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格平

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绿色行动

——辽宁环境科研教育「123工程」论文集(2)



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“123 工程”论文集
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曲格平先生简介

曲格平先生 1930 年 6 月生于山东肥城，是中国环境保护事业的开创者和奠基人之一，著名环境学家，现任中华环境保护基金会名誉理事长。

作为中国政府代表团成员，他出席了 1972 年在斯德哥尔摩召开的联合国人类环境会议，以及 1992 年在里约热内卢召开的联合国环境与发展大会。曾任中国常驻联合国环境署首任代表，国家环境保护局首任局长，全国人大环境与资源保护委员会第八届、第九届主任委员等职。他在认真总结国内外环保经验的基础上，系统地提出了适合中国国情的、具有独创性的环境保理论和政策思路。

40 多年来，他为开拓和发展中国的环境保护事业，夙兴夜寐、殚精竭虑，做出了不可磨灭的贡献。他曾荣获联合国环境大奖、“金方舟”大奖、“蓝色星球奖”、“爱丁堡公爵保护奖”等国际奖项。同时他还是联合国可持续发展高级咨询委员会委员、全球环境基金高级顾问，北京大学、清华大学、牛津大学、哈佛大学等多所国内外知名院校的兼职和客座教授。

（摄影：中华环境保护基金会宣传联络部 李力）

格平绿色行动
——辽宁环境科研教育“123工程”论文集(2)

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序 言

“格平绿色行动”是由中华环境保护基金会名誉理事长曲格平先生出资倡导、创建的可持续资助及扶助型公益慈善行动，具有扶贫、生态改善、助学等特点，其内容包括捐资助学活动，“格平绿色行动”是可持续发展理论在环境保护公益事业中的一次具体实践。

周生贤部长在第二次全国环保科技大会上指出：加强科技创新，推动环保事业发展，是时代赋予环境科技工作者的神圣使命。科学技术是第一生产力，科技人才是社会发展的动力和源泉。国家实施振兴东北老工业基地和沿海经济带战略，要求我们必须培养和造就一批专业的环境保护科技人才来处理好经济与环境、社会与环境、资源与环境之间的协调关系。

中华环境保护基金会辽宁代表处开展的“格平绿色行动——辽宁环境科研教育‘123工程’”，旨在促进辽宁环境保护和教育事业的发展、大力培养环保科技人才，是实现环境科技人才培养与环境管理需要的有机结合。“123工程”起到了连接环境、科研和教育的桥梁作用。

“123工程”结合人才培养的实际，对在辽宁省境内就学的本科生、硕士研究生和博士研究生进行选择性的资助，充分发挥了环保公益资助金的社会效益，体现了环保部的科技工作要求和曲格平老先生的慈善助学精神。

在环境保护公益资助领域，中华环境保护基金会辽宁代表处开展的辽宁环境科研教育“123工程”是一个有益的尝试，“123工程”的持续开展，必将推动辽宁环保科技专业人才的茁壮成长。

作为中国第一家专门从事环境保护事业的全国性公募基金会，我们将一如既往地支持辽宁代表处持续开展好“123工程”。希望“123工程”以解决辽宁省乃至全国环境保护工作的实际需要为根本，以实现节能减排和改善环境质量为核心，突出抓好重点环境保护科研项目的资助，高度重视科研成果的实际应用，为政府全面提升环境科技支撑做出更大的贡献。

中华环境保护基金会副理事长 秘书长 李伟

前 言

由中华环境保护基金会辽宁代表处实施的“格平绿色行动——辽宁环境科研教育‘123工程’”是指：中华环境保护基金会辽宁代表处每年出资100万元，分别资助10名博士研究生每人3万元、20名硕士研究生每人2万元、30名环境保护专业应届本科生每人1万元。资助金用于补助研究生的环境保护课题研究经费及补助品学兼优、家庭贫困本科生学习和生活费用的一项环境保护公益活动。

2009年，首届“123工程”实施后，收到辽宁省高等院校和科研院所的大量课题申请。经过认真讨论评审，共选出10个博士研究生、20个硕士研究生的研究课题和30名本科大学生给予资助，2010年，辽宁代表处分别在沈阳和大连，对获得第一届“123工程”资助的本科生、硕士和博士研究生发放了资助金。目前，“123工程”已举行五届，辽宁共有300名优秀的环保学子获得资助。

“123工程”取得了良好的成效。目前，已有《沈抚环境与经济同城发展战略研究》等60余篇环保科研课题公开发表，获得各项专利30余项，在SCI发表论文40余篇。其中，中国医科大学博士生董光辉凭借基金会资助完成的《新型“复合型空气污染”对儿童呼吸系统疾病的影响》课题，一年内在SCI刊物上发表17篇论文，课题成果用于科研和临床指导，董光辉于2011年被学校破格晋升为副教授和博士生导师；沈阳建筑大学硕士研究生郭琦正是由于得到了“123工程”的资助，使其完成了《本钢一铁厂的定留与重塑》研究课题，并已在本钢一铁厂的更新改造中得到应用；沈阳航空航天大学硕士研究生问鼎同学的《基于无人机的风电类建设项目竣工环保验收方法》研究课题，成功应用于辽宁的风电类建设项目环境保护竣工验收工作。沈阳药科大学的本科生吴永奇同学得到资助后，发奋学习，成绩优秀，本科毕业后，被保送读研。

“123工程”对加快辽宁省环境保护工作和教育事业的发展、培养和储备环境科技人才、促进辽宁生态省建设起到了积极作用。中华环境保护基金会副理事长、秘书长李伟认为：“123

工程”结合人才培养的实际，对本科、硕士、博士进行选择性的资助，充分发挥了环保公益资助金的社会效益。“123 工程”的持续开展，将推动辽宁环保专业人才的培养。辽宁省环境保护厅朱京海厅长说：要借助中华环境保护基金会的公益资助在辽宁实施好“123 工程”，用这项公益工程，打造一所环境保护科技队伍的“黄埔军校”。

《格平绿色行动——辽宁环境科研教育“123 工程”论文集》正是收录了历届“123 工程”部分获得资助学生的论文，以此来宣传中华环境保护基金会的社会影响、展示基金会资助成果、鼓励指导更多的学子们参与到环境保护事业中来。

限于编者的水平和经验不足，成书仓促，缺点错误在所难免，敬请各位读者批评指正。

编 者

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Study of the performance on magnesia base catalyst used for flue gas denitration^{*}

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Abstract: To control contamination of flue gas emission NO_x , the author mixed method of preparing new magnesia base catalyst and researched on denitrifying flue gas by directness catalytic decomposition. Usually removal rate of NO is regarded on the main evaluation criteria. The author analysed calcination temperature of catalyzer manufacturing process, the temperature of flue gas desorption tower, bed height and efficiency denitrification of NO concentration. And discussed the result of both denitrate fore-and-aft of FT-IR and XRD. The experiment indicate that magnesia base catalyst is available to directness catalytic decomposition denitrificate flue gas. Through denitrification rate is from 85% to 95%. The ingredient of magnesia base catalyst is made from magnesia, firming agent, addition agent. Height denitration bed was 4~5 cm, denitration reaction temperature is 130~170°C. Through the analyses above, presumed that magnesia base catalyst exist activating of vice, and analyzed this vice preliminarily.

Keywords: denitrification, NO_x flue gas, direct catalytic decomposition, catalyst

1 Introduction

SO_2 and NO_x in burning coal emission flue gas is the main source of the air pollution in China. Nowadays it's the focus research to get rid of SO_2 and NO_x and turns them economically and effectively into useful materials to be the focus of studying. At present, industry adopts different kinds of denitrate technology to dispose flue gas, each of them have advantage and disadvantage applicable conditions, for instance, adsorption method^[1], reduction method^[2,3], decomposition method^[4,5], and oxidation method^[6,7] and so on^[8]. To the realities of increasing of flue gas emission, resources lacking and environmental protection funds shorting in China. We explored a new way of denitration and to researched by direct catalytic decomposition method. Denitration research was carried out through development of the catalysis adsorbent. And the method remove NO_x through decomposing and reducing NO_x into nitrogen and oxygen.

In condition of imitating flue gas, the author explored magnesia base catalyst how to dispose NO_x (the principal ingredients are NO, accounts for 90%) in flue gas by way of direct catalytic decomposition, anylized the affection to denitrify by disposal condition, and anylized reaction theory primarily.

^{*} This research was supported by Liaoning environmental research and education "project 123"(CEPF2008-123-1-1), the Key Laboratory of Eco-remediation of Contaminated and Resources Reuse (Ministry of Education) foundation of China (NO.ERCERR0807), the Science and Technology Program of Shenyang of China (1071057-9-00) .

2 Experiment

2.1 Catalyst Preparation

Preparation of magnesia base catalyst by mixing method^[9-11], Main raw materials was the magnesia of the southeast Dashiqiao of Liaoning Province of China, curing agent (analytical reagent A.R. Shenyang City Xinhua reagent factory), Additives (industrial products). Figure 1 shows the process.

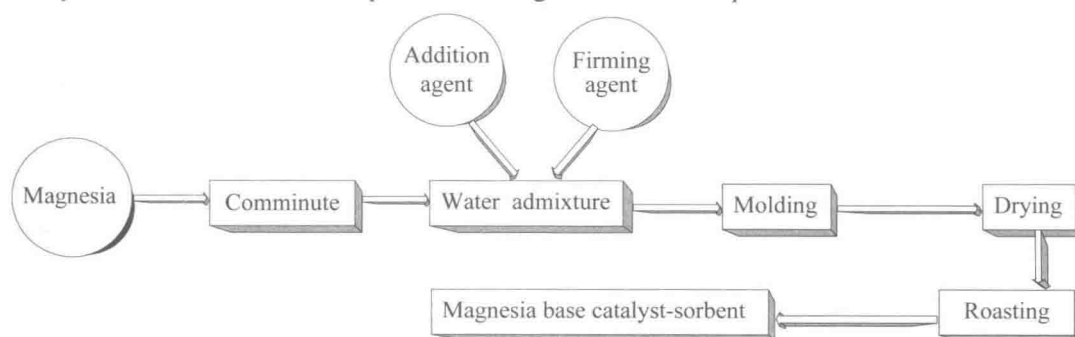


Fig.1 Flow chart of preparation of magnesia base catalyst

Experimental conditions is listed as follow: Magnesia was crushed into powder, particle size of the powder was less than 0.076 mm. All kinds of raw material was blended for 10 minutes after mixing them. Then pressed the mixture into cylinder-style particle, and average diameter of the particle was about 3 mm, average height was 5 mm. The rate of activation heating was control within 10~20°C/min. Temperature of mixture was raised to 500°C in the air, then fixed the temperature for 3 hours.

2.2 Catalyst Characterization

In the furnace, the author used digital temperature to prepare. magnesia base catalyst's catalytic activity was characterized by NO removal rate (the ratio of that the difference of NO concentration of the simulation flue gas enters and out the denitrating tower, and enters'). Fourier transform infrared spectroscopy (Thermo Nicolet Corporation) was used to analyse FT-IR of magnesia base catalyst of before and after Denitration. X'Pert Pro diffraction (PANalytical B.V.) was used to analyse the characterization results of X-ray diffraction (XRD).

2.3 Catalytic Experiments

Dynamic fluid systems was adopted in denitration simulation experiments. The system was made up of gas installations, gas-mixing device, denitrification tower and the NO_x on-line monitoring devices, and so on. Figure 2 shows processes of experimental.

Simulated flue gas devices is made up of N₂, NO and O₂ cylinders and the corresponding valve, a flow meter and the buffer bottles composition etc. Reactor gas imports simulated flue gas composition, NO, O₂ and water vapor volume fraction of 0.1%, 6%, 3% and the remainder is N₂ balance gas. Magnesia base catalyst was used by 7~8 g in the experiment, space velocity is 2 720 h⁻¹, the reaction was carried on at special temperature. Active evaluation of catalysts was made by fixed bed catalytic reactor at normal pressure. finally, simulated flue gas entered NO_x on-line monitoring of the system. Flue gas analyzer (Germany, MRU95/3CD) and hydrochloric acid and naphthalene diamine spectrophotometry was to monitor concentration of NO_x on-line, before and after

denitrification^[12,13].

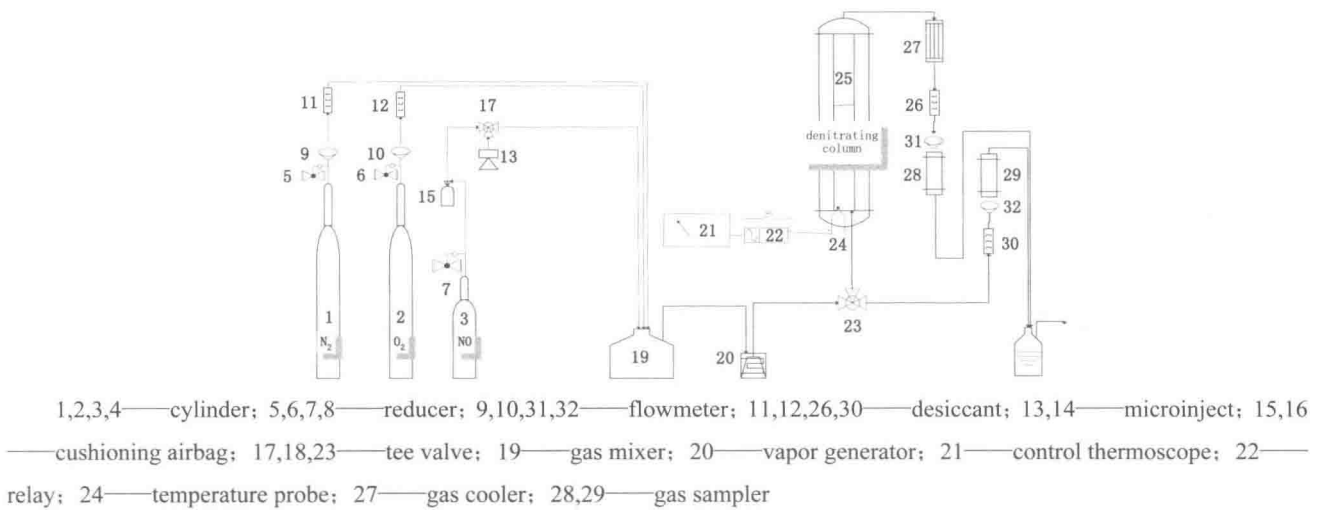


Fig.2 Schematic diagram of experimental apparatus of denitrification by magnesia base catalyst

3 Results and discussion

3.1 Effect of Calcination Temperature

Figure 3 shows different denitrification rate of magnesia base catalyst at different calcination temperature. In the range of 250°C to 350°C, NO removal rate increases according to the increasing of calcination temperature. NO removal rate instable with much vibration when bed temperature is at 90°C. The NO removal rate by catalyst decreases when roasting temperature gets 650°C except bed temperature in 130°C.

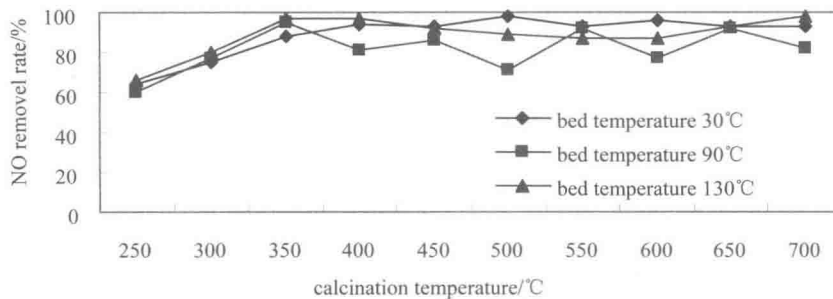


Fig.3 Effect of NO removal rate by temperature changing after 2 hours calcination temperature

Figure 3 shows that gradualde composition of organic matter and crystal water in magnesia base catalyst-sorbent increase factor of porosity, pore volume and specific surface area in activation stage when roasting temperature is below 350°C^[9]. Magnesa is getting activation at the same time, and activity defects come into being. Temperature increasing promotes catalytic activity^[14,15]. Volatile evaporate completely when roasting temperature above 350°C, and magnesia is nearly activated completely. Catalytic activity keeps stable when roasting temperature is in the range of 350°C to 650°C, and crystal catalyst particles enlarge gradually with temperature increasing. As roasting temperature exceed 650°C, higher temperature result in catalyst surface sinter. Besides specific area degression, density increase, specific gravity increase, the activating vice gradually turned,

and result in degression of magnesia base catalyst activity. In the meantime, catalyzer in the proximity of bed temperature at 90°C , denitrate efficiency is in a state of instability. Because under this temperature simulation flue gas of vapor in the fringe of seethe and aerification, and water in catalyst surface forms the liquid membrane to impact catalyzer normal catalytic activity. But denitrate efficiency keep stabilize when bed temperature in 130°C , and this is to explanat higher bed temperature in favor of catalytic conversion of magnesia base catalyzer.

3.2 Effect of Calcination Time

When the calcination temperature is at 400°C , the influence of calcination time on NO removing rate in Figure 4. As a whole, the removal rate of NO is inferiority in initial stage (less 2 h). Subsequently, 3 to 4 h run up to higher level, and begin decrease in five hours later.

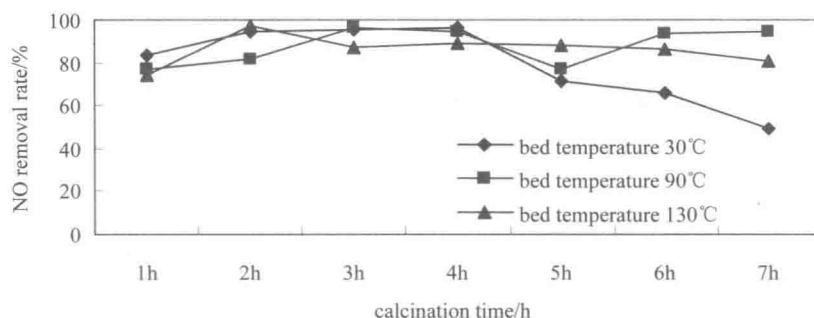


Fig.4 Effect of calcinations time on the NO removal rate at 400°C

Such as Figure 4, Roasting activated, through thermal decomposition reaction in initial stage, and remove magnesia base catalyst's chemical combined water, thereafter organic matter, etc. the volatile material or impurity begun to volatilize or decompose. At the same time the chemical bond between magnesia and firming agent crystal further integration, and the surface forms the active defects. Going on with roasting, original pores became larger and generate new pores. Pore structure upgrowth arisen, and it make in favour of enhanced magnesia base catalyst-sorbent of adsorption activity. As time go on, it result in microlite relevance sinter, in the meantime it has improved catalyzer mechanical strength. when calcination time exceed 5 h, catalyst excessive sintering, resulting in reduced activity, denitrification efficiency decreased. According to Figure 4, activity decline quickly when bed temperature lower. it is obvious that calcination time longer to make catalyzer easy sinter. Sintered catalyst activity scale reduced after fritting, the active accommodation is reduced, the influence of denitrification is relatively large. So in the range of flue gas normal temperature, the temperature more higher and the magnesia base catalyst's catalytic activity is more actively, and denitrate efficiency would better.

3.3 Influence of Adsorber Bed Temperature

In Figure 5, catalyzer denitrate efficiency was evaluated respectively when denitrating column bed temperature was at $40, 70, 90, 110, 130, 150, 170^{\circ}\text{C}$ and the figure also shows that NO removal rate increased gradually with the raise of bed temperature. At 130°C , it gets a climax. Denitrate efficiency keeps stable in the rang of $97.2\% \sim 99.6\%$ when temrperature overpasses 130°C . Such embodies that catalytic activity is not obvious at low temperature, it make denitrification rates lower and catalytic activity enhanced beyond 130°C . Catalytic activity is activated at higher temperature and gets a ideal status. On the curve of lowest bed (4 cm) in the figure, the temperature drops followed by a raise. The bottom value is at 90°C because such temperature just lies the

edge on which water evaporates and boils quickly. At this temperature, the film of contact surface between catalyst and NO may present a instable status among liquid phase, solid phase and gas phase, and such status cannot exert the reductive effect of NO by the vice on O lattice imperfection of activity MgO crystal surface. The film of contact surface is liquid below 70°C and it's gas above 110°C, they are all unique stable structure.

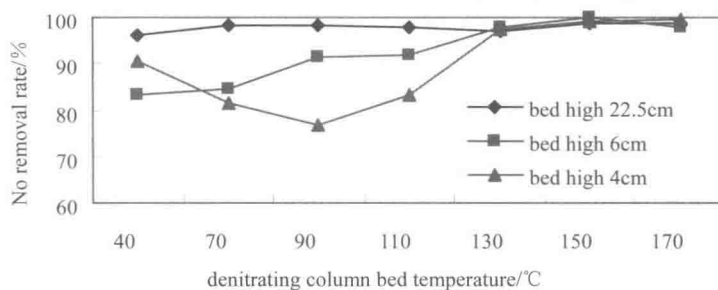


Fig.5 Influence of amount of bed temperature and bed height on denitrification efficiency

3.4 Bed Height Impact on the Efficiency of Denitrification

At the temperature of 130°C, Bed height has small effect on the NO removal efficiency when bed height is above 4 cm. The NO removal efficiency falls down with the lower of bed height when bed height is below 4 cm. According to Figure 6. It is because adsorption catalysis action of catalyzer to NO need touch time and response time, and catalytic activity perfect only when bed height is above 4 cm. According to there variables of bed height in Figure 5, selected bed height take small effect on NO removal efficiency. There is some difference at lower temperature, while there is almost little difference above 130°C, the distance between the maximum and minimum is just in 1%. It presents that catalytic activity is still inevident when NO is adsorpted and catalyzed at lower temperature. Flue gas contact time is longer when bed height is higher, hence catalytic time is prolonged and removal efficiency is increased. When temperature is beyond 130°C, catalytic activity is improved a little and becomes stable, at the same time it behaves more evident catalytic activity, hence NO is removed much more. In this research, bed height has been a lower level, so bed height in other experiments of the research was fixed on 5 cm.

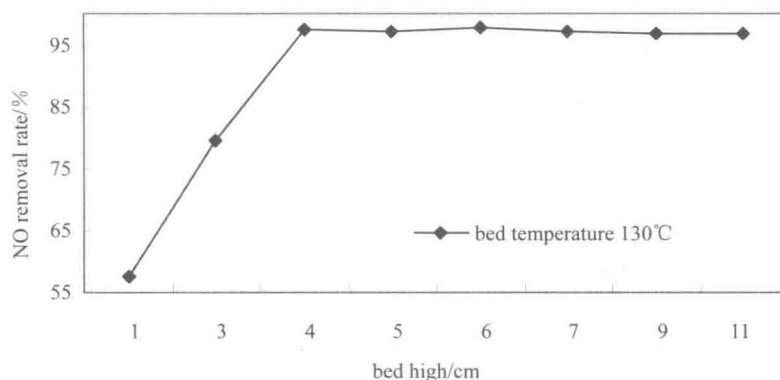


Fig.6 Influence of amount of bed height on denitrification efficiency

3.5 NO Concentration on the Efficiency of Denitrification

In chang of NO removal efficiency is not evident from Figure 7. It takes on increasing trend at 110°C. These

show that the reaction is relatively stable at 130℃, and activity is higher than that of 110℃. So, high temperature of denitiation column is advantage of reaction, and it shows that the reaction has some activation energy, such activation energy accelerates the reaction at special temperature. It also implicates that the reaction is chemical adsorption than physical adsorption. It also implicates that it's an evidence to direct catalytic decomposition, and it suggests that the surface of magnesia base catalyst may exist activating vice.

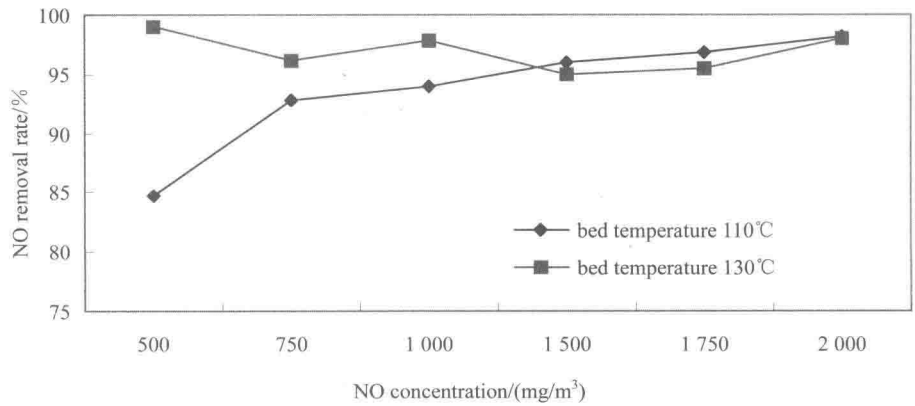
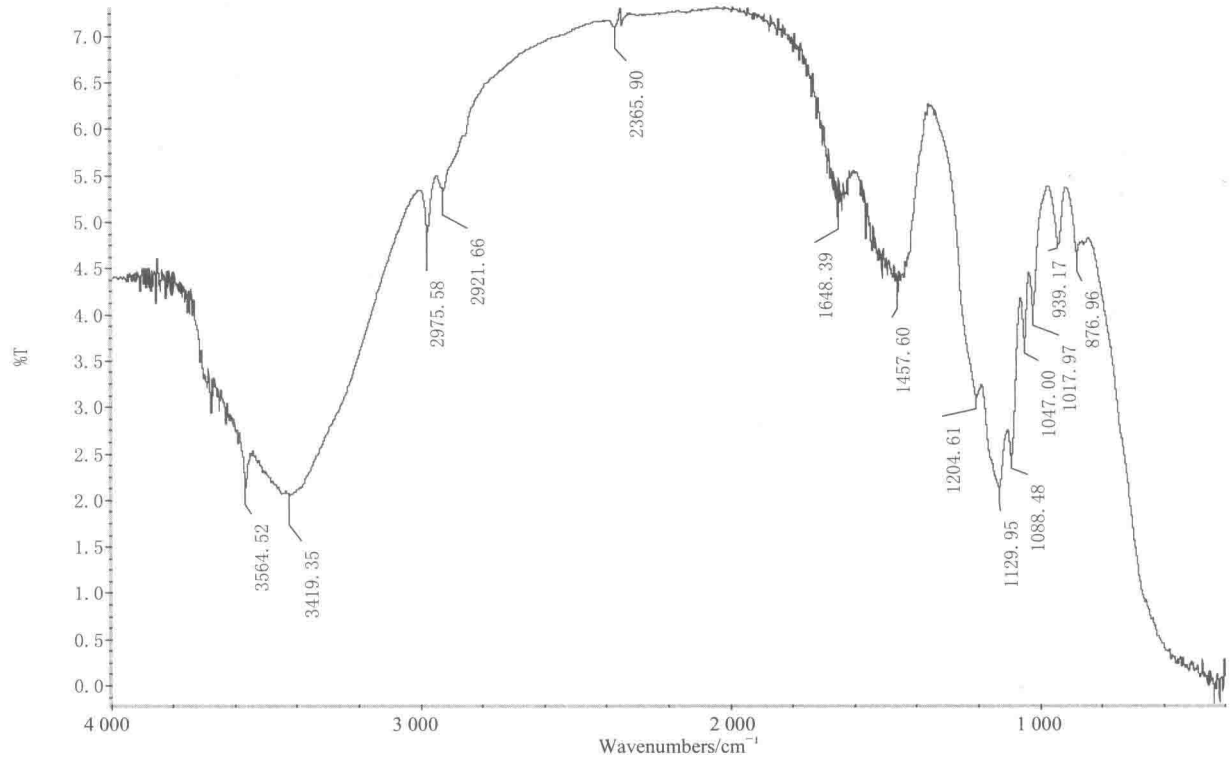


Fig.7 Influence of amount of NO on denitrification efficiency

3.6 Fourier transform infra-red spectra (FT-IR) analysis

Figure 8 shows before and after magnesia base catalyst denitrate Fourier transform infra-red spectra spectrogram. Main characteristic peaks of two Figures have no change, several peak is affected by steam and pressed thickness in test, and their changes exist in whole peak height.



a Before denitrification

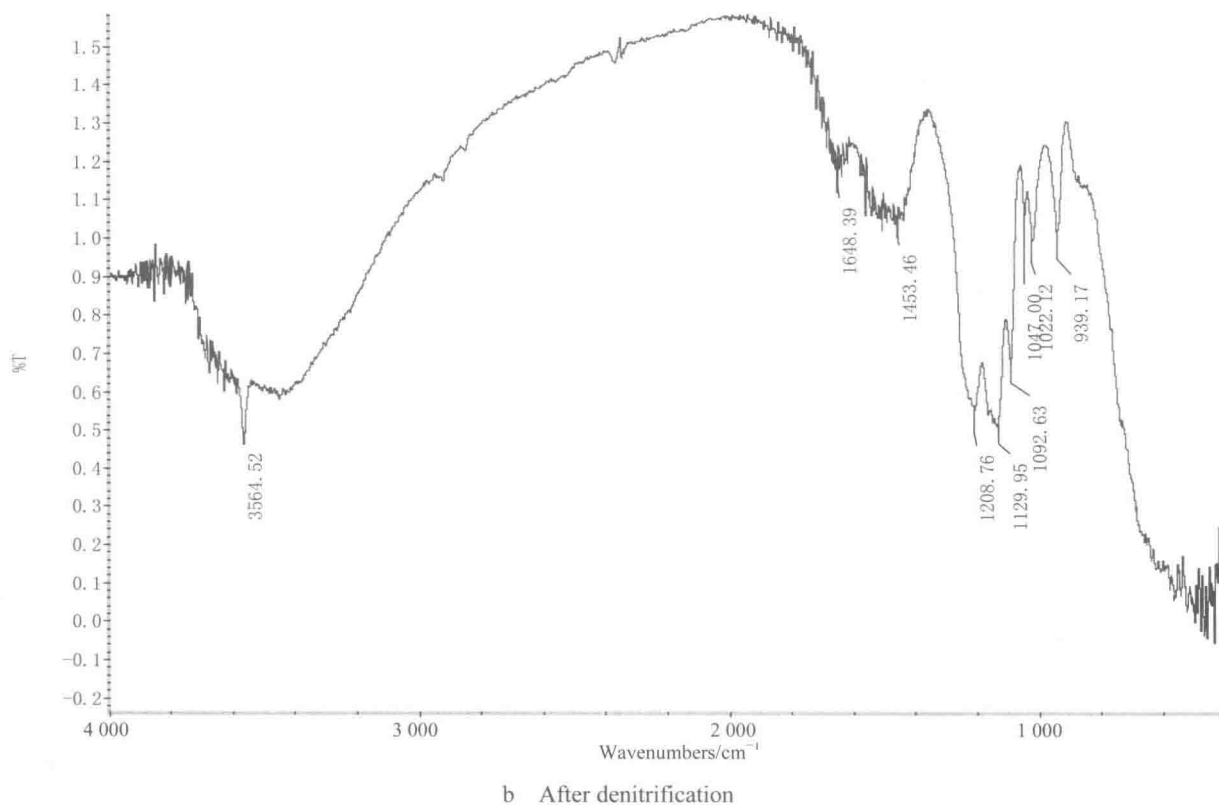
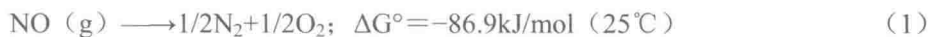


Fig.8

Figure 8 shows that there is weak absorption peak at frequency of $1\,080\text{ cm}^{-1}$, and there is at there is strong absorption peak at $1\,210\sim 1\,025\text{ cm}^{-1}$. This suggests that there is antisymmetric magnesium salt in it. At the same time, there are absorption peak at 528 cm^{-1} and 410 cm^{-1} . It suggests that it contains MgO. Figure 5b shows that NH_3^+ for stretching vibration doesn't exist because there isn't dispersive absorption spectrum between $3\,300\sim 2\,000\text{ m}^{-1}$, and there isn't symmetry, antisymmetric vibration and angle vibration in this range. But NH_2^+ for stretching vibration may exit because there is strong dispersive absorption spectrum. Alkylamine has three groups of peaks near $2\,970\text{ m}^{-1}$, $2\,800\text{ m}^{-1}$ and $2\,480\text{ m}^{-1}$. Figure 8a shows that this peak has existed before new preparation catalyst is denitrated, so it's a result of component translation after rosating. The change of every peak is not evident at every stage of the experiment, this implicate that catalyst's structure doesn't change, so catalyst's feature doesn't change in the process of denitrification. And NO_x adsorption is just a mid-procedure in denitrification.process, and magnesia base catalyst's flaw play a key role on the removal of NO_x . Hence we can conclude that NO_x is decomposed directly by catalyst into N_2 and O_2 :



The result shows that it was directness catalytic decomposition reaction that of experimental study magnesia base catalyst in denitrification process.

3.7 X-ray Diffraction (XRD) Analysis

Figure 9 shows the XRD of magnesia base catalyst after denitrification. Judging from the height and area of X-ray diffraction patterns, we can conclude that the amount of magnesia is more than that of firming agent. The