由配气系统、燃烧系统、温控系统以及烟气分析系统组成, 反应器为水平放置的长 100 cm、内径 4.5 cm 的石英管, 炉温控制精度为 1 °C, 烟气分析利用 GASMET DX4000 型红外气体分析仪, 每 5 s 记录一次数据。实验时称取 0.5 g 的样品, 均布于瓷舟内。载气流量为 3 L/min, 程序升温速率为 20 K/min, 终温 800 °C。

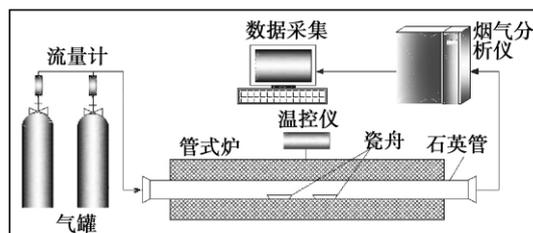


图 1 实验系统示意图

Fig. 1 Schematic drawing of the test system

实验所用烟煤为某电厂的设计煤种, 污泥样品为某污水处理厂生活污水。工业分析采用 SDTGA

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-2000 工业分析仪;元素分析采用 Elementar Vario EL CHNS 元素分析仪。样品中氯含量采用艾士卡混合剂熔样-硫氰酸钾滴定法测定,结果如表 1 所示。

表 1 煤和污泥的工业分析和元素分析结果  
Tab.1 Industrial and elementary analytic results of coal and sludge

	烟煤	污泥
工业分析/%		
M <sub>ar</sub>	6.60	20.00
A <sub>ad</sub>	22.28	50.98
V <sub>ad</sub>	29.69	40.24
FC <sub>ad</sub>	45.62	5.99
元素分析/%		
C	54.40	19.63
H	4.00	3.32
O	7.47	11.19
N	1.04	3.21
S	0.54	0.70
Cl/g · kg <sup>-1</sup>	0.5429	0.8907

注: O 含量由差减法计算得到。

定义 Cl→HCl 转化率为:

$$\eta_{Cl \rightarrow HCl} = \left( 1 - \frac{\text{释放的 HCl 摩尔数}}{\text{样品中 Cl 摩尔数}} \right) \times 100\%$$

定义 HCl 脱除率为:

$$\eta_{HCl} = \left( 1 - \frac{\text{加CaCO}_3 \text{后 HCl 释放浓度}}{\text{加CaCO}_3 \text{前 HCl 释放浓度}} \right) \times 100\%$$

## 2 实验结果及讨论

### 2.1 HCl 释放规律研究

#### 2.1.1 污泥掺混比例对 HCl 释放的影响

关于煤和污泥燃烧时 HCl 的生成机理,李寒旭等人研究表明<sup>[8]</sup>:在 200 °C 左右时,HCl 主要由吸附在样品裂隙中的氯离子解析生成;400 °C 左右时,HCl 的释放与煤的有机结构有关,涉及到 C-H 键和 C-Cl 键的断裂;700 °C 以上,HCl 的释放主要与样品中无机氯化物有关。

图 2 为空气气氛中不同污泥掺烧比例的样品升温至 800 °C 的 HCl 释放曲线。结果显示:HCl 的释放主要集中在 350 ~ 420 °C,即 HCl 的释放主要源自于有机物质的裂解。另外,随着污泥掺烧比例的增加,HCl 的释放浓度峰值更大(烟煤无明显峰值),

峰值出现的温度无明显差别。

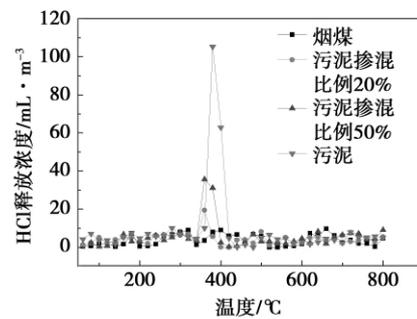


图 2 不同污泥掺烧比例下 HCl 释放随温度变化曲线

Fig. 2 Curves showing a change of HCl release with temperature in various proportions of sludge mixed in combustion

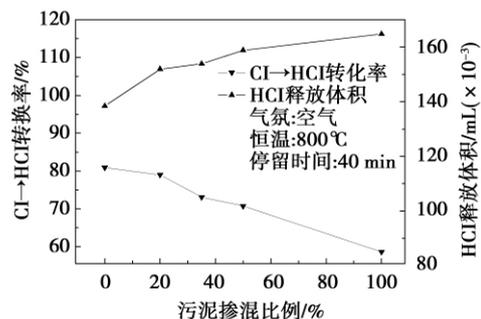


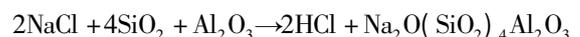
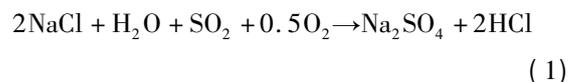
图 3 不同掺烧比例的 Cl→HCl 转化率和 HCl 释放体积

Fig. 3 Cl→HCl conversion rate and HCl release volume in various proportions of sludge mixed in combustion

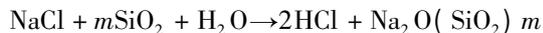
从图 3 看出:随着污泥掺烧比例的增加,样品 HCl 释放浓度增加,但 Cl→HCl 转化率逐渐降低。

污泥的 Cl 含量远远高于烟煤,所以随着污泥掺烧比例的增加,样品的 Cl 含量增加,这是导致 HCl 释放体积变大的主要原因,如表 2 所示。

另外,实验污泥全水远远高于烟煤,达到了 20%。随着掺烧比例的增加,带入燃烧过程的水蒸气增加,不仅促进了反应式(4)的逆反应,还促进了反应式(1)~式(3)向正反应方向进行<sup>[9-10]</sup>,导致 HCl 释放体积增加。



(2)



(3)

根据勒夏特列原理<sup>[11]</sup>,增加反应物的量,反应向正方向进行,但是反应物转化率下降。因此随着污泥掺烧比例的增加,样品中 Cl 含量增加,HCl 释放浓度也增加,但 Cl→HCl 转化率却降低。

### 2.1.2 温度对 HCl 释放的影响

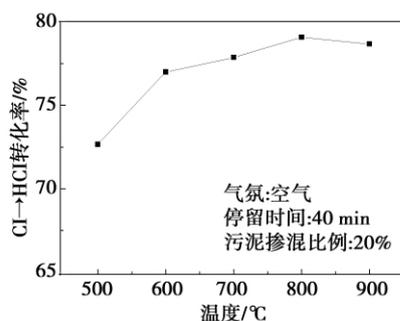


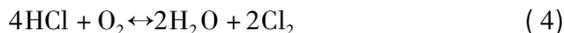
图4 不同温度下 Cl→HCl 转化率

Fig. 4 Cl→HCl conversion rates at different temperatures

图4显示了样品在不同温度下停留40 min的 Cl→HCl 转化率。结果表明:温度对 Cl→HCl 转化率的影响表现出阶段性的特征,随着恒温的升高,Cl→HCl 转化率先是迅速增长,然后缓慢增长,最后甚至下降。

2.1.1 节的研究结果表明,500 °C 以下为 HCl 释放主要温度区段,即污泥与煤中大部分的氯在低温下就以 HCl 形式析出,因此随着温度升高,转化率增加变缓。

此外,在高温下,反应式(4)会非常活跃的向正方向进行,生成的 HCl 中一部分会转化成 Cl<sub>2</sub>,导致 HCl 的释放量在高温下并没有显著增加。以往的研究中<sup>[12]</sup>,高温下,Cl→Cl<sub>2</sub>的转化率为 2%~5%,即:



### 2.1.3 不同氧气浓度对 HCl 释放的影响

从图5看出,以 20 K/min 的升温速率升温至 800 °C 时,污泥在空气气氛中 HCl 释放峰值比氮气气氛更大,但都在 400 °C 左右,李晓东等人的研究结果也证明了这个结论<sup>[4]</sup>。

图6的结果表明,随着载气中氧气比例的增加,样品中 Cl→HCl 转化率逐渐增大,但氧气比例超过 20% 后有所回落。

随着氧气通入比例的增加,管式炉内由惰性气氛转变为氧化性气氛,更有利于 HCl 的析出<sup>[4]</sup>,且

反应式(5)向有利于 HCl 生成的方向进行,因此 HCl 的释放增加。当氧气比例为 20% 以上时,管式炉中变为强氧化性气氛,反应式(4)会向正方向反应,因此 HCl 有一部分会转变为 Cl<sub>2</sub>,而且反应式(4)的影响大<sup>[13]</sup>,导致 Cl→HCl 转化率降低,则:

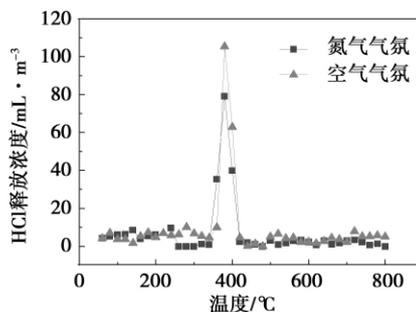
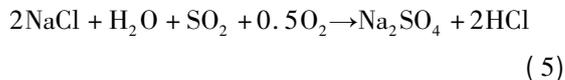


图5 污泥在不同气氛下 HCl 释放随温度变化曲线

Fig. 5 Curves showing a change of HCl release of sludge with temperature in various atmospheres

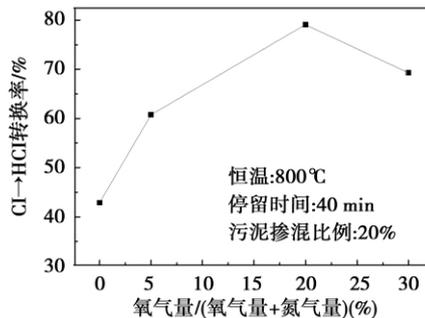


图6 800 °C 时不同氧浓度下 Cl→HCl 转化率

Fig. 6 Cl→HCl conversion rates at various oxygen concentrations at 800 °C

### 2.1.4 不同升温速率对 HCl 释放的影响

图7为空气气氛中,污泥从常温升温至 800 °C 时 HCl 的释放过程曲线。结果表明:升温速率越高, HCl 释放峰值越大,且越早出现,这与以往研究结论一致<sup>[14]</sup>。2.1.1 节的研究结果显示 HCl 的释放主要源自于有机物质的裂解,那么,升温速率越高,达到高温就越快,有机物质内产生断键也就越快,而 C-H 键和 C-Cl 键都是极易断裂的键<sup>[14]</sup>,从而,高升温速率下,较短的时间就出现了 HCl 释放的高峰。

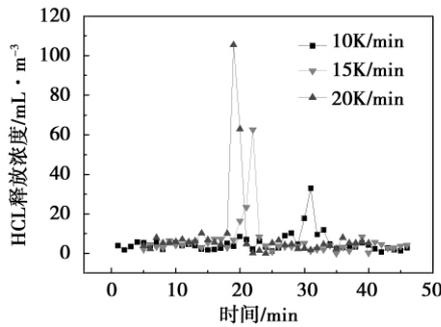


图7 污泥不同升温速率 HCl 释放随时间变化曲线

Fig. 7 Curves showing a change of HCl release of sludge with temperature at various temperature rise speeds

### 2.2 HCl 脱除规律研究

#### 2.2.1 不同 Ca/(S + 0.5Cl) 摩尔比对 HCl 脱除的影响

将 CaCO<sub>3</sub> 与污泥掺烧比例为 20% 的混合燃料均匀混合后在水平管式炉燃烧,研究了 CaCO<sub>3</sub> 添加量和温度对 HCl 脱除的影响。CaCO<sub>3</sub> 脱除 HCl 的反应机理为:

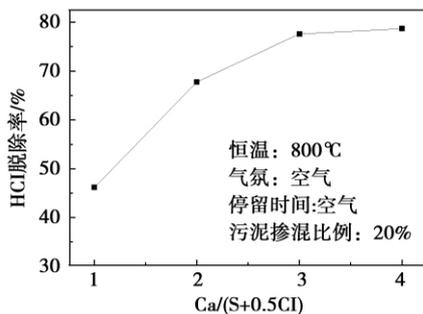
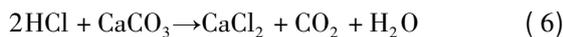


图8 800°C 时不同 Ca/(S + 0.5Cl) 摩尔比 HCl 脱除率

Fig. 8 HCl removal rates at various Ca/(S + 0.5Cl) molar ratios at 800°C

从图8可以看出:随着 Ca/(S + 0.5Cl) 摩尔比的增大,HCl 脱除率增加,但增长逐渐减缓。这是因为反应式(6)在开始阶段受化学反应控制,CaCO<sub>3</sub> 的增加有利于其进行。但随着反应的进行,吸收剂颗粒的表面逐渐形成产物层,且孔隙易被生成的 CaCl<sub>2</sub> 填满,整个反应的速率主要与 HCl 气体在产物层中的扩散速率有关,所以加大吸收剂,HCl 的去除率增长变缓<sup>[15]</sup>。另外,Ca/(S + 0.5Cl) 摩尔比为

2 时,HCl 脱除率已达 68%,剩余 HCl 浓度有限,因此增加吸收剂,反应式(6)只会少许进行,也导致 HCl 脱除率增长变缓。

#### 2.2.2 温度对 HCl 脱除的影响

图9为 Ca/(S + 0.5Cl) 摩尔比为 2 时,不同温度下 HCl 脱除率对比情况。结果表明:随着燃烧温度升高,HCl 脱除率大幅度增大,700 °C 时达到最大。同样,Daoudi 等人研究发现<sup>[16]</sup>:CaCO<sub>3</sub> 去除 HCl 的最佳温度是 600 ~ 850 °C。这是因为随着温度升高,反应式(6)中 CaCl<sub>2</sub> 的生成速率变快,CaCl<sub>2</sub> 阻塞气孔现象也会好转<sup>[17]</sup>,所以 HCl 的脱除率越来越高。

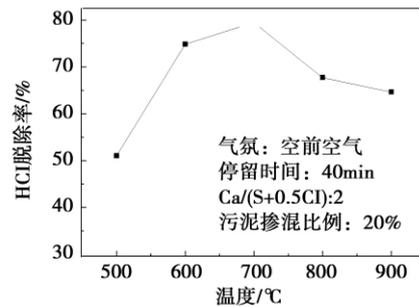


图9 不同温度下 HCl 脱除率

Fig. 9 HCl removal rates at various temperatures

但是,700°C 后温度继续升高,大量水蒸气的存在,污泥灰和煤灰中较大含量的 SiO<sub>2</sub> 与生成的 CaCl<sub>2</sub> 发生反应式(7),导致 HCl 脱除率下降:



D. Lawrence 研究也发现<sup>[18]</sup>:反应式(7)中,水分含量越高,CaCl<sub>2</sub> 的质量损失越大,且温度越高,上述反应越彻底。

### 3 结论

(1) 污泥掺烧比例增加,HCl 释放体积增加,而 Cl → HCl 转化率逐渐降低。

(2) 升高温度促进了 HCl 释放,但 600 °C 后 Cl → HCl 转化率变化不大。

(3) 载气中氧气比例升高促进 HCl 释放,但氧气比例达到 20% 后,由于部分 HCl 转化为 Cl<sub>2</sub>,Cl → HCl 转化率降低。

(4) 高升温速率下,HCl 释放峰值更大,且更早出现。

(5) 随着 Ca/(S + 0.5Cl) 摩尔比的增大,HCl 脱除率增加,但增长逐渐减缓,且 Ca/(S + 0.5Cl) 摩

尔比为 2 时, 700 °C 是 HCl 脱除最佳温度。

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face produce more crystal nuclei and the crystal distribution is irregular ,resulting in a cross and superimposed growth. **Key words:** CaCO<sub>3</sub> crystallization fowl coupon test ,crystal morphology ,crystal size distribution

基于流固耦合的化容补水泵性能分析 = **Analysis of the Performance of a Chemical Vessel Make-up Water Pump Based on a Fluid-solid Coupling** [刊 ,汉] ZHU Li-kai ,HU Jing-ning ,ZHANG Jun-hui ,LI Hao ( Engineering Technology Research Center for Fluid Machinery ,Jiangsu University ,Zhenjiang ,China ,Post Code: 212013) // Journal of Engineering for Thermal Energy & Power. - 2012 27( 4) . - 483 ~ 488

Three groups of models were designed for chemical vessel supplementary water pumps and three-dimensional models were established by using the software Pro/E. CFD software was used to conduct a simulation and a test was performed of the hydraulic models to acquire an optimum model by comparing the test results with the simulation ones. The link of the CFD with the Static Structural and Modal was established by using the Ansys Workbench to conduct an analysis of the optimum model. On the basis of the three-dimensional steady numerical calculation results of the pump and by making use of the sequence coupling technology ,an iterative calculation was performed of the solid and fluid domain to analyze the static stress and vibration mode of the impeller. It has been found that under the action of the hydraulic pressure ,the maximal displacement due to the blade deformation occurs at a place nearing the trailing edge of the blade. Due to an action of the balance holes ,the equivalent stress of the blade is relatively uniform and small. Under the action of the pressure difference before and after the wheel ,the hub has a relatively big equivalent stress. Under the design operating condition ,the deformation of the hub has a conspicuous influence on the vibration mode. **Key words:** chemical vessel supplementary ( CVS) water pump ,static stress ,mode ,fluid-solid coupling

烟煤掺烧污泥 HCl 的排放和脱除实验研究 = **Experimental Study of HCl emissions and Removal During Combustion of Coal Mixed and Diluted with Sewage Sludge** [刊 ,汉] CUI Hao ,ZHANG Cheng ,XIA Ji ,CHEN Gang( National Key Laboratory on Coal Combustion ,Central China University of Science and Technology ,Wuhan ,China ,Post Code: 430074) //Journal of Engineering for Thermal Energy & Power. - 2012 27( 4) . - 489 ~ 493

Power generation by burning coal mixed and diluted with sewage sludge is a sludge treatment method with a relatively good prospect. However ,a great amount of HCl gas produced during the combustion may result in such harms to the heating surfaces as corrosion and erosion. By making use of a horizontal tube type reactor ,the authors studied the influence of such factors as temperature rise speed ,combustion temperature ,proportion of sludge mixed and diluted

and reaction atmosphere etc. on the HCl release. Furthermore, the influence of the  $\text{CaCO}_3$  adding amount and temperature on the HCl removal efficiency was investigated and by using a GASMET DX4000 model infrared gas analyzer, the concentration of the HCl released was real-time measured. It has been found that with an increase of the proportion of the sludge mixed and diluted, the volume of the HCl released will increase while the conversion rate of Cl to HCl will decrease. To increase the temperature will promote the HCl release, however, after  $600\text{ }^\circ\text{C}$ , the conversion rate of Cl to HCl will change little. The proportion of oxygen in the carrier gas will make a part of HCl converted into  $\text{Cl}_2$ , resulting in a drop of the conversion rate of Cl to HCl in the sample. A high temperature rise speed will lead to a large peak value of HCl release. To increase the  $\text{Ca}/(\text{S} + 0.5\text{Cl})$  molar ratio will increase the HCl removal rate. When the  $\text{Ca}/(\text{S} + 0.5\text{Cl})$  molar ratio is 2,  $700\text{ }^\circ\text{C}$  will be regarded as the optimum temperature for HCl removal. **Key words:** sewage sludge, bituminous coal, HCl emissions, HCl removal

造纸污泥与污水污泥的表观干燥动力学研究 = **Study of the Apparent Drying Kinetics of Papermaking and Sewage Water Sludge** [刊, 汉] LOU Bo, QIAN Wei, WU De-zhi (Guangdong Provincial Key Laboratory on Green Energy Source Technologies, College of Electric Power, South China University of Science and Technology, Guangzhou, China, Post Code: 510640) // Journal of Engineering for Thermal Energy & Power. - 2012, 27(4). - 494 ~ 497

To study the apparent drying kinetics of sludge is of major significance for guiding sludge treatment. For this purpose, a contrast test of paper-making sludge and sewage water sludge with a same specific surface area of  $3.24\text{ cm}^2/\text{g}$  at four temperatures ( $30\text{ }^\circ\text{C}$ ,  $70\text{ }^\circ\text{C}$ ,  $102\text{ }^\circ\text{C}$  and  $130\text{ }^\circ\text{C}$ ) was performed and a fitting of the kinetic equation was conducted. It has been found that the drying rate constant  $k$  is an important parameter for studying the drying kinetics of sludge and the drying rate constant of the paper-making sludge is bigger than that of sewage water sludge. The activated energies of the paper-making and sewage water sludge are  $26.84\text{ kJ/mol}$  and  $27.423\text{ kJ/mol}$  respectively, indicating that the activated energy of the sewage water sludge is slightly bigger than that of the paper-making sludge. This depends on the fact that the viscosity of the sewage water sludge is comparatively big. **Key words:** paper-making sludge, sewage water sludge, drying kinetics, activated energy, drying rate

柴油机燃用不同比例菜籽油生物柴油和乙醇柴油的对比试验研究 = **Contrast Experimental Study of a Diesel Engine Burning Rapeseed Oil-based Bio-diesel and Ethanol-diesel in Various Proportions** [刊, 汉] LI Hui-fen, HUANG Jin-cheng, HUANG Hao-zhong (College of Mechanical Engineering, Guangxi University, Nanning, China, Post Code: 530004), CHEN Guo-dong (Xingjian College of Science and Liberal Arts, Guangxi Univer-