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亚硫酸钠循环法烟气脱硫工艺实验研究

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摘　要: 以处理量为 1 800 m^3/h 的亚硫酸钠循环法烟气脱硫装置为实验对象, 着重研究吸收液 pH 值、液气比 L/G、吸收液成份和脱硫剂初始浓度等因素对脱硫效率的影响, 并对脱硫剂再生过程和再生清液的脱硫特性进行研究。结果表明: 吸收液 pH 值决定硫成份在溶液中的状态, 从而影响脱硫效率, 当 pH＞6 时, 脱硫效率高且随 pH 增大变化平缓; 当 pH＜6 时, 脱硫效率随 pH 减少急剧降低; 该工艺可以在较低的液气比(L/G＝0.25～1.25 L/m^3)下保持较高的脱硫效率(η＞90%); 同等条件下, 与 NaOH 和 Na_2CO_3 相比较, Na_2SO_3 的脱硫能力稍低; 吸收液 Na_2SO_3 浓度在 5%～10%范围内脱硫效率大于 90%; 脱硫剂再生反应过程能很快完成, 再生脱硫剂脱硫效率稳定, 与新鲜脱硫剂相比脱硫效率略低。

关　键　词: 烟气脱硫; 亚硫酸钠; 循环
中图分类号: X701.3　　文献标识码: A

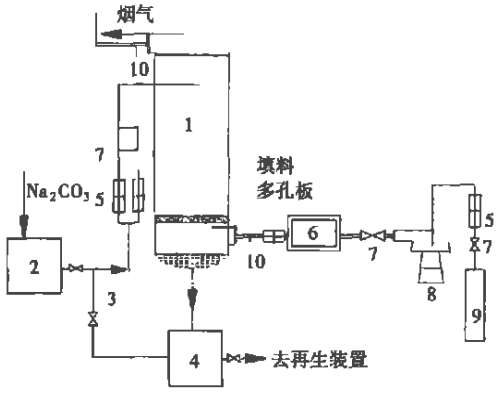
1　引　言

我国二氧化硫污染严重, 每年给国家造成巨大损失, 严重威胁人们的健康生活[1]。国内绝大部分燃煤火力发电厂未安装脱硫设备, 国家“十五”期间加大了二氧化硫污染治理力度, 强制要求新建燃煤电厂上配套脱硫装置, 同时要求对老电厂进行脱硫改造。与此同时, 国家对火力发电厂二氧化硫排放标准进行了修改, 2003 年新出台的电厂锅炉二氧化硫排放限值也更加严格。此外, 现阶段对燃煤工业炉窑的脱硫还没有全面展开, 其二氧化硫总排放量与电厂相当。所以, 目前我国烟气脱硫任务繁重, 同时也意味着烟气脱硫技术市场需求旺盛[2]。国内现有燃煤电厂的烟气脱硫技术主要依赖进口, 湿法脱硫技术是目前主流的烟气脱硫方法, 现阶段石灰石－石膏法占垄断性地位, 其特点是脱硫效率高, 系统投资和运行成本高。由于采用钙基脱硫容易结垢, 严重时会堵塞设备, 导致脱硫设备不能正常运行[3～7]。本文研究的亚硫酸钠循环法烟气脱硫技术, 吸收剂为可再生易溶性碱性溶液, 可以避免设备的结垢和堵塞, 运行时气液比较低, 可望达到降低脱硫成本的目的。

2　实验装置

实验室脱硫系统如图 1 所示, 钢瓶出来的 SO_2 与空气混合形成的模拟烟气, 经电加热器预热到预定温度后进入脱硫塔与吸收液反应, 部分脱硫吸收液去再生池再生。

脱硫塔结构基本参数: 烟气进、出口直径均为 200 mm, 塔内径 D＝400 mm, 塔高 H＝2 750 mm, 多孔气相导流板开孔直径为 d＝10 mm, 开孔率为 40%, 在孔板上覆有 50 mm 厚的陶瓷拉西环散堆填料, 孔板位于距离烟气进口中心线 150 mm 上方, 风机最大风量为 1 800 m^3/h。



1－吸收塔; 2－溶解池; 3－循环泵; 4－储液池; 5－流量计; 6－电加热器; 7－阀门; 8－风机　9－SO_2 钢瓶; 10－测试点

图 1　脱硫系统实验装置

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3　实验结果及讨论

3.1　pH 值和 L/G 的影响

湿法烟气脱硫过程中, 在吸收塔内 SO_2 从气相传递到液相, 进行的脱硫反应是相间传质过程。脱硫反应过程中硫成份在溶液中分别以 SO_3^{2-}、HSO_3^- 和 SO_4^{2-} 等阴离子形式存在, 其中 SO_4^{2-} 由脱硫过程中氧化反应产生, 其占总的硫成份比例非常小。吸收液中 SO_3^{2-} 和 HSO_3^- 浓度与溶液中的 pH 值关系密切, 图 2 为这两种离子浓度与溶液 pH 的关系, 由图可知, 当 pH 逐渐减小时, 溶液中 SO_3^{2-} 不断转化为 HSO_3^-, 吸收液的有效吸收 SO_2 成份不断减少, 导致系统脱硫效率降低。因此, 为保持一定的脱硫效率, 必须不断补充新鲜碱液来调整溶液 pH 值, 使溶液中有效脱硫成份 SO_3^{2-} 浓度保持在一定范围。脱硫吸收过程吸收液 pH 值对脱硫效率的影响如图 3 所示, 在 pH>6 的情况下, 脱硫效率基本达到最大, 且随溶液 pH 增大变化平缓; 而当 pH<6 时, 系统脱硫效率随 pH 降低急剧下降, 这是因为这时溶液中 HSO_3^- 浓度急剧增大, 导致脱硫能力降低。因此, 为保证一定的脱硫效率, 吸收液 pH 值宜控制在 6 以上。

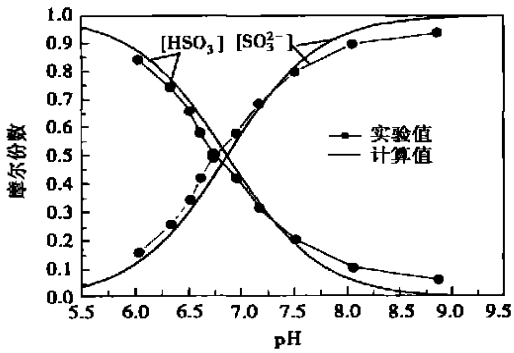

图 2　吸收液中 pH 与硫成份的关系

液气比 L/G 的大小直接关系到两相吸收反应效果, 在采用石灰石—石膏湿法脱硫时, 为保证一定的脱硫效果, 其运行液气比 L/G 通常在 20~30 L/m³ 之间。本文采用的易溶性碱液吸收效果良好, 在液气比 L/G=0.25~1.25 L/m³ 范围内, 脱硫效率均大于 90%, 说明该法在降低循环泵动力消耗方面存在较大优势, 随 L/G 的增大, 气液两相接触条件改善, 脱硫效率逐渐增大, 如图 4 所示。

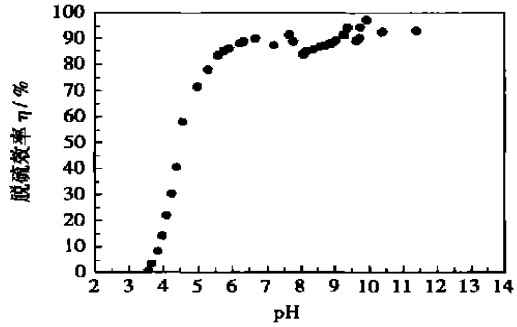

图 3　吸收液 pH 值对脱硫效率的影响

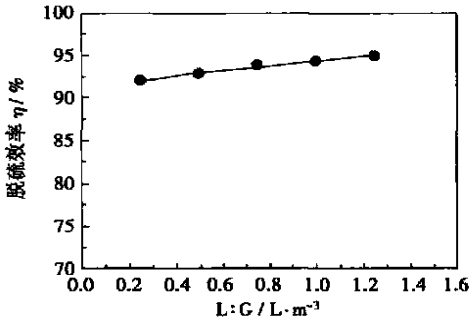

图 4　L/G 对脱硫效率的影响

3.2　脱硫剂的影响

脱硫剂 Na_2SO_3 初始浓度选择也非常重要, 本文分别以质量浓度 1%、5%和 10% 的 Na_2SO_3 初始吸收液为对象进行脱硫对比实验。结果表明, Na_2SO_3 初始浓度在 5%~10%间能有较好的脱硫效果, 过稀的初始吸收液脱硫效率降低较快, 实际应用时不宜采用, 如图 5 所示。

亚硫酸钠循环法脱硫系统吸收液中存在 3 种有效脱硫成份分别为 NaOH、Na_2CO_3 和 Na_2SO_3, 为比较三者的脱硫效果, 本实验分别单独采用它们作为初始吸收液, 其质量浓度均为 10%, 在液气比 L/G=0.54 L/m³ 时, 测得它们脱硫效率如图 6 所示。同等条件下, 与 NaOH 和 Na_2CO_3 相比, 脱硫剂采用 Na_2SO_3 时, 脱硫效率有所降低, 但系统脱硫效率也能在90%以上。其原因是, 以 NaOH 和 Na_2CO_3 为初始液, 在吸收一定 SO_2 后, 吸收液中间产物都是 Na_2SO_3, 其脱硫效果比 Na_2SO_3 好。文献[4] 用 Na^+ 浓度来表示脱硫液的有效吸收成份, 由于吸收液中 Na^+ 可以和多种阴离子结合, 也就是说, 当 Na^+ 浓度

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不变时溶液的吸收能力变化很大，因此用 Na^+ 浓度并不能真实反映吸收液的脱硫能力，真正决定吸收液脱硫能力的是吸收液中阴离子的形态。

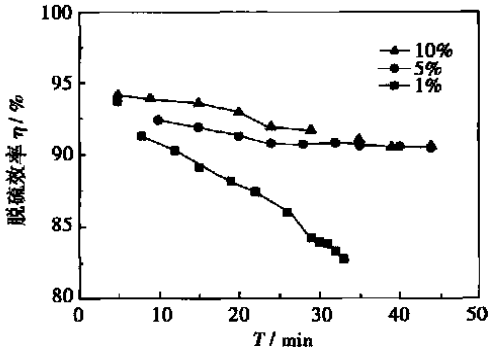

图 5 Na_2SO_3 浓度对脱硫效率的影响

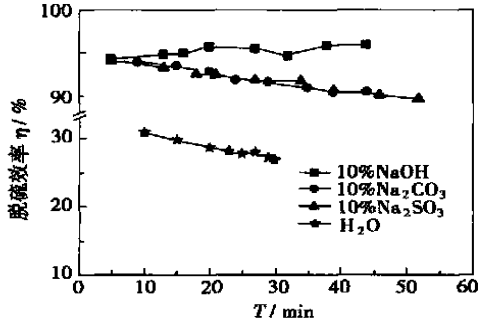

图 6 不同脱硫剂对脱硫效率的影响

3.3 脱硫剂再生

本实验采用 $\rho = 1.25$ g/mL 的 $Ca(OH)_2$ 悬浮液再生 Na_2SO_3，在不同 $Ca(OH)_2$:$NaHSO_3$ 摩尔比的情况下，连续搅拌，中和反应过程再生液 pH 值随时间变化如图 7 所示。由实验结果知，各种工况下，中和再生反应较快，随 $Ca(OH)_2$ 量的增加，反应完成更快。当 $Ca(OH)_2$:$NaHSO_3$ 摩尔比为 0.8 或更多时，再生液 pH 值达到 12 以上，此时再生吸收液成份中 NaOH 比例不断增加。在保证脱硫效率的前提下，从控制再生过程 $Ca(OH)_2$ 用量和防止吸收液输送管道设备防碱腐蚀角度考虑，再生时控制 $Ca(OH)_2$: $NaHSO_3$ 摩尔比在 0.5 ~ 0.8 之间比较合适。

为检验再生脱硫清液的脱硫效果，本文进一步对再生的脱硫剂进行脱硫实验，图 8 为 $Ca(OH)_2$: $NaHSO_3 = 0.6$ 时再生脱硫液的脱硫效果实验结果。由图可知，再生液脱硫效率稳定在 90% 左右，与新鲜脱硫液比较(见图 5 和图 6)，脱硫效率略低，其原因与脱硫过程中脱硫剂的氧化消耗等因素有关，通过补充少量的新鲜脱硫液，脱硫效率可以达到新鲜吸收液的水平。

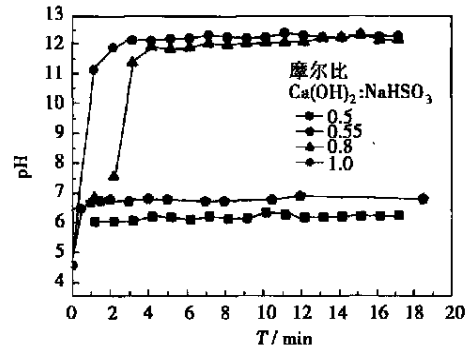

图 7 再生过程再生液 pH 随时间变化

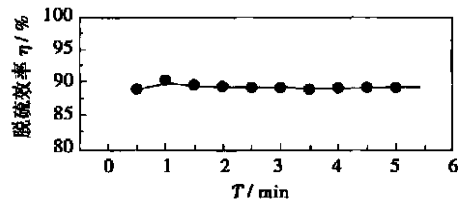

图 8 再生液脱硫效果

4 结 论

(1) 吸收液 pH 值与溶液中有效硫成份阴离子关系密切，为保证较高的脱硫效率(大于 90%)，吸收液 pH 值必须大于 6。

(2) 本工艺在较低液气比 L/G (0.25 ~ 1.25 L/m^3)下运行，脱硫效率大于 90%；与 NaOH 和 Na_2CO_3 相比较，以 Na_2SO_3 为脱硫吸收液时，脱硫效率稍有降低；脱硫剂 Na_2SO_3 溶液合适浓度范围为 5% ~ 10%。

(3) 采用 $Ca(OH)_2$ 悬浮液的再生过程反应能很快完成，再生过程中 $Ca(OH)_2$:$NaHSO_3$ 摩尔比宜控制在 0.5 ~ 0.8 之间，再生吸收液脱硫效果稳定。

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能量位垒, 即前面求取的活化能 33 650 J/mol。由此可知, 由于温度的升高, 在高温的条件下该反应不是很难进行, 常温下发生反应的几率就很小。因为在常温下能碰撞到铁表面的氧分子中, 只有很小一部分能量特别高的氧分子才能走完这个反应历程[5]。

在455 ℃下存在的主要氧化物成份 Fe_3O_4 和 Fe_2O_3[6], 由于特定试验条件与其它因素的影响, 很难确定两种氧化物的确切含量。故设反应后的氧化物为 Fe_xO_y, 该式仅代表生成氧化物的铁氧比例的形式, 不代表某种存在的氧化物成份, 忽略其它杂质成份生成的氧化物。又根据 455 ℃下氧化前后铁元素和氧元素的原子百分含量, 计算出氧化物的形式为 $Fe_{1.04}O$, 从而根据实验的氧化增重量计算出的氧化膜中被氧化的铁的量。从氧化动力学曲线可知在 102 h 的时候氧化曲线基本趋于平坦, 计算出氧化过程中的平均氧化速率为 22.06 mg/(dm^2·d)。在 102 h 后随着时间的增加氧化增重并不明显, 但在实际锅炉的运行中存在冲刷磨损以及多种腐蚀性气体的腐蚀等多方面因素的影响, 氧化仍可能继续恶化, 进而对锅炉造成严重的影响。

4　结　论

(1) 锅炉水冷壁主要材料 20 g 的试片在 380 ~ 480 ℃温度下氧化动力学曲线成双曲线(单支)函数分布, 并且随着温度的升高氧化速率加快, 符合金属腐蚀规律。利用热分析动力学的方法求得 20 g 在炉膛温度为 810 ~ 920 ℃(壁温 380 ~ 480 ℃)范围内的反应活化能 E = 33 650 J/mol, 指前因子 A = 3.971 s^{-1}。

(2) 对试片进行 SEM 扫描观察, 并用 EDS 能量色谱分析元素成份, 结合氧化动力学曲线得出了在该试验条件下氧化过程中的平均氧化速率为 22.06 mg/(dm^2·d)。

(3)本试验以空气作为试验介质, 在实际锅炉运行时, 炉膛内的氧气要比空气中含氧量低。但是考虑到煤粉燃烧的过程中, 炉膛内的氧含量分布也是不均匀的, 某些区域水冷壁的氧化速率可以参考试验中得出的结果, 在进一步的试验中将考虑到不同气氛的问题。

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循环流化床烟气脱硫装置对电除尘器影响研究=**A Study of the Influence of the Flue Gas Desulfurizer of a Circulating Fluidized Bed on an Electrostatic Precipitator**[刊, 汉] /ZHAO Xu-dong, XIANG Guang-ming (Thermal Energy Engineering Department, Tsinghua University, Beijing, China, Post Code: 100084), WU Shao-hua (Harbin Institute of Technology, Harbin, China, Post Code: 150001) //Journal of Engineering for Thermal Energy & Power. —2005, 20(4). —377 ~380

The flue gas suspension desulfurization system of a double circulating fluidized bed was designed and constructed for a 75 t/h coal-fired boiler. Prior to and after the operation of the desulfurization system a comparison analysis of the related characteristics at the inlet of an electrostatic precipitator was conducted. Such characteristics include flue gas temperature, humidity, chemical composition, dust particle diameter, outward appearance and specific resistance, etc. It has been found that after the operation of the desulfurization system the humidity of the flue gas at the inlet of the electrostatic precipitator has increased while its temperature and dust particle specific resistivity decreased, thus enhancing the system desulfurization efficiency and the dust removal efficiency of the electrostatic precipitator. Also investigated was the impact of the flue gas desulfurizer of the circulating fluidized bed on the operation stability and dust removal efficiency of the electrostatic precipitator. Meanwhile, several countermeasures for operation improvement are also proposed. **Key words**: flue gas desulfurization of a circulating fluidized bed, electrostatic precipitator, dry method of desulfurization

镁离子法测定除雾器出口烟气携带液滴量方法=**The Use of Magnesium Ion Method for the Determination of Droplet Quantity Entrained by Flue Gas at the Outlet of a Demistor**[刊, 汉] /LI Sen, ZHAO Qu-lan, XU Tong-mo, et al (Energy & Power Engineering Institute under the Xi'an Jiaotong University, Xi'an, China, Post Code: 710049) //Journal of Engineering for Thermal Energy & Power. —2005, 20(4). —381 ~383

Based on the technological features of the desulfurization by wet limestone-gypsum method the authors have revealed through a study that Mg2+ concentration in slurry is equal to the concentration of Mg2+ in the droplet entrained by the flue gas at a demister outlet. On the basis of this conclusion a constant speed sampling was performed at the demister outlet with 5m3 of flue gas being extracted. On the basis of a determination of the Mg 2+ concentration in a micro-fiber filter wash liquid in the sampling system, in a sampling-tube wash solution and in a blank micro-fiber filter wash liquid, as well as the slurry density in an absorption tower and the concentration of Mg^{2+} in an absorption-slurry filtering liquid, calculated was the liquid droplet quantity entrained by the flue gas at the demister outlet. **Key words**: magnesium ion, demister, liquid droplet, constant speed sampling

亚硫酸钠循环法烟气脱硫工艺实验研究=**Experimental Study of Wet Flue Gas Desulfurization by Using a Sodium Sulfite Circulation Method**[刊, 汉] /JIANG Li-qiao (Thermal Science and Energy Engineering Department, China National University of Science & Technology, Hefei, China, Post Code: 230026), ZHAO Dai-qing, CHEN En-jian (Guangzhou Institute of Energy Conversion under the Chinese Academy of Sciences, Guangzhou, China, Post Code: 510070) //Journal of Engineering for Thermal Energy & Power. —2005, 20(4). —384 ~386, 401

With the wet flue gas desulfurizer based on a sodium sulfite circulation method (processing capacity 1800 m^3/h) serving as an object of experimentation a detailed study was conducted of the impact of several factors on the desulfurization efficiency. Such factors include: pH value of absorption liquid, liquid/gas ratio, the initial composition of the absorption liquid and the initial concentration of a desulfurizing agent, etc. Furthermore, the regeneration process of the desulfurizing agent and the desulfurizing characteristics of the regenerated clean liquid were also studied. The results of the study indicate that the pH value of the absorption liquid can decide the state of sulfur element in a solution, thereby influencing the desulfurization efficiency. When pH is greater than 6, the desulfurization efficiency will be high and only change mildly with an increase in pH value. When pH is less than 6, the desulfurization efficiency will drastically decrease with a decrease in pH value. Under the present technological process it is possible to maintain a relatively high desulfurization efficiency (ηgreater than 90%) at a relatively low liquid-gas ratio (L/G = 0.25 ~1.25 L/m^3). Under equivalent conditions, as compared with NaOH and Na2CO3, the desulfurization capacity of Na2SO3 is slightly lower. When the concentration of the absorption liquid Na_2CO_3 is within the range of 5% - 10% the desulfurization efficiency will be greater than 90%. The regeneration reaction process of the desulfurizer can be completed very rapidly. The desulfurization effi-

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ciency of the regenerated desulfurizing agent is stable. Compared with a fresh desulfurizung agent the desulfurization efficiency is slightly lower. **Key words**: flue gas desulfurization, sodium sulfite, cycle

基于 ROLS 算法的 RBF 神经网络燃料电池电特性建模＝**Electric-characteristic Modeling of a Fuel Cell Based on ROLS Algorithm and RBF (Radial Based Function) Neural Network Identification Technique**[刊, 汉] /MIAO Qing, CAO Guang-yi, ZHU Xin-jian (Fuel Cell Research Institute under the Shanghai Jiaotong University, Shanghai, China, Post Code: 200030) //Journal of Engineering for Thermal Energy & Power. —2005, 20(4). —387 ~389

An innovative method is presented for the electric-characteristic modeling of a direct methanol fuel cell (DMFC) through the use of ROLS algorithm-based RBF (radial based function) neural network identification technique. With the operating temperature of the cell serving as an input and the voltage/electric current density of the cell serving as an output 1200 groups of experimental data were utilized as training and test samples to set up under various operating temperatures a dynamic response model of the cell voltage/electric current density. Simulation results indicate that the modeling method by using the RBF neural network identification technique is effective with the established model featuring a relative high precision. **Key words**: direct methanol fuel cell, radial based function, neural network identification, ROLS algorithm

基于神经网络的电除尘器效率在线确定方法＝**A Method for the On-line Determination of the Efficiency of a Neural Network-based Electrostatic Precipitator**[刊, 汉] /LI Da-zhong, TIAN li (North China Electric Power University, Baoding, China, Post Code: 071003), ZHANG Zheng-wei, et al (Datang Xuchang Longgang Power Generation Co. Ltd., Yuzhou, Henan Province, China, Post Code: 452500) //Journal of Engineering for Thermal Energy & Power. —2005, 20(4). —390 ~393

There exist numerous factors, which can affect the efficiency of an electrostatic precipitator. This also makes it difficult to conduct an on-line determination of the precipitator efficiency. In view of the above the authors have proposed a new method for setting up a model of electrostatic precipitator efficiency with the help of a neural network. In this kind of neural network model it is only necessary to input such operating parameters as boiler steam output, flue gas flow to be processed, ash, dust particle diameter and dust specific resistance, etc and one can readily realize the on-line determination of the electrostatic precipitator efficiency. The results of a simulation indicate that this neural network-based model has a fair effectiveness approximating to an actual system, thus providing a useful reference for the further modeling and optimized control of an electrostatic precipitator system. **Key words**: neural network, electrostatic precipitator, dust collection efficiency

辐射通道一维稳态温度场数值模拟＝**Numerical Simulation of the One-dimensional Steady-state Temperature Field of a Radiation Channel**[刊, 汉] /HAN Jia-de, LU Yi-ping (Institute of Mechanical & Power Engineering under the Harbin University of Science & Technology, Harbin, China, Post Code: 150080), ZHANG Pei-ting (Heilongjiang Research Institute of Special Equipment Inspection, Harbin, China, Post Code: 150040) //Journal of Engineering for Thermal Energy & Power. —2005, 20(4). —394 ~396

To determine the temperature distribution in a radiation-convection channel, a mathematical model was set up on the basis of energy conservation theory to reflect the heat transfer of air in a tube to a circular tube wall under radiation and convection nonlinear boundary conditions. A finite difference method is presented for solving the one-dimensional steady-state heat exchange of tube wall temperature and in-tube cooling air temperature. For the radiation heat exchange calculation adopted was a radiation heat transfer factor-based Monte Carlo method. The impact of relevant parameters on the temperature distribution in the radiation channel is analyzed. The parameters being studied include radiator surface temperature, the ratio of piping length to radius, the flow speed of cooling air in the tube, etc. The results of the calculation indicate that the surface temperature of the radiator represents a major factor influencing the maximum temperature in the radiation channel. The method under discussion can provide temperature field data for the detailed thermodynamic characteristics calculation of a radiation channel. **Key words**: radiation heat transfer, finite difference method, numerical simulation, temperature field

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