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**D E C I S I O N**  
**of 22 May 2000**

**Case Number:** T 0706/95 - 3.3.5

**Application Number:** 86906556.5

**Publication Number:** 0242394

**IPC:** C01B 21/00

**Language of the proceedings:** EN

**Title of invention:**

Reduction of nitrogen-based pollutants through the use of urea solutions containing oxygenated hydrocarbon solvents

**Patentee:**

FUEL TECH, INC.

**Opponent:**

Noell-KRC Umwelttechnik GmbH

**Headword:**

Reduction of nitrogen-based pollutants/FUELTECH

**Relevant legal provisions:**

EPC Art. 123(2), 54(1), 56

**Keyword:**

"Main and first auxiliary requests - novelty (no) - new use (no) - additional effect of a known process or use"

"Second, fifth and sixth auxiliary requests - amendment not allowable"

"Third and fourth auxiliary requests - obvious modifications"

**Decisions cited:**

G 0002/88

**Catchword:**

The discovery that the **same known means** lead to an additional

effect when they are used for the **same known purpose (i.e. known use)** of reducing the concentration of nitrogen oxides in the same effluent cannot confer novelty to this known use (see point 2.5).



Case Number: T 0706/95 - 3.3.5

**D E C I S I O N**  
**of the Technical Board of Appeal 3.3.5**  
**of 22 May 2000**

**Appellant:** FUEL TECH, INC.  
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**Respondent:** Noell-KRC Umwelttechnik GmbH  
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**Decision under appeal:** **Decision of the Opposition Division of the  
European Patent Office posted 6 June 1995  
revoking European patent No. 0 242 394 pursuant  
to Article 102(1) EPC.**

**Composition of the Board:**

**Chairman:** R. K. Spangenberg  
**Members:** M. M. Eberhard  
M. B. Günzel

## Summary of Facts and Submissions

- I. European patent No. 242 394 based on application No. 86 906 556.5 was granted on the basis of thirteen claims. Granted claim 1 reads as follows:

"1. A process for maintaining low ammonia concentrations, while reducing the concentration of nitrogen oxides in an oxygen-rich effluent from the combustion of a carbonaceous fuel, the process comprising:

injecting an aqueous solution of urea and an oxygenated hydrocarbon into said effluent at an effluent temperature above 1600°F (871°C), the concentration of the urea in solution and the size of the droplets in the dispersion being effective to achieve reduction of nitrogen oxide levels in the effluent and the concentration of oxygenated hydrocarbon in the dispersion being effective to reduce the level of free ammonia in the effluent."

- II. The respondent (opponent) filed a notice of opposition requesting revocation of the patent on the grounds of lack of novelty and lack of inventive step. In support of his arguments, the respondent relied *inter alia* on JP-A-82 365/1979 (D6) and a German translation thereof (D6a).
- III. The opposition division revoked the patent on the ground of lack of novelty. It held that the process of claim 1 as granted and the use as defined in claim 1 of the first auxiliary request submitted on 17 February 1995 both lacked novelty over the disclosure of D6. The use of the oxygenated hydrocarbon in the method of D6

at a temperature above 1600°F to reduce the level of free ammonia in the effluent was considered to form part of the information which the skilled person could inherently obtain from D6. Furthermore, the disclaimer introduced into each claim 1 of the second, third and fourth auxiliary requests submitted on 17 February 1995 did not render the claimed processes novel over the disclosure of D6.

IV. The appellant (proprietor of the patent) lodged an appeal against this decision. Two auxiliary requests were filed together with the statement of grounds of appeal on 16 October 1995 as well as an English translation of D6 (hereinafter D6b). The respondent informed the board by a letter dated 20 February 1998 that the opposition was withdrawn. On 2 December 1998 the appellant filed six auxiliary requests in replacement of the previous auxiliary requests. Claim 1 of the first auxiliary request reads as follows:

"1. The use of a process comprising:

injecting an aqueous solution of urea and an oxygenated hydrocarbon into said effluent at an effluent temperature above 1600°F (871°C), the concentration of the urea in solution and the size of the droplets in the dispersion being effective to achieve reduction of nitrogen oxide levels in the effluent and the concentration of oxygenated hydrocarbon in the dispersion being effective to reduce the level of free ammonia in the effluent

for maintaining low ammonia concentrations, while reducing the concentration of nitrogen oxides in an oxygen-rich effluent form (sic) the combustion of a carbonaceous fuel."

Claim 1 of the second auxiliary request differs from claim 1 of the main request (granted claim 1) only by the incorporation of the phrase "**and also minimizing carbon-based pollutants**" at the beginning of the claim so that it is directed to "a process for maintaining low ammonia concentrations **and also minimizing carbon-based pollutants**, while reducing the concentration of nitrogen oxides ...".

Claim 1 of the third auxiliary request differs from claim 1 of the main request in that the sentence "**wherein the weight ratio of oxygenated hydrocarbon to urea is within the range of 1:2 to 2:1**" has been added at the end of the claim.

Claim 1 of the fourth auxiliary request differs from claim 1 of the main request by the following additional features incorporated at the end of the claim: "**wherein the droplets within the dispersion are uniformly within the range of from about 10 to 10.000  $\mu\text{m}$  in diameter.**"

Claim 1 of the fifth auxiliary request reads as follows:

"1. The use of a process comprising:

injecting an aqueous solution of urea and an oxygenated hydrocarbon into said effluent at an effluent temperature above 1600°F (871°C), the concentration of the urea in solution and the size of the droplets in the dispersion being effective to achieve reduction of nitrogen oxide levels in the effluent and the concentration of oxygenated hydrocarbon in the dispersion being effective to reduce the level of free ammonia in the effluent, wherein the weight ratio of

oxygenated hydrocarbon to urea is within the range of from 1:2 to 2:1

for maintaining low ammonia concentrations and minimizing carbon-based pollutants, while reducing the concentration of nitrogen oxides in an oxygen-rich effluent form (sic) the combustion of a carbonaceous fuel."

Claim 1 of the sixth auxiliary request differs from claim 1 of the fifth auxiliary request in that the sentence "**wherein the droplets within the dispersion are uniformly within the range of from about 10 to 10.000 µm in diameter and**" has been incorporated after the phrase "being effective to reduce the level of free ammonia in the effluent".

In a communication pursuant to Article 11(2) RPBA, the appellant was informed of the preliminary opinion of the board about the seven requests on file. The appellant's attention was drawn in particular to specific points to be discussed at the oral proceedings. The appellant did not present comments on the reasons given in the communication. Oral proceedings were held on 22 May 2000 in the absence of the appellant and of the previous respondent. The appellant had informed the board on 18 May 2000 that he would not attend the oral proceedings.

V. The appellant's arguments can be summarised as follows:

Regarding claim 1 of the main request, the appellant argued that D6/D6b was totally silent about the possibility of maintaining low ammonia concentrations. Rather, following the equations on page 7 of D6, especially equation (5), it would seem that the amount

of ammonia was increased and not decreased in the process of D6. Furthermore, D6 was also silent about the droplet size of the dispersion. The concentrations of urea and oxygenated hydrocarbon used in the examples of D6 did not fall within the concentrations defined in claim 1. According to claim 1 urea had to be present in a concentration effective to achieve the reduction of the **total** nitrogen oxide levels and the concentration of the oxygenated hydrocarbon had to be effective to reduce the level of free ammonia in the effluent. Urea and the oxygenated hydrocarbon were both used in a concentration of 35% in the example of the patent in suit, whereas the concentrations of urea and ethanol in D6 were 4.2 wt.% and 0.6 wt.% respectively. Furthermore, D6, contrary to the patent in suit, did not teach a skilled person to introduce urea for reducing the **total** amount of NO<sub>x</sub>, but rather taught using urea mainly for transforming NO to NO<sub>2</sub>. Thus, the concentration of urea used in D6 did not necessarily fall within claim 1. Claim 1 was clearly novel over D6 in accordance with decision G 2/88 (OJ EPO, 1990, 93). In this decision the alleged "doctrine of inherence" had not been accepted. The skilled person would not have derived from D6 the technical teaching underlying the patent in suit, namely to provide a process for maintaining low ammonia concentrations, while reducing the concentration of nitrogen oxides in an oxygen-rich effluent from the combustion of a carbonaceous fuel. D6 was completely silent on the reduction of ammonia in the effluent and thus this feature represented a "hidden technical feature" which had not been available to the public. Furthermore, the technical teaching of the patent in suit was very different from that of D6. In D6 the alcohol was used in order to oxidise NO to NO<sub>2</sub> and permit removal of the latter by scrubbing. D6 was



completely silent about using an alcohol in a selective-non-catalytic reduction. D6 taught that in each and every instance the oxidation step was followed by a wet scrubbing step. The primary technical teaching of D6 was to provide an alternative manner of oxidising NO.

Claim 1 of the first auxiliary request was admissible following decision G 2/88 and the same arguments applied to this request. As D6 did not disclose the exemplified process to be useful for maintaining low ammonia concentrations while reducing the concentration of nitrogen oxides in an oxygen-rich effluent from the combustion of carbonaceous fuel, the subject-matter of claim 1 of this request was new with respect to D6.

Novelty was even more given for each claim 1 of the auxiliary requests 2 to 6 since nothing was mentioned in D6 about minimizing carbon-based pollutants, about the ratio oxygenated hydrocarbon/urea or the droplet size. The means of realisation were different in terms of concentration, droplet size, temperature range for effective operation and the specific combination of all three. D6 neither described the importance of these features nor enabled their selection for any process other than for use with wet scrubbing as a final step to reduce NO<sub>2</sub>.

- VI. Before withdrawing the opposition, the previous respondent had contested the appellant's arguments put forward in the statement of grounds of appeal.
  
- VII. The appellant requested in his letter dated 1 February 2000 that the decision of the opposition division be set aside and that the patent be maintained on the

basis of the granted claims, as the main request, or as auxiliary requests, on the basis of one of the auxiliary requests 1 to 6 filed on 2 December 1998, in consecutive order. The appellant further requested on 18 May 2000 that a decision be taken on the basis of the requests, documents and written submissions presently on file.

### **Reasons for the Decision**

1. The appeal is admissible.

#### *Main request (granted claims)*

2. The question arises whether or not the process according to claim 1 is novel over the disclosure of D6/D6b.
  - 2.1 D6/D6b discloses a process for reducing the concentration of nitrogen oxides in an oxygen-rich effluent such as an oxygen-rich exhaust gas from a boiler. It describes experiments in which an aqueous solution of urea (4.2 wt.% urea), an aqueous solution of ethanol (0.6 wt.%) or a mixture thereof was injected into the effluent at different temperatures within the range from 500 to 1000°C to cause a gas phase reduction. The weight ratio of urea to ethanol in the mixture was 2.5:1. With the mixture of urea and ethanol the gas phase reduction started at 500°C and became more active when increasing the temperatures up to 1000°C. Figure 2 shows the ratio of nitrogen transformation at temperatures of 500, 600, 700, 800, 900 and 1000°C. At temperatures of 900°C or 1000°C this ratio is well above 60%. NO<sub>2</sub> was formed in addition to

nitrogen because of the presence of oxygen. D6/D6b teaches that this gas phase reduction leads to a maximum denitrification ratio of about 80%, this limitation resulting from the formation of NO<sub>2</sub> in the presence of oxygen. The said ratio can be improved by reducing and decomposing the formed NO<sub>2</sub> with an aqueous solution of urea or by absorbing it with alkaline adsorbents (see D6b, page 6, line 9 to page 7 line 9; page 8, example; whole page 9; page 10, first and second paragraph; Figures 1 and 2).

2.2 The concentration of urea in the mixture falls within the ranges indicated in dependent claim 13 of the patent in suit. As a reduction of the nitrogen oxide level was obtained in the experiments of D6/D6b which were carried out at a temperature of 900°C or higher (see Figure 2; page 9, last paragraph; page 10, second paragraph), it must be assumed that the size of the droplets fulfilled the conditions stated in claim 1, ie a size "effective to achieve reduction of the nitrogen oxide levels in the effluent".

2.3 The appellant argued in the statement of grounds of appeal that the concentrations of urea and oxygenated hydrocarbon used in the examples of D6 were different from those of the claimed process. He supported this argument in particular by referring to the example of the patent in suit where urea and the oxygenated hydrocarbon are both used in a concentration of 35%. These arguments cannot be accepted by the board. As concentrations of 35% are not stated in claim 1, the latter is not limited to concentrations of urea and oxygenated hydrocarbon of about 35%. Furthermore, the urea concentration and the urea/ethanol ratio disclosed in D6 lie within the ranges stated in dependent

claims 13, 4 and 5 of the patent in suit.

- 2.4 The appellant further argued that D6 did not teach introducing urea for the purpose of reducing the total amount of  $\text{NO}_x$  but rather indicated using urea mainly for transforming  $\text{NO}$  to  $\text{NO}_2$  and that D6 was silent about using an alcohol in a selective non-catalytic reduction. In the appellant's view the primary technical teaching of D6 was to provide an alternative manner of oxidising  $\text{NO}$ . These arguments are not convincing for the following reasons. According to claim 1 of D6/D6b a nitrogen oxide-containing gas is brought into contact with urea and an alcohol in the presence of oxygen to cause a gas phase reaction in which a part of the nitrogen oxide is transformed into nitrogen gas and a greater part of the remaining nitrogen oxide is transformed into nitrogen dioxide. However, the disclosure of D6 is not limited to the subject-matter as defined in the claims. It is further disclosed in D6 that the gas phase reduction of nitrogen oxide into  $\text{N}_2$  leads to a maximum denitrification of **about 80%** at the higher temperatures as already indicated above. It is directly and unambiguously derivable from Figure 2 of D6 and the corresponding explanations in the description that the injection of urea or of the mixture of urea and alcohol into the effluent at a temperature of for example  $900^\circ\text{C}$  or higher in the presence of oxygen **leads to the reduction of a major part (up to 80%) of the nitrogen oxide to nitrogen** and not mainly to the oxidation of  $\text{NO}$  into  $\text{NO}_2$ . The fact that a minor part of the nitrogen oxide is transformed into  $\text{NO}_2$ , which may be further reduced or decomposed with an aqueous solution of urea or absorbed with alkaline adsorbents, is not relevant to the novelty issue since, on the one hand, the

claimed process does not exclude a multi-stage treatment and, on the other hand, it is not stated in claim 1 to which extent the nitrogen oxide level in the effluent is reduced.

- 2.5 It is not indicated in D6/D6b that low ammonia concentrations are maintained in the effluent or that the concentration of ethanol in the dispersion is effective to reduce the level of free ammonia in the effluent. However, it is observed that both the ethanol/urea ratio and the urea concentration used in the experiments of D6 fall within the ranges defined in dependent claims 4, 5 and 13 of the patent in suit. As the process for reducing the concentration of nitrogen oxides at a temperature of 900°C disclosed in D6 is applied to the same effluent as in the patent in suit and is performed using a temperature and urea and ethanol concentrations which all lie within the ranges defined in claims 1, 4, 5 and 13 of the patent in suit, it must be inferred therefrom that the ethanol concentration used in D6 also causes a reduction of the level of free ammonia in the effluent. The appellant's argument that in view of equation (5) on page 7 of D6, it would seem that the amount of ammonia was increased and not decreased in D6 is not convincing. As pointed out by the respondent, it was known to the skilled person before the priority date that ammonia and NO are converted to N<sub>2</sub> and water in the presence of oxygen especially in a temperature region of about 1600°F (871°C). This was not contested by the appellant. In these circumstances, the board is not convinced that a skilled person would have derived from D6 that the ammonia concentration in the effluent might be increased when the process is carried out at a temperature of 900°C.

The appellant further argued that the process of claim 1 was clearly novel over D6 in accordance with decision G 2/88 (OJ EPO, 1990, 93) since D6 did not disclose the reduction of the free ammonia level in the effluent and this feature represented therefore a "hidden technical feature" which had not been made available to the public. The board cannot agree with these arguments for the following reasons. As already pointed out above, it is not stated in D6 that the alcohol reduces the level of free ammonia in the effluent. However, the claimed subject-matter involves the **same use or purpose** as in D6, ie the reduction of the concentration of nitrogen oxides in an oxygen-rich effluent from the combustion of a carbonaceous fuel, and the **same means of realisation** as in D6, ie the injection of an aqueous solution of urea and an oxygenated hydrocarbon into the effluent at the same temperatures and in the same concentrations. The effluent in which the urea and oxygenated hydrocarbon are injected is also the same as in D6. Thus, in the present case, not only the means of realisation but also the use or purpose are the same as in the prior art document. The present case is therefore not comparable to the situation considered in G 2/88 where the claim was directed to a **new use** of a known compound, based on a newly discovered technical effect. The discovery that the **same known means** lead to an additional effect, ie the reduction of the level of free ammonia in the effluent, when they are used for the **same known purpose (ie known use)** of reducing the concentration of nitrogen oxides in the same oxygen-rich effluent from the combustion of a carbonaceous fuel cannot confer novelty to this known use since both the said means of realisation and the said use or purpose remain the same.

2.6 It follows from the above that the subject-matter of claim 1 according to the main request lacks novelty over the disclosure of D6. As claim 1 of the main request does not meet the requirement of novelty set out in Articles 52(1) and 54 EPC, the main request is not allowed.

*First auxiliary request*

3. Claim 1 of this request is not correctly formulated since reference is made to "said effluent" in the third line although an effluent was not previously mentioned. However, as reference is made in the last two lines of this claim to "an oxygen-rich effluent form (sic) the combustion of a carbonaceous fuel", the board considers that the claim can be understood as meaning that the "said effluent" referred to at the beginning of the claim is in fact the effluent as defined at the end of the claim.

3.1 Claim 1 of this request is directed to the use of a process comprising specific process features for maintaining low ammonia concentrations, while reducing the concentration of nitrogen oxides in an oxygen-rich effluent from the combustion of a carbonaceous fuel. The said specific features are the same as those stated in claim 1 of the main request. The effluent and the purpose or use are also the same as those defined in claim 1 of the main request. Therefore, this claim does not differ in its substance from claim 1 of the main request. It follows that the reasons given above in points 2.1 to 2.5 apply likewise to claim 1 of the first auxiliary request and that this claim is also considered to lack novelty over the disclosure of D6.

*Second auxiliary request*

4. Claim 1 of this request differs from claim 1 of the main request by the incorporation of the features "**and minimizing carbon-based pollutants**" into the claim. The board expressed doubts in the communication dated 9 February 2000 regarding the allowability of this amendment. The patent in suit and the original application disclose that there is a "need for a process which enables the reduction of nitrogen-based pollutants by operating under efficient oxygen-rich conditions which minimize carbon-based pollutants, and yet permits the control of ammonia levels in the final effluent" (see respectively page 2, line 57 to 58 and page 4, lines 16 to 21). Therefore, according to the description oxygen-rich conditions minimize carbon-based pollutants. However, it is not clear to the board for which reasons it would be directly and unambiguously derivable from this statement that the injection of an aqueous solution of urea and oxygenated hydrocarbon into an oxygen-rich effluent from the combustion of a carbonaceous fuel, at temperatures above 1600°F also minimizes carbon-based pollutants. The application as filed contains no additional information from which it might be directly derived that the said injection of urea and oxygenated hydrocarbon enables not only reducing the concentration of nitrogen oxides and the level of free ammonia in the effluent but also, at the same time, minimizing carbon-based pollutants. The appellant himself did not indicate on which part of the original description the claimed amendment was based (see point 3 of the communication dated 9 February 2000). Furthermore, he did not present any comment in reply to the doubts expressed in the said communication. In these



circumstances, the board is still not convinced that the amendment in claim 1 of the second auxiliary request meets the requirements of Article 123(2) EPC. Therefore this request cannot be granted.

*Third auxiliary request*

5. The amendments in claim 1 of this request meet the requirements of Articles 123(2) and (3) EPC. The subject-matter of claim 1 is new over the disclosure of D6 since the latter discloses an ethanol/urea weight ratio of 1:2.5 (ie 0.4), and thus a ratio which lies outside the claimed range of 1:2 to 2:1 (ie 0.5 to 2).
  
6. Concerning the issue of inventive step, the board considers in agreement with the appellant that D6/D6b represents the closest prior art. Neither the patent in suit nor the file contains evidence showing that an improvement is achieved by using a weight ratio of oxygenated hydrocarbon/urea of 1:2 to 2:1 instead of 1:2.5 in D6/D6b. In these circumstances, the problem to be solved with respect to D6 can be seen in the provision of another process for reducing the concentration of NO<sub>x</sub> in oxygen-rich effluents from the combustion of a carbonaceous fuel while maintaining low ammonia concentrations.

It is proposed to solve this problem by a process as defined in claim 1 of the third auxiliary request, ie by a process which differs from D6/D6b only in that the weight ratio of oxygenated hydrocarbon/urea is within the range 0.5 to 2 instead of 0.4. In view of the data in the patent in suit, it is credible that this problem has actually been solved by the claimed process.

The question whether or not it was obvious to the skilled person to increase the ratio of 1:2.5 disclosed in D6 in view of the teaching of this document has to be answered positively. D6 teaches that a weight ratio ethanol/urea of 1:2.5 was used in the experiments carried out at temperatures from 500 to 1000°C. It can be inferred from D6 that this value represents only an example and not the upper limit of the appropriate range for the ethanol/urea ratio. Therefore, the skilled person reading D6 would have expected that values of the ethanol/urea ratio which are either somewhat smaller or higher than the exemplified value of 1:2.5 would also be suitable for the gas phase reduction of the nitrogen oxides into nitrogen at the disclosed temperature of for example 900°C. In these circumstances and further taking into account that the lower limit of 1:2 stated in claim 1 is not far removed from the value of 1:2.5 given as an example in D6, it is considered that the choice of the oxygenated hydrocarbon/urea ratio stated in claim 1 was an obvious solution of the above stated technical problem in view of the teaching of D6. It follows that the process according to claim 1 of the third auxiliary request does not involve an inventive step. As claim 1 does not meet the requirements of Articles 52(1) and 56 EPC, this request must also fail.

*Fourth auxiliary request*

7. Claim 1 of this request meets the requirements of Article 123(2) and (3). It is assumed to the appellant's benefit that the process according to claim 1 of this request is new with respect to D6 taking into account that D6 does not expressly mention that "the droplets within the dispersion are uniformly

within the range of from about 10 to 10.000  $\mu\text{m}$  in diameter".

8. As the appellant has provided no evidence that the choice of a droplet size of 10 to 10 000  $\mu\text{m}$  is critical or results in an improvement over other possible droplet sizes, the technical problem with respect to the closest prior art D6 is the same as that stated above in connection with claim 1 of the third auxiliary request. It is also credible that this problem has been solved by the claimed process. However, it is self-evident that a uniform mixing of the urea solution with the effluent is desirable in order to achieve an efficient gas phase reduction. As pointed out by the respondent and not contested by the appellant, it formed part of the general knowledge before the priority date that the droplet size is important to enable uniform mixing of the effluent gases with the reactant, ie urea. In these circumstances, the board considers that in view of D6 it would have lain within the competence of the skilled person to determine, by routine experiments, the appropriate range of suitable droplet sizes in relation to the chosen temperatures in order to obtain an efficient reduction of the nitrogen oxide in the effluent. Therefore, the process according to claim 1 of this request lacks an inventive step and the fourth auxiliary request is not allowable.

*Fifth and sixth auxiliary requests*

9. Claim 1 of the fifth auxiliary request and claim 1 of the sixth auxiliary request both include the amendment "**and minimizing carbon-based pollutants**", which has been examined above in connection with the amendments in claim 1 of the second auxiliary request (see

point 4). Therefore, the considerations in point 4 above apply likewise to each claim 1 of the fifth and sixth auxiliary requests. It follows that these claims are considered to contravene the provisions of Article 123(2) EPC. Therefore, the fifth and sixth auxiliary requests must also fail.

**Order**

**For these reasons it is decided that:**

The appeal is dismissed.

The Registrar:

The Chairman:

S. Hue

R. Spangenberg