Catalytic ammonia oxidation to nitrogen (I) oxide

Abstract: The process of synthesis of nitrous oxide by low-temperature catalytic oxidation of NH has been investigated for organic synthesis. The investigation has been carried out by the stage separation approach with NH oxidation occurring in several reaction zones, which characterized by different catalytic conditions. The selectivity for N₂O was 92–92.5% at the ammonia conversion of 98–99.5% in the optimal temperature range.

Keywords: Mn-Bi-Ce-Cu-O catalyst; ammonia oxidation; nitrous oxide, technological parameters, pilot testing.
The process of low-temperature catalytic oxidation of NH has been investigated for production of N₂O for organic synthesis. The investigation has been carried out by the stage separation approach with NH oxidation occurring in several reaction zones, which characterized by different catalytic conditions.

**Introduction.** In the recent years, nitrous oxide has become used as a mild oxidizer for partial oxidation of hydrocarbons, for example oxidation of benzene to phenol. That is why, the process for direct ammonia oxidation is of interest to numerous researchers [1–4]. For these applications the substantial attention is focused on the improvement of the N₂O production technology. Nitrogen (I) oxide is produced for medical purposes by thermal decomposition of ammoniac salt peter. The hazards of the existing methodology of the thermal decomposition of ammonia nitrate involve the possibility of explosion of ammonia nitrate at heating and speeding up of the side reactions with increasing temperature.

The catalytic oxidation of ammonia to N₂O at low temperatures (200–400°C) is a promising and more economically efficient technique. The catalytic oxidation of ammonia by technical oxygen to N₂O has been investigated by N. I. Il’chenko [4]. N₂ and N₂O are formed during low temperatures ammonia oxidation in reactions 4NH₃ + 3O₂ = 2 N₂ + 6H₂O and 2NH₃ + 2O₂ = N₂O + 3H₂O.

In the course of these reactions the formation of unstable intermediate particles (OH, ONH₂, ONH₃, NH₂, N, NH etc.) is also possible in parallel and sequential stages. In the present work we study different steps of the ammonia oxidation process and determine the influence of each step on the product yield. For this purpose, the ammonia oxidation reaction steps are carried out in separate reaction spaces, each having a different catalyst. To some extent, this approach avoids parallel reactions that may occur within a single reaction space and that are responsible for nitrogen unbounding. The influence of various technological parameters and various catalysts [5] on the ammonia oxidation process and production of nitrogen (I) oxide can be investigated as well.

Experimental. The measurements have been carried out in a reactor with two spatially separated catalysts. This allowed us to investigate individual stages of ammonia oxidation and the influence of various stages on the product yield as well as to avoid the parallel reactions that can occur in same reaction space and result in unbounding of nitrogen. The influence of technological parameters on the process of ammonia oxidation to N₂O is studied for a variety of catalysts.

The catalysts for ammonia oxidation were prepared from oxides of Mn, Bi, Cu and Ce as well as from nitrates of these metals by sol-gel method. Before ammonia oxidation the catalysts were heated to 200°C for two hours at the presence of an oxygen flow. A layer of catalyst 2 was stacked on the quartz grid of the reactor. A layer of broken quartz glass with particle sizes from 3 to 4 mm was located above the catalyst layer. A layer of catalyst 1 was stacked on top of the broken quartz glass layer. The design of the device allowed us to vary the distance between the catalysts up to 55 mm. The experiments show that in the system containing two separate catalyst layers, the N₂O yield can be sharply increased by 7 to 15% depending on the separation distance between layers. However, the some negative phenomena were observed in parallel with increase of the yield. If the catalyst 1 begins to work in an autonomous thermal mode, the ammonia oxidation process takes place solely on catalyst 1, and the temperature of this catalyst sharply increases. As a result the temperature of catalyst 2 decreases below the ignition temperature and catalyst 2 stops working. If the catalyst 2 is heated externally, then catalyst 2 switch on and dominates in the catalysis. As a result catalyst 2 uncontrollably heats up and catalyst 1 stops working as soon as its temperature decreases below the ignition temperature.

In order to eliminate the mutual influence of the catalysts and provide the more stable working conditions a series of experiments have been carried out in a quartz reactor (d = 20 mm). The design of reactor allowed us to increase the distance between the catalysts up to 90 mm. In this way, the temperature of one of catalysts was fixed in the range 200–225 °C, while the temperature of the other catalyst was slowly changed. The outlet nitrous gases were tested for the content of N₂O by gas chromatography method.

**Results and Discussions.** A low-temperature oxidation of ammonia exhibits several unusual feature. At low temperatures the reaction mainly proceeds to N₂ and N₂O, while at high temperatures NO is produced much more efficiently. Formation of unstable intermediate products, such as O, OH, ONH₂, NH₂, N, NH, HNO etc., is also possible at low temperatures. The dependence of the N₂O yield on the temperature differences between the two catalysts was studied.

The data were obtained with Mn-Bi-Cu-Ce-O as catalyst 1 and manganese (II) oxide promoted by palladium [3] as catalyst 2. The study shows the maximum yield at ΔT=0. The date temperature dependences of the
Catalytic ammonia oxidation to nitrogen (I) oxide

nitrogen (I) oxide yield at various gas flow rate show that at optimum temperatures and low concentrations of ammonia in the gas mixture the N2O yield can reach 98–99%.

At high ammonia concentration the N2O yield can be decreased considerably. The N2O yield reaches 80–82% for a given catalyst at the linear gas flow rate v = 0.005 m/s and NH3 concentration of 10%. Under these conditions an optimal temperature of the process is in the range 210–270 °C. The N2O yield reaches 98–99% at optimal temperature and 4.14% ammonia concentration in gas mixture. It is necessary to note that the competing side reactions leading to the formation of nitrogen do not take place at the optimal temperature. At temperatures below 200 °C the reaction rate is decreased considerably and NH3 remains in the gas mixture even after passing through the catalyst.

The transformation of NH3 to nitrogen (I) oxide at various temperatures and the linear gas flow rate v = 0.015 m/s and the temperature dependence of the degree of transformation of NH3 to N2O and the yield of the higher oxides of nitrogen were studied. The data indicate that an optimal temperature of transformation of NH3 to N2O at the ammonia concentration of 10% can be found in the range 340–480 °C, while at the ammonia concentration of 7.7% it is in the range 330–370 °C. With the ammonia concentration 4.4% the degree of transformation of NH3 into N2O is very close to 98–99% at temperatures above 340 °C. For the ammonia concentrations 10%, 7.7% and 4.4% the parallel side reactions occur at temperatures above 250, 290 and 330 °C, accordingly.

Comparison of the N2O yields at various levels of ammonia specific loading on the catalyst and at various NH3 concentrations gives the following results. With the specific ammonia loading on the catalyst 11.76 kg/m² and NH3 concentration of 7.7%, the N2O yields 75% at 280 °C that avoids formation of the higher nitrogen oxides. The same yield was obtained at the specific ammonia loading on the catalyst 11.4033 kg/m². At the 10% NH3 concentration in gas mixture and the specific ammonia loading on the catalyst 15.27 kg/m², the N2O yield at 280 °C was 57%. Thus, at the ammonia concentration of 10% the N2O yield is lower than at the ammonia concentration of 7.7%. Moreover, the higher nitrogen oxides can be formed at 280 °C and NH3 concentration of 10%. A pure nitrogen (I) oxide without the higher oxides is formed at the NH3 concentration of 10% only if temperature is decreased to 250°C. However, at this temperature the degree of NH3 transformation is 40% and the N2O yield is only 7.9 kg/m². At the NH3 concentration in the initial gas mixture equal 4.4% and the specific loading on the catalyst equal 6.7 kg/m², N2O yield is as low as at the NH3 concentration of 7.7%. The concentration of NH3 in gas mixture is a more valuable parameter in the oxidation process than the catalyst specific loading.

An increase of catalyst specific loading at the ammonia concentration of 10% results in a sharp decrease in the degree of transformation of NH3 into N2O. The N2O outlet sharply decreases as well. For example, the growth of catalyst specific loading by factor of 5 increases the N2O yield only by factor of 2.45. At the same time with the ammonia concentration of 7.5% a factor of 5 growth of the catalyst specific loading increases the N2O yield in 4.44 times. With a further decrease of the ammonia concentration to 4.1–4.4%, the N2O yield is increased 5.2 times at 5.3 times growth of the catalyst specific loading. With a simultaneous decrease of the ammonia concentration and increase of the catalyst specific loading, the N2O yield is increased as well. The temperature interval within which the N2O yield is near 98–99% is reduced. For example, if the gas flow rate is 0.005 m/s and ammonia concentration in gas mixture is 7.6%, the width of the favorable temperature interval is about 140 °C. If the gas flow rate is increased to 0.025 m/s, the width of the favorable temperature interval decreases to 60 °C.

Conclusions. The results of the investigation of the low-temperature NH3 oxidation process using two catalysts located in separate reaction zones allows us to control and optimize the yield of N2O. The proposed design of the reaction chamber provides the increase of the N2O yield by 7–15% under autothermally conditions of NH3 oxidation. The degree of NH3 oxidation into N2O is increased without any detectable nitrogen unbounding. The results of the investigation of the ammonia oxidation process using several catalysts and a variety of gas mixture flow rates, temperatures and catalyst specific loadings show that under optimal conditions a practically complete transformation of ammonia into N2O is possible.

References:

Application of gis technology in the analysis of relief the south slope of Greater Caucasus

Abstract: We consider the evolutionary development of geographic information systems (GIS) in the Republic and the concept of providing a high-rise in the South slope of the Greater Caucasus (Qabala).

Keywords: geodesic height, normal height, geodetic measurements.

Mustafayev Yasin Isimoglu, Baku State University Baku, Azerbaijan
E-mail: yasin@geoid.az

Применение гис- технологий при анализе рельефа южного склона большого Кавказа

Аннотация: Рассмотрено эволюционное развитие географических информационных систем (ГИС) в Республике и концепция системы высотного обеспечения на территории Южного склона Большого Кавказа (Габала).

Ключевые слова: геодезическая высота, нормальная высота, геодезические измерения.

Введение. Абсолютная высота всех точек в пространстве стран СНГ принимаются от уровня Балтийского моря, являясь составной и неотъемлемой частью решения общих задач геодезии. На современном этапе развития географической науки установлены связи между нивелирными сетями Республики со странами СНГ, входящих в единую сеть нивелирования Европы (UELN), участие которого требует определения оптимальных высот в общеземной системе.


Austrian Journal of Technical and Natural Sciences

№ 9–10  2015
September–October

Vienna
2015
Austrian Journal of Technical and Natural Sciences
Scientific journal
№ 9–10 2015 (September–October)

ISSN 2310-5607

Editor-in-chief
Petra Busch, Austria

Consulting editors
Jolanta Lewicka, Poland
Alajos Fazekas, Hungary

International editorial board
Egor Rachynski, Ukraine
Rostislav Komarov, Russia
Slavka Konstantinova, Bulgaria
Jennifer Mathieson, Scotland
Hong Han, China
Alessandro Massaro, Italy

Proofreading
Kristin Theissen

Cover design
Andreas Vogel

Additional design
Stephan Friedman

Editorial office
European Science Review
“East West” Association for Advanced Studies and Higher Education GmbH, Am Gestade 1
1010 Vienna, Austria

Email:
info@ew-a.org

Homepage:
www.ew-a.org

Austrian Journal of Technical and Natural Sciences is an international, German/English/Russian language, peer-reviewed journal. It is published bimonthly with circulation of 1000 copies. The decisive criterion for accepting a manuscript for publication is scientific quality. All research articles published in this journal have undergone a rigorous peer review. Based on initial screening by the editors, each paper is anonymized and reviewed by at least two anonymous referees. Recommending the articles for publishing, the reviewers confirm that in their opinion the submitted article contains important or new scientific results.

Instructions for authors
Full instructions for manuscript preparation and submission can be found through the “East West” Association GmbH home page at: http://www.ew-a.org.

Material disclaimer
The opinions expressed in the conference proceedings do not necessarily reflect those of the «East West» Association for Advanced Studies and Higher Education GmbH, the editor, the editorial board, or the organization to which the authors are affiliated.

© «East West» Association for Advanced Studies and Higher Education GmbH
All rights reserved; no part of this publication may be reproduced, stored in a retrieval system, or transmitted in any form or by any means, electronic, mechanical, photocopying, recording, or otherwise, without prior written permission of the Publisher.

Typeset in Berling by Ziegler Buchdruckerei, Linz, Austria. Printed by «East West» Association for Advanced Studies and Higher Education GmbH, Vienna, Austria on acid-free paper.
Contents

Section 1. Architecture ............................................................................. 3
Kaplinska Mariana
The transport problem of the market squares regeneration in the historic towns and cities of Galicia ............ 3

Section 2. Biology.................................................................................. 6
Bekk Viktor Viktorovich, Mosina Lyudmila Vladimirovna, Jandarova Julia Aleksandrovna
Biological activity of soils of landfills as an indicator their ecological condition (in the example Salarievo landfill) ...................................................... 6

Section 3. Information technology ................................................................. 10
Aizat Abdyshova
Applied Semantic search with Microsoft SQL Server 2012 ........................................ 10
Grigoryev Alexey Mikhailovich, Khokhlov Igor Alexandrovich, Matviychuk Alexander Rostislavovich, Belousov Alexey Nikolaevich
Corporate cloud of UbR RAS .............................................................................13

Section 4. Mathematics............................................................................ 18
Drushinin Victor Vladimirovich, Lazarev Alexey Alexandrovich
The extension of euler-fermat's theorem and the rationale of carmichael numbers .............................18

Section 5. Mechanics .............................................................................. 21
Vasenin Valery Ivanovitch, Bogomjagkov Alexey Vasilevitch
Investigation of the work of the storeyshaped gating system .................................................21
Dolnyiak Yaroslav, Stypnytskyy Vadym
Rheological modeling and analysis of the influence of technological factors on the 2nd kind's residual stresses formation ................................................................................ 31

Section 6. Medical science ......................................................................... 35
Bisko Nina, Galeb Al-Maali, Mustafin Kairat, Suleimenova Zhanara, Saduyeva Zhazira
Ganoderma lucidum as potential source of biologically active compounds .................................35
Dovlatov Zyaka Asaf ogly, Seregin Alexander Vasilyevich, Loran Oleg Borisovich
Comparison of different types of synthetic suburethral tape for the treatment of stress urinary incontinence in women .................................................................38
Zhurba Oleg Oleksandrovich
Routine and emergency switching to artificial circulation in coronary artery bypass surgery ..................41
Pasiechvili Nana Merabovna
Modern approach to reduce perinatal morbidity and mortality in infectious diseases fetus and newborn ........ 44

Section 7. Mechanics .............................................................................. 48
Kassimov Azad Tursibekovitch
New mechanical regularity in the two-body problem and the explanation of the experimental results PLANC, BICEP2, the phenomenon of superfluidity and other questions of physics .................48

Section 8. Food processing industry .......................................................... 57
Salihanova Dilnoza Saidakbarovna, Pardaev Gulomnazar Eshbaevich, Eshmetov Izzat Do'simbatovich, Agzamkhodjaev Anvarxodja Ataxodjaevich
Preparation of modified carbonic adsorbent for purification of cottonseed oil ................................. 57

Section 9. Agricultural sciences ................................................................. 61
Normuratov Obek Ulugberdievich, Holiyarow Jasur Ziyadullaevich, Hasanov Hayitmurod Abdirashidovich, Mamayusupova Aziz Chorshanbievna, Ibrohimov Giyosiddin Baxriddin ugli
Removal of cotton basic nutrients and utilization of phosphorus from phosphate and compound fertilizers . . . . . .61
Section 10. Technical sciences .......................................................... 65
Jouravel Valentin Ivanovitch, Marichev Vladimir Rjurickovitch
The Identification oil's pollutions sources ....................................... 65
Marichev Vladimir Rjurickovitch, Marichev Nicolay Vladimirovitch
Modern technologies of information and telecommunication systems design ........................................ 69
Masalitina Nataliya Yurevna, Savenkov Anatoliy Sergeevich
Catalytic ammonia oxidation to nitrogen (1) oxide .......................... 71
Mustafayev Yasin Isimoglu
Application of gis technology in the analysis of relief the south slope of Greater Caucasus ...................... 74
Fuchs Sophia Leyvikovna, Pinaeva Lyudmila Nikolayevna, Devyaterikova Svetlana Vladimirovna
Wastewater from the electroplating of cobalt ions ........................... 76
Provatar Alexey Gennadievich
Additive technologies in engine-building ....................................... 78
Rudnev Sergey Georgievitch
Options of discrete capacity in postharvest technology of cereals ......................................................... 82
Khakimov Sherkul Shergazievich, Mardonov Botir Mardonovich
Modeling of movement of foreign impurities soft along the chopping drum during cleaning of raw cotton from small litter ................................................................. 86
Kholiddinov Ilkhombek Khosilzhonovich
Monitoring of the electric power quality characteristics in the low-voltage power grids ......................... 90
Section 11. Chemistry .............................................................. 96
Aliyeva Adelya Mansur, Efendi Arif Javanshir, Rustamova Jeyran Teymur, Kojarova Lyudmila Ivanovna, Magerramova Lala Gyulbala
Oxidative-dehydrogenation of n-amil and izo-amil alcohols on the catalysts of alloys and hydrides alloys Zr with V, Mo, Fe .............................................................................. 96
Appazov Nurbol Orynbassaruly, Akhataev Nurlibek Akarystanuly, Dzhiembayev Bulat Zhazkenovich, Baramisova Gulnara Tursimetovna, Buhukbayev Kairymbek Sabitovich, Tusipova Ulmeken Salamatovna
New growth-stimulating drugs containing N, SE ................................ 101
Gaibnazarov Sunnatilla Bohodirzhanovich
Study physic-chemical characteristic polymer applicable in bore solution ............................................. 104
Djandullaeva Munavara Saparbaevna, Kabulova Lola Baltamuratovna, Atakuziev Temirjan Azim ugly
Silicate brick on resource-saving technologies ................................ 107
Kabulova Lola Baltamuratovna, Djandullaeva Munavara Saparbaevna, Atakuziev Temirjan Azim ugly
Atmospheric stability of Portland cement with 20 and 30% of tuffite additives which were burnt under 600 ° C .................................................................................. 110
Mirzaakhmedova Mavlyuda Ahmedjanovna
Technology of extracting demulsifiers for nonionic oil-water dispersions .................................................. 114
Muhamadiev M. G., Khazratkulova S. M., Mahkamov M. A.
Physic-chemical characteristics of polymers on the basis N-replaced acrylamides of natural ox acids .......... 118
Yusifova Aida Rafig qizi, Rafiyev Azad Natig oglu
Biostability of used lubricating oils for high-speed engines with spark ignition ..................................... 121
Section 12. Electrical engineering .................................................. 125
Zaikov Vladimir Petrovich, Mescheryakov Vladimir Ivanovych, Zhuravlov Yurii Ivanovych,
Reliability-oriented design of thermoelectric cooling devices ................................................................. 125