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Method for the removal of NOx and N2O from the residual gas in nitric acid production

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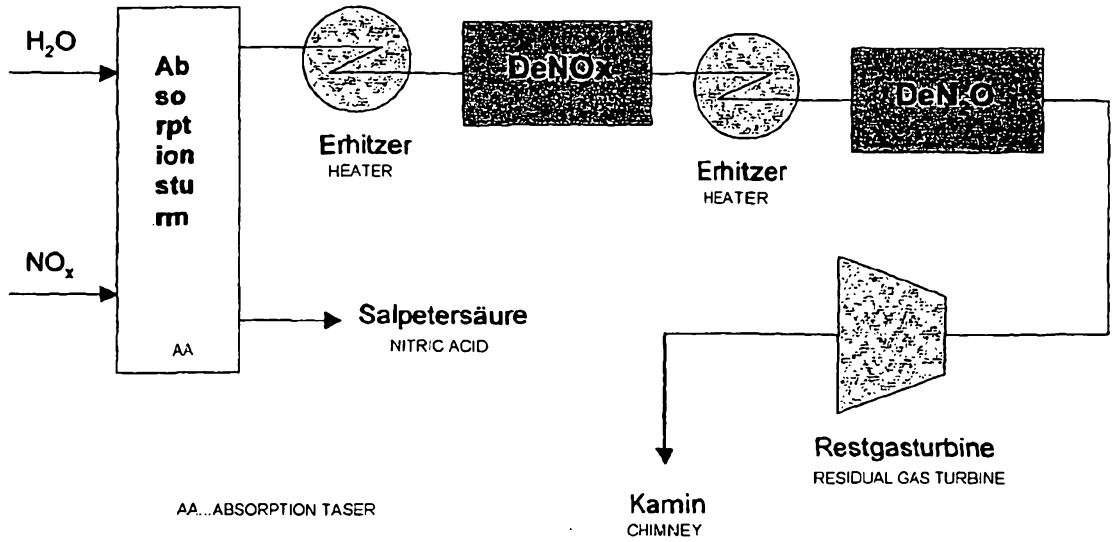
As printed

(54) Title: METHOD FOR THE REMOVAL OF NO_x AND N_2O FROM THE RESIDUAL GAS IN NITRIC ACID PRODUCTION

(54) Bezeichnung: VERFAHREN ZUR BESEITIGUNG VON NO_x UND N_2O AUS DEM RESTGAS DER SALPETERSÄURE-PRODUKTION



(57) Abstract: A method for the removal of NO_X and N_2O from the residual gas in nitric acid production is disclosed. The method comprises passing the residual gases leaving the absorption tower, through a two-stage combination before introduction into the residual gas turbine. In the first stage the NO_x content of the gas is reduced and in the second stage the N_2O content is reduced. The NO_x/N_2O ratio, before entry into the second stage, is in the range from 0,001 to 0,5 and said gas is brought into contact with a catalyst in the second stage. Said catalyst comprises essentially one or several iron-loaded zeolites.

(57) Zusammenfassung: Beschrieben wird ein Verfahren zur Minderung der NO_x- und N_2O-Konzentration aus dem Restgas der Salpetersäureproduktion. Das Verfahren umfasst die Führung des den Absorptionsturm verlassenden Restgases vor Eintritt in die Restgasturbine durch eine Kombination zweier Stufen. Dabei wird in der ersten Stufe der NO_x-Gehalt und in der zweiten Stufe der N_2O-Gehalt des Gases reduziert, das NO_x/N_2O-Verhältnis vor Eintritt des Gases in die zweite Stufe liegt im Bereich von 0,001 bis 0,5 und dieses Gas wird in der zweiten Stufe in Kontakt mit einem Katalysator gebracht, welcher im wesentlichen einen oder mehrere mit Eisen beladene Zeolithe enthält.

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For an explanation of the two-letter codes and the other abbreviations, reference is made to the explanations ("Guidance Notes on Codes and Abbreviations") at the beginning of each regular edition of the PCT Gazette.

Description

Process for eliminating NO_x and N_2O from the residual gas from nitric acid production

The present invention relates to a process for eliminating NOx and N_2O from the residual gas from nitric acid production.

Industrial production of nitric acid HNO_3 by catalytic combustion of ammonia produces a waste gas loaded with nitrogen monoxide NO, nitrogen dioxide NO_2 (together termed NOx), and also nitrous oxide N_2O. While NO and NO_2 have long been recognized as compounds having relevance to environmental toxicity issues (acid rain, smog formation), worldwide limits having been set for maximum permissible emissions of these materials, the focus of environmental protection has in recent years increasingly also been directed toward nitrous oxide, since it makes a not inconsiderable contribution to the decomposition of stratospheric ozone and to the greenhouse effect.

After the adipic acid industry has reduced emissions of nitrous oxide, nitric acid production is the largest source of industrial emissions of nitrous oxide. For reasons of environmental protection, therefore, there is an urgent requirement for technical solutions for reducing nitrous oxide emissions as well as NOx emissions during nitric acid production.

There are numerous versions of processes for eliminating NOx from the waste gas from nitric acid production (termed here the DeNOx stage), such as chemical scrubbing, adsorption processes, or catalytic reduction

processes. Ullmann's Encyclopedia of Industrial Chemistry, Vol. A 17, VCH Weinheim (1991) (D1) gives an overview. Emphasis should be given here to selective catalytic reduction (SCR) of NOx by means of ammonia to give N_2 and H_2O. Depending on the catalyst, this reduction can proceed at temperatures of from about 150°C to about 450°C, and permits more than 90% NO_x decomposition. This is the version of NOx reduction mostly utilized during nitric acid production, but, like the other versions, does not lead to any reduction in N_2O content.

For this purpose the current prior art requires a separate, second catalyst stage, advantageously combined with the DeNOx stage.

An example of the process based on this approach is described in US-A-5,200,162, which claims the decomposition of N_2O in a waste gas which also comprises NOx, downstream of a DeNOx stage. Here, at least one substream of the waste gas which leaves the N_2O decomposition stage is cooled and returned thereto in order to avoid overheating of this stage due to the exothermic nature of the N_2O decomposition process. The invention relates to waste gases whose N_2O content is up to 35% by volume, e.g. to waste gases from adipic acid production.

A process put forward by Shell describes the integrated elimination of NOx and N_2O in the residual gas from nitric acid production (Clark, D.M.; Maaskant, O.L.; Crocker, M., The Shell DeNOx System: A novel and cost effective NOx removal technology as applied in nitric acid manufacture and associated processes, presented at Nitrogen '97, in Geneva, 9-11th February 1997, (D2)).

The Shell reactor system is based on what is called a lateral flow reactor principle, where even relatively low temperatures (from 120°C) are possible for the

operation of the DeNOx stage. An amorphous metal oxide catalyst is used for removing N_2O.

When appropriate catalysts are arranged in the residual gas leaving the absorption column with a temperature of from 20 to 30°C, the latitude for possible operating temperatures is prescribed by the operating temperature of the residual gas turbine.

Specifically, for reasons associated with the technical and economic running of the entire process, the residual gas turbine should most advantageously be operated with entry temperatures < 550°C and with maximum ΔT and Δp.

This is particularly important for eliminating N_2O, since according to current prior art this requires markedly higher temperatures than those needed during catalytic reduction of NOx. The cost-effectiveness of this option is therefore linked to adequate catalyst activity.

Kapteijn F.; Rodriguez-Mirasol, J.; Moulijn, J.A., Appl. Cat. B: Environmental 9 (1996) 25-64, (D3) gives an overview of the numerous catalysts which have been demonstrated to be suitable in principle for decomposing and reducing nitrous oxide.

Metal-exchanged zeolite catalysts (US-A-5,171,533), inter alia, appear particularly suitable for decomposing N_2O.

The zeolites used here are prepared by ion exchange in an aqueous solution comprising metal salts. The metals used for the ion exchange are from the group: copper, cobalt, rhodium, iridium, ruthenium, and palladium. The copper zeolites are highly sensitive to water vapor and rapidly lose their activity under those conditions (M.; Sandoval, V.H.; Schwieger, W.; Tissler, A.; Turek, T.;

Chemie Ingenieur Technik 70 (1998) 878-882, (D5)), while the other metals listed here are relatively expensive.

Using iron-doped zeolite of Fe-ZSM5 type under appropriate conditions, as described in Table 1 in US-A-5,171,533, in the absence of NOx, H_2O, and O_2 at 450°C, only 20% N_2O decomposition was achieved.

The activity of Fe-ZSM-5 for decomposing N_2O is, however, markedly increased in the presence of appropriate amounts of NO, this being attributed to a reaction forming NO_2 as in $NO + N_2O \rightarrow N_2 + NO_2$, catalyzed by Fe-ZSM-5 (Kapteijn F.; Marban, G.; Rodrigeuez-Mirasol, J.; Moulijn, J.A., Journal of Catalysis 167 (1997) 256-265, (D6); Kapteijn F.; Mul, G.; Marban, G.; Rodrigeuez-Mirasol, J.; Moulijn, J.A., Studies in Surface Science and Catalysis 101 (1996) 641-650, (D7)).

In the absence of NOx, higher activity was found for Cu or Co-exchanged zeolites than for the corresponding Fe zeolites.

In the descriptions set out in the prior art (D6, D7) of N_2O decomposition in the presence of an Fe-ZSM-5-catalyst at 400°C, use is usually made of equimolar amounts of NO and N_2O. In D6 and D7, the effect of NOx on N_2O decomposition falls constantly as NO/N_2O ratio sinks, and therefore N_2O decomposition becomes unsatisfactory when the NO/N_2O ratio is below 0.5.

The best results are found when the molar ratio NO/N_2O is 1 or greater than 1.

According to the authors, when this catalyst is used for N_2O reduction in the waste gas from nitric acid production, the NO_2 formed could be returned to the process for obtaining HNO_3. Depending on the version of

the process, the NOx concentration and N_2O concentration in the waste gas are about 1 000 ppm.

WO 99/34901 relates to iron-containing zeolites based on ferrierite for reducing N_2O-containing gases. The catalysts used here comprise from 80 to 90% of ferrierite, and also binders. The water content of the gases to be reduced is in the range from 0.5 to 5%. When various zeolite types are compared, the best results for decomposition of N_2O at temperatures of from 375 to 400°C were obtained using zeolites of FER (ferrierite) type (97% N_2O decomposition at 375°C and $NO/N_2O = 1$). Substantially less decomposition was found when using zeolites of pentasil (MFI) type or mordenite (MOR) type. Indeed, the maximum N_2O decomposition achievable under the above conditions when iron-containing MFI zeolites were used was only 62%.

In the light of the known prior art, the present invention seeks to provide an economic process, in particular for HNO_3 production, which permits not only high levels of NO_x decomposition but also satisfactory N_2O decomposition.

In particular, good results for N_2O decomposition are to be obtained even when the NOx/N_2O ratio is sub-stoichiometric, in particular at the ratios which result after NOx content reduction, i.e. at a ratio < 0.5, preferably < 0.1.

The present invention seeks to achieve the above and provides a process for reducing the NO_x concentration and N_2O concentration from the residual gas from nitric acid production, where the residual gas leaving the absorption column is passed, prior to entry into the residual gas turbine, through a combination of two stages, the first stage (DeNOx stage) reducing the NOx content and the second stage (DeN_2O stage) reducing the N_2O content of the gas, and where the NOx/N_2O ratio prior to entry

of the gas into the second stage [lacuna] in the range from 0.001 to 0.5, preferably in the range from 0.001 to 0.2, in particular in the range from 0.01 to 0.1, and in the second stage this gas is brought into contact with a catalyst which is substantially composed of one or more iron-loaded zeolites.

Catalysts used according to the invention are composed substantially of one or more iron-loaded zeolites, preferably > 50% by weight, in particular > 70% by weight. For example, alongside an Fe-ZSM-5 zeolite there may be another iron-containing zeolite present in the catalyst used according to the invention, e.g. an iron-containing zeolite of the MFI type or MOR type. The catalyst used according to the invention may moreover comprise other additives known to the skilled worker, e.g. binders.

The catalysts used for the DeN_2O stage are preferably based on zeolites into which iron has been introduced via solid-phase ion-exchange. The usual starting materials here are the commercially available ammonium zeolites (e.g. NH_4-ZSM-5) and the appropriate iron salts (e.g. $FeSO_4 \times 7\ H_2O$), these being mixed intensively with one another by mechanical means in a bead mill at room temperature. (Turek et al.; Appl. Catal. 184, (1999) 249-256; EP-A-0 955 080). These citations are expressly incorporated herein by way of reference. The resultant catalyst powders are then calcined in a furnace in air at temperatures in the range from 400 to 600°C. After the calcination process, the Fe zeolites are thoroughly washed in distilled water, and the zeolites are filtered off and dried. The resultant Fe zeolites are finally treated with the suitable binders and mixed, and extruded to give, for example, cylindrical catalyst bodies. Suitable binders are any of the binders usually used, the most commonly used here being aluminum silicates, e.g. kaolin.

According to the present invention, the zeolites which may be used are iron-loaded zeolites. The iron content here, based on the weight of zeolite, may be up to 25%, but preferably from 0.1 to 10%. Particularly suitable zeolites here are of the type MFI, BETA, FER, MOR, and/or MEL. Precise details concerning the build or structure of these zeolites are given in the Atlas of Zeolithe Structure Types, Elsevier, 4th revised Edition 1996, which is expressly incorporated herein by way of reference. According to the invention, preferred zeolites are of MFI (pentasil) type or MOR (mordenite) type. Zeolite Fe-ZSM-5 type are particularly preferred.

According to the present invention, DeN_2O catalysts are arranged in combination with an upstream DeNOx stage, between the absorption column and the residual gas turbine, in such a way that the residual gas leaving the absorption column is first passed at temperatures < 400°C, in particular < 350°C, into a reactor (first stage) in which the NOx content of the gas is reduced to, for example, < 100 ppm (cf. Figure 2). The operating pressure for this first stage is preferably from 1 to 15 bar, in particular from 4 to 12 bar.

The upstream DeNOx stage corresponds to a process usually used in nitric acid plants according to the prior art for reducing the amount of NOx emissions. However, the remaining NOx content of the residual gas has to be sufficiently high to permit the cocatalytic effects of NO or NO_2 to be active in the downstream DeN_2O stage.

If the DeN_2O stage is operated without upstream DeNOx, i.e. if the entering stream has approximately equimolar amounts of NO and N_2O, return of the NO_2 formed by $NO + N_2O \rightarrow N_2 + NO_2$ into the HNO_3 process is uneconomic, due to the relatively low NO_2 concentration, < 2 000 ppm.

The N_2O content of the gas remains substantially unaltered in the DeNOx stage. After leaving the first stage, therefore, the NO_x content of the gas is usually from 1 to 200 ppm, preferably from 1 to 100 ppm, in particular from 1 to 50 ppm, and its N_2O content is from 200 to 2 000 ppm, preferably from 500 to 1 500 ppm. The resultant NOx/N_2O ratio after leaving the DeNOx stage is from 0.001 to 0.5, preferably from 0.001 to 0.2, in particular from 0.01 to 0.1. The water content of the gas, both after leaving the absorption column and, respectively, the DeNOx stage and after leaving the DeN_2O stage, is usually in the range from 0.05 to 1%, preferably in the range from 0.1 to 0.8%, in particular in the range from 0.1 to 0.5%.

The residual gas conditioned in this way is then passed into the downstream DeN_2O stage, where decomposition of the N_2O into N_2 and O_2 is brought about by utilizing a cocatalytic effect of NOx in the presence of the appropriate zeolite catalyst.

Surprisingly, it was found that in the presence of the iron-containing zeolite catalysts used according to the invention N_2O decomposition is drastically increased (cf. Figure 1) even in the presence of small amounts of NO_x, i.e. when the molar NO_x/N_2O ratio is < 0.5. An effect which becomes markedly more pronounced as the temperature increases. According to the present invention, therefore, at 450°C, for example, a molar NO_x/N_2O ratio of 0.01 is still sufficient to lower the N_2O concentration from 72% to 33% in the presence of an Fe-ZSM-5 catalyst. This is all the more astounding since in the prior art the accelerated decomposition of N_2O is attributed to the abovementioned stoichiometric reaction of N_2O with NO. If the temperature is sufficient it appears that if the NO_x/N_2O ratio is small NO_x adopts the role of a homogeneous cocatalyst which accelerates N_2O decomposition as in $N_2O \rightarrow N_2 + 1/2\ O_2$.

If the NO_x/N_2O ratio is within the above mentioned limits, maximum decomposition of N_2O is possible in the downstream DeN_2O stage. As soon as the ratio falls away below 0.001, N_2O decomposition also sinks to values which become unsatisfactory (cf. Example 5). The content of N_2O in the process of the invention after leaving the DeN_2O stage is in the range form 0 to 200 ppm, preferably in the range from 0 to 100 ppm, in particular in the range from 0 to 50 ppm.

As now claimed, according to one aspect, the present invention provides a process for reducing the NO_x concentration and N_2O concentration from the residual gas form nitric acid production, where the residual gas leaving the absorption column is passed, prior to entry into the residual gas turbine, through a combination of two stages, the first stage reducing the NO_x content by catalytic reduction, and the second stage reducing the N_2O content of the gas by decomposition into nitrogen and oxygen, and where the molar NO_x/N_2O ratio prior to entry of the gas into the second stage is in the range from 0.001 to 0.5, and in the second stage this gas is brought into contact with a catalyst which comprises one or more iron-loaded zeolites, the operating pressure in the second stage being from 4 to 12 bar.

Preferably the first stage is operated using the SCR process.

The operating temperature for the DeN_2O stage here is in particular determined by the desired degree of decomposition of N_2O and the amount of NO_x present in the residual gas, but also, as is known to the skilled worker and like almost all catalytic waste gas purification processes, highly dependent on the catalyst loading, i.e. on the waste gas throughput based on the amount of catalyst. The operating temperature for the second stage is preferably in the range from 300 to 550°C, in particular in the range from 350 to 500°C, the pressure being in the range from 1 to 15 bar, in particular from 4 to 12 bar. As pressure rises, the cocatalytic action of NO_x on N_2O decomposition becomes greater, and increase of pressure therefore permits a further drop in operating temperature.

In determining or setting the operating temperature, account also has to be taken of the content of oxygen and H_2O. This content can vary within certain limits, depending on the mode of operation and on the version of the process used for nitric acid production, and inhibits N_2O conversion. The O_2 content is in the range from 1 to 5% by volume, in particular in the range from 1.5 to 4% by volume.

N_2O decomposition of > 90%, in particular > 95%, can therefore be achieved at temperatures in the range from 300 to 550°C, preferably from 350 to 500°C, using the iron-containing zeolite catalysts used according to the invention. As temperature rises it is even possible to achieve satisfactory N_2O decomposition when the NO_x/N_2O ratio is 0.01.

By combining a DeNOx stage and a DeN_2O stage, the process of the invention permits the NO_x content and N_2O content of the residual gas to be reduced to minimal values during nitric acid production. The arrangement of the DeNOx stage prior to the DeN_2O stage and between absorption column and residual gas turbine moreover makes the process of the invention very economic, due to the continuously rising temperature profile.

Furthermore, the arrangement of both stages prior to the decompression turbine makes the conduct of the process particularly advantageous, since both stages can be operated at superatmospheric pressure (between 4 and 11 bar, depending on the version of the HNO_3 process), resulting in a reduction of the volume of reactor and, respectively, catalyst effectively needed.

Furthermore, since the DeNOx stage operates even at relatively low temperatures, sufficient reduction of NO_x content during plant start-up is also ensured when only little process heat is available.

Another advantage of arranging both stages between absorption column and residual gas turbine in a continuously rising temperature profile is that the residual gas leaving the inventive combination can be introduced, without prior cooling, and without any other measures for waste gas purification, directly to the residual gas turbine for ideal reclamation of compressive and thermal energy.

Examples:

DeNOx stage:
The DeNOx catalyst used as described with NH_3 as reducing agent upstream of the DeN_2O catalyst was a conventional SCR catalyst based on V_2O_5-WO_3-/TiO_2 (cf.,

for example, G. Ertl, H. Knözinger J. Weitkamp: Handbook of Heterogeneous Catalysis, Volume 4, pages 1633-1668). This was operated at a temperature of 350°C. Depending on the amount of NH_3 introduced, various NO_x contents and therefore NO_x/N_2O ratios were set at the outlet from the DeNOx stage.

DeN_2O stage:
An iron-containing MFI catalyst was prepared by solid-phase ion exchange, starting from a commercially available ammonium-form zeolite (ALSI-PENTA, SM27). Detailed information concerning the preparation process may be obtained from: M. Rauscher, K. Kesore, R. Mönnig, W. Schwieger, A. Tißler, T. Turek, Appl. Catal. 184 (1999) 249-256.

The catalyst powders were calcined in air for 6 h at 823 K, washed and dried overnight at 383 K. Extrusion to give cylindrical catalyst bodies (2 × 2 mm) followed after addition of appropriate binders.

The experiments were carried out in a flux apparatus operated at steady state with on-line analysis, the space velocity in each case being 10 000 h^{-1}.
The composition of the feed was:

1 000 ppm	NO_x
1 000 ppm	N_2O
0.5% vol	H_2O
2.5% vol	O_2
remainder	N_2

The following residual concentrations of NO_x and N_2O were obtained by varying the amount of NH_3 added:

Example	Amount of NH_3 added	Resultant NO_x concentration (after DeNOx stage at 350°C)	Resultant NO_x/N_2O ratio (after DeNOx stage)	Resultant N_2O concentration (after DeN_2O stage at 475°C)
1	500 ppm	500 ppm	0.5	40 ppm
2	800 ppm	200 ppm	0.2	54 ppm
3	950 ppm	50 ppm	0.05	81 ppm
4	990 ppm	10 ppm	0.01	99 ppm
5	1 000 ppm	< 1 ppm	< 0.001	462 ppm

As can be seen from the examples given, a high level of N_2O decomposition is possible up to an NO_x/N_2O ratio of 0.001, in particular 0.01. If the ratio sinks below this limit, the decomposition level becomes inadequate, since there is no longer sufficient cocatalytic function of NO_x.

Throughout this specification and the claims which follow, unless the context requires otherwise, the word "comprise", or variations such as "comprises" or "comprising", will be understood to imply the inclusion of a stated integer or step or group of integers or steps but not the exclusion of any other integer or step or group of integers or steps.

The reference to any prior art in this specification is not, and should not be taken as, an acknowledgment or any form of suggestion that the prior art forms part of the common general knowledge in Australia.

THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:

1. A process for reducing the NO_x concentration and N_2O concentration from the residual gas from nitric acid production, where the residual gas leaving the absorption column is passed, prior to entry into the residual gas turbine, through a combination of two stages, the first stage reducing the NO_x content by catalytic reduction, and the second stage reducing the N_2O content of the gas by decomposition into nitrogen and oxygen, and where the molar NO_x/N_2O ratio prior to entry of the gas into the second stage is in the range from 0.001 to 0.5, and in the second stage this gas is brought into contact with a catalyst which comprises one or more iron-loaded zeolites, the operating pressure in the second stage being from 4 to 12 bar.

2. The process as claimed in claim 1, characterized in that the iron-loaded zeolite(s) present in the catalyst are of MFI, BEA, FER, MOR and/or MEL type.

3. The process as claimed in claim 2, characterized in that the iron-loaded zeolite(s) are of MFI type.

4. The process as claimed in claim 3, characterized in that the zeolite is an Fe-ZSM-5.

5. The process as claimed in at least one of the preceding claims, characterized in that the temperature of the first stage is < 400°C, preferably < 350°C.

6. The process as claimed in at least one of the preceding claims, characterized in that the temperature of the second stage is in the range of 300 and 550°C, preferably in the range of 350 and 500°C.

7. The process as claimed in at least one of the preceding claims, characterized in that both stages are operated at a pressure in the range of from 4 to 12 bar.

8. The process as claimed in at least one of the preceding claims, characterized in that the first stage is operated using the SCR process.

9. The process as claimed in at least one of the preceding claims, characterized in that, after leaving the absorption column and prior to entry into the first or second stage, use is made of a residual gas whose water content is in the range from 0.05 to 1% by volume, in particular in the range from 0.1 to 0.8% by volume.

10. The process as claimed in at least one of the preceding claims, characterized in that, prior to entry into the second stage, use is made of a residual gas whose NO_x content is in the range from 1 to 200 ppm and whose N_2O content is in the range from 200 to 2 000 ppm.

11. A process for reducing the NO_x concentration and N_2O concentration from the residual gas from nitric acid production substantially as hereinbefore described with reference to the examples and the accompanying figures.

DATED THIS 31st day of March, 2004.

UHDE GMBH

By Its Patent Attorneys

DAVIES COLLISON CAVE

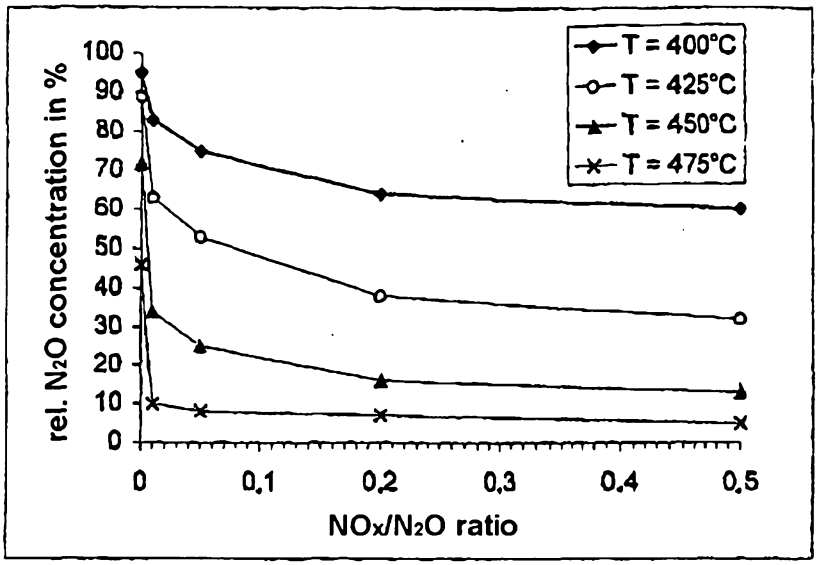

Figure 1

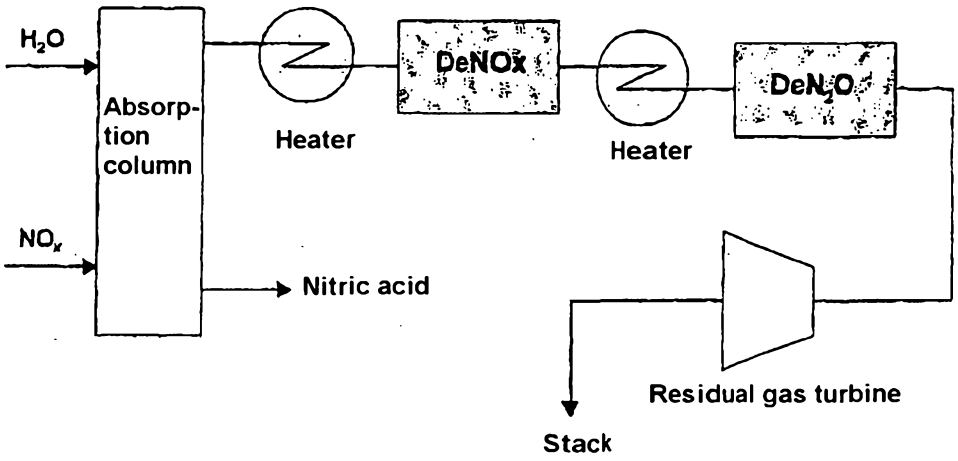

Figure 2