D G M K Research Report 616-1

AdBlue as a Reducing Agent for the Decrease of NO_x Emissions from Diesel Engines of Commercial Vehicles



Deutsche Wissenschaftliche Gesellschaft für Erdöl, Erdgas und Kohle e.V.

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German Society for Petroleum and Coal Science and Technology

DGMK-Research Report 616-1

AdBlue as a Reducing Agent for the Decrease of NO_x Emissions from Diesel Engines of Commercial Vehicles

Abstract:

The Parliament of the European Union has issued stricter emission standards (Euro 4 and 5) for diesel engines of HD (heavy duty) commercial vehicles for the years 2005 and 2008.

After comparing and testing various technical options the European manufacturers of HD commercial vehicles have agreed on one method, the *"selective catalytic reduction"* (SCR) and on one reducing agent, *AdBlue*, an aqueous solution of urea.

The European automobile industry plans to successively equip HD vehicles with the SCR technology. The automobile industry assumes that mineral oil industry and trade will make available the reducing agent AdBlue via a separate tanking system at their public service stations – eventually in a balanced distribution throughout Europe.

This report describes the activities of national and international committees engaged in the introduction of the SCR/urea technology. It also outlines questions as yet unresolved with respect to the new technical and logistic requirements.

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Deutsche Wissenschaftliche Gesellschaft für Erdöl, Erdgas und Kohle e.V.

DGMK-Forschungsbericht 616-1

AdBlue als Reduktionsmittel für die Absenkung der NO_x-Emissionen aus Nutzfahrzeugen mit Dieselmotor

Kurzfassung:

Das Parlament der Europäischen Union hat für die Jahre 2005 und 2008 verschärfte Abgasnormen für Dieselmotoren von schweren Nutzfahrzeugen (EURO 4 und EURO 5) verabschiedet.

Nach Vergleich und Erprobung verschiedener technischer Möglichkeiten haben sich die europäischen Hersteller von schweren Nutzfahrzeugen auf ein Verfahren, die "selektive katalytische Reduktion" (SCR) und ein Reduktionsmittel "wässrige Harnstofflösung" (AdBlue) geeinigt.

Die europäische Automobilindustrie plant, ab dem Jahr 2004 schwere Nutzfahrzeuge mit der SCR-Technik sukzessive auszurüsten.

Die Automobilindustrie geht davon aus, dass die Mineralölwirtschaft das Reduktionsmittel AdBlue über ein separates Betankungssystem an ihren öffentlichen Tankstellen schließlich flächendeckend zur Verfügung stellt.

Der Bericht schildert die Aktivitäten nationaler und internationaler Gremien, die sich mit der Einführung der SCR/Harnstoff-Technik befassen. Er beschreibt die noch ungeklärten Fragen im Hinblick auf die neuen technischen und logistischen Anforderungen.

Die Studie berücksichtigt die vorliegenden Informationen mit Stand bis Juli 2003.

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0 Summary

The Parliament of the European Union has issued stricter emission standards (Euro 4 and 5) for HD diesel engines for the years 2005 and 2008. Compared to current limits an emissions reduction of 60 % for nitrogen oxides and of 80 % for particulate matter will be required.

After comparing and testing various technical options the European manufacturers of HD commercial vehicles have agreed on **one** method for meeting these standards: the *"selective catalytic reduction"*(SCR technology), which converts nitrogen oxides into innocuous nitrogen and water in the presence of a reducing agent. The European automobile industry has also agreed on **one** reducing agent: an aqueous solution of urea, which shall be distributed under the trade name *"AdBlue"*.

The main reason for this decision of the HD commercial vehicle industry was the fact that the SCR/urea technology appears to be the only method which enables the necessary decrease of exhaust emissions while maintaining engine optimization with regard to performance and fuel economy. Furthermore this method can be applied to all engines.

The present report describes the production, properties and environmental impact of urea as well as of its aqueous solution. Since the solution is corrosive, certain materials are required for its handling. Product quality and properties of AdBlue are defined in the DIN standard 70070, which should form the basis for a later EN or ISO standard.

According to a survey of the Association of the Automobile Industry (VDA), the annual demand for AdBlue for the use in HD commercial vehicles is estimated to increase from initially 500 t to over 2.5 million t in the year 2012. The cost of AdBlue is mainly determined by the costs of energy and raw materials in the production of urea, also by the costs for distribution; the latter depend on the means of distribution and the investments probably required at the service stations.

The present report describes the activities of national and international committees working on the introduction of the SCR/urea technology. Beginning in the year 2004, the European automobile industry plans to equip their HD trucks with the SCR converters. It is proposed that the supply of these vehicles with AdBlue will be accomplished in three phases: initially by filling stations of larger fleet owners, followed by filling stations along magistrale routes in Europe and finally also by public filling stations in a balanced distribution in Europe. The automobile industry assumes that AdBlue will be distributed through a separate tanking system.

Mineral oil industry and trade will have to decide whether they will offer the reducing agent required for the SCR/urea technology, at their service stations, and whether they will invest in the necessary infrastructure of additional tanks, piping, pumps and filling nozzles, or whether they prefer to supply AdBlue in mobile containers, from which the truck drivers can fill up their AdBlue tanks.

This report lists a number of as yet unresolved issues in the area of technical and logistic requirements and the field tests initiated by DGMK for their resolution. The results of these tests will be described in the follow-up report 616-2.

Zusammenfassung

Das Parlament der Europäischen Union hat für die Jahre 2005 und 2008 verschärfte Abgasnormen für Dieselmotoren von schweren Nutzfahrzeugen verabschiedet, die für die Emission von Stickoxiden eine Reduktion um 60 % und die von Partikeln um 80 % gegenüber den zur Zeit geltenden Grenzwerten verlangen (EURO 4 und EURO 5).

Nach Vergleich und Erprobung verschiedener technischer Möglichkeiten haben sich die europäischen Hersteller der schweren Nutzfahrzeuge auf **ein** Verfahren zur Einhaltung dieser Grenzwerte geeinigt: die "selektive katalytische Reduktion" (SCR-Verfahren), bei der die Stickoxide in Gegenwart eines Reduktionsmittels in unschädlichen Stickstoff und Wasserdampf umgesetzt werden. Die europäische Automobilindustrie hat sich darauf verständigt, einheitlich eine wässrige Lösung von Harnstoff als Reduktionsmittel einzusetzen, die unter der Bezeichnung "AdBlue" vertrieben werden soll.

Maßgeblich für diesen Beschluss der Nutzfahrzeughersteller ist die Tatsache, dass die SCR/Harnstoff-Technik das einzige Verfahren darstellt, welches die erforderliche Absenkung der Abgasemissionen bei gleichzeitiger Optimierung des Motors hinsichtlich Leistungsverhalten und Kraftstoffverbrauch ermöglicht. Außerdem ist dieses Verfahren bei allen Motoren anwendbar.

Im vorliegenden Bericht werden Herstellung, Eigenschaften und Umweltverhalten sowohl des Harnstoffs als auch seiner wässrigen Lösung beschrieben. Die Lösung ist korrosiv, weshalb bei ihrer Handhabung bestimmte Materialien verwendet werden müssen. Produktqualität und Eigenschaften von AdBlue sind in der Norm DIN 70070 definiert, die die Grundlage für einen späteren EN- oder ISO-Standard bilden soll.

Nach einer Umfrage des Verbandes der Automobilindustrie wird der jährliche Bedarf an AdBlue für den Einsatz in schweren Nutzfahrzeugen von über 2,5 Mio t im Jahre 2012 prognostiziert. Faktoren, die die Kosten von AdBlue bestimmen, sind vor allem die Energie- und Rohstoffkosten zur Herstellung des Harnstoffs sowie die Distributionskosten für AdBlue, die wiederum durch die Art der Verteilung und die damit eventuell verbundenen Investitionen an der Tankstelle beeinflusst werden.

Der vorliegende Bericht schildert die Aktivitäten nationaler und internationaler Gremien, die sich mit der Einführung der SCR/Harnstoff-Technik befassen. Berücksichtigt wurden die Informationen bis Juli 2003.

Die europäische Automobilindustrie plant, ab dem Jahr 2004 Lastkraftwagen über 3,5 t mit dem SCR-Katalysator auszurüsten. Nach ihren Vorstellungen soll die Versorgung dieser Fahrzeuge mit AdBlue in drei Phasen erfolgen: anfangs über die Betriebstankstellen größerer Fahrzeugflotten, dann auch über Autobahn-Knotenpunkte und schließlich europaweit und flächendeckend auch über das öffentliche Tankstellennetz der Mineralölkonzerne und -händler. Sie geht dabei davon aus, dass AdBlue über ein separates Betankungssystem zur Verfügung gestellt wird. Mineralölindustrie und -handel müssen entscheiden, ob sie das für die SCR/Harnstoff-Technik benötigte Reduktionsmittel an ihren Tankstellen anbieten und die dafür erforderlichen Investitionen für zusätzliche Tanks, Leitungssysteme und Zapfvorrichtungen aufbringen wollen, oder ob sie es bevorzugen, AdBlue in ortsbeweglichen Behältern vorzuhalten, aus denen die LKW-Fahrer dann ihre AdBlue-Tanks auffüllen können.

Der Bericht schildert die noch ungeklärten Fragen im Hinblick auf technische und logistische Anforderungen und die zu ihrer Klärung von der DGMK initierten Feldtests, deren Ergebnisse in einem Folgebericht 616-2 beschrieben werden sollen.

1 Fundamentals of the after-treatment of exhaust gases from diesel engines of commercial vehicles

1.1 Euro standards for the reduction of exhaust gas emissions

By the end of 1999 the Parliament of the European Union has issued stricter emission standards for diesel engines of heavy duty (HD) commercial vehicles [1]: According to the Euro 4 standard, which will become effective in October 2005, emissions of nitrogen oxides (NO_x) will have to be reduced by 30 %, emissions of particulate matter (PM) by 80 % vis-à-vis the current limits of Euro 3. In a second step (Euro 5), these values will have to be decreased further in 2008. In addition, exhaust gas limits for carbon monoxide (CO) and unburnt hydrocarbons (HC), which are already very stringent, will have to be further reduced according to Euro 4 (see table 1).

| Pollutant | Euro 3 | Euro 4 | Euro 5 |
|------------------------------------|-----------|-----------|-----------|
| | from 2001 | from 2005 | from 2008 |
| Nitrogen oxides (NO _x) | 5,0 | 3,5 | 2,0 |
| Carbon monoxide (CO) | 2,0 | 1,5 | 1,5 |
| Unburnt hydrocarbons (HC) | 0,66 | 0,46 | 0,46 |
| Particulate matter (PM) | 0,1 | 0,02 | 0,02 |

Tab. 1: Exhaust gas limits of the EU standard 99/96/EG for HD commercial vehicles (> 3,5 t; > 85 kW) in g/kWh

The background for this tightening of exhaust gas limits is the EU guideline for air quality, which aims at further reduction of air pollution by industrial plants and motor vehicles. In 1999, road traffic was the main contributor of nitrogen oxide emissions with 51 %, whereas industrial furnaces as well as public power and heating plants contributed 28 %, only [2]. They form nitric acid in the atmosphere, which causes the so-called acid rain in combination with sulfuric acid. Prolonged exposure to higher concentrations of nitrogen oxides causes irritations of the respiratory system in humans. They are also precursors for the formation of photo oxidants, e.g. ozone.

Particulate matter contains polycyclic aromatic hydrocarbons (PAH), some of which are suspected to be carcinogenic.

Therefore, the new limits of Euro 4 and 5 for exhaust gas emissions shall reduce the emission of pollutants.

1.2 Reduction of exhaust gas emissions

There are several different technical solutions to the problem of reducing exhaust gas emissions. The German Association of the Automobil Industry (VDA) describes the following options in its annual report of 2002:

- Internal measures to the engine: shape of the cylinder, mixing of the fuel/ignition, valve timing
- Catalytic converter for the reduction of CO, HC and NO_x emissions while operating the gasoline engine in a stoichiometric proportion of $\lambda = 1$
- Converter plus operation of the engine with excess air ($\lambda > 1$) for reduction of NO_x emissions
- Oxidation catalyst for decreasing CO, HC and also particulate matter emissions to a certain extent
- Exhaust gas recycling (EGR) for the reduction of NO_x emissions
- Storage catalyst with separate control for reduction of NO_x emissions
- Selective catalytic reduction (SCR) to decrease NO_x emissions (with an additive as a reducing agent)
- Particle filter with continuous or discontinuous regeneration (with or without additive as a regeneration aid)

The above methods can also be used in various combinations. According to the VDA, considerable development work is still required for some of these techniques (storage catalyst, SCR and particle filter) [3].

However, there is a dilemma when trying to reduce particle and NO_x emissions simultaneously, because there is a correlation between the formation of NO_x on one hand and of the remaining pollutants on the other: It is possible to reduce NO_x emissions by internal measures, such as EGR, which lowers the temperature in the cylinder of the engine. However, emissions of particles, unburnt hydrocarbons and carbon monoxide would then increase. In addition, efficiency and effectiveness of the diesel engine would be impaired and therefore fuel consumption and carbon dioxide emissions will increase. If, however, the combustion in the engine is optimized with regard to efficiency and performance, then formation of particulate matter and NO_x will increase to such an extent that the imminent Euro standards cannot be met.

1.3 The SCR method

The above mentioned SCR technique offers a solution of this dilemma. If the engine operates under conditions which optimize the combustion with regard to efficiency, then SCR will convert NO_x emissions into innocuous substances by means of a reducing agent on a ceramic catalyst [4]. Ammonia is a suitable agent for this purpose. The chemical reaction in the exhaust gas stream behind the engine at the ceramic catalyst causes a reduction of the nitrogen oxides NO₂ and NO by ammonia (NH₃), according to the following equations:

 $6 \text{ NO}_2 + 8 \text{ NH}_3 \rightarrow 7 \text{ N}_2 + 12 \text{ H}_2\text{O}$ 2 NO + O₂ → 2 NO₂ NO + NO₂ + 2 NH₃ → 2 N₂ + 3 H₂O

It follows from these equations, that nitrogen dioxide (NO₂) reacts directly with ammonia (NH₃), whereas initially nitrogen monoxide (NO) is partly oxidized to NO₂ (overall the speed of the NO_x reduction is fastest, when NO and NO₂ are present at a ratio of 1:1). Other than water (vapour), this reaction produces only elementary nitrogen (N₂), which with 80 % constitutes the main natural component of the atmosphere.

1.4 Urea as a reduction agent

Ammonia is a very aggressive and noxious gas. According to the opinion of the "Federal Ministry for Environment, Nature Conservation and Safety of Nuclear Power", the use of ammonia in public transportation would require the establishment of a very extensive system of regulations encompassing e.g. environmental protection plus work and traffic safety [5]. Therefore it is preferable to use less hazardous chemical compounds instead of ammonia, which release ammonia in the exhaust stream only. One such substance is urea, which will form ammonia at elevated temperature in the presence of vapour, according to the following equation:

 $(NH_2)_2CO + H_2O \rightarrow CO_2 + 2 NH_3$

However, urea is not applied as a pure substance, because it forms is a solid white powder, which cannot easily be added to the exhaust gases in exact quantities. Therefore an aqueous solution of urea should be used, which will be described in more detail in the next chapter.

The technology of selective catalytic reduction with urea as a reducing agent has stood the test for denitrification of exhaust gases in stationary units, such as power plants and waste incineration facilities. For its application in mobile systems, like vehicles, some further development is needed – as mentioned above.

It should be mentioned at this point that basically other chemical compounds, which release ammonia, can be used in the SCR technology. Ammonium carbamate is one example; its suitability was investigated in a research project of the Institute for Chemical Technology at the University of Karlsruhe [6]. In addition, other reducing agents can be also used for the conversion of nitrogen oxides into innoxious nitrogen, such as natural gas or higher hydrocarbons (e.g. gasoline).

1.5 Combined system of exhaust gas treatment on the basis of the SCR method

In order to optimize the emissions performance of a diesel engine, the following devices can be combined: the exhaust gases flow through an oxidation catalyst behind the engine, in which NO is partially converted into NO_2 , and unburnt hydrocarbons are oxidized to CO_2 and water. Subsequently, after addition of urea solution and its hydrolysis to NH_3 and CO_2 , the mixture of NO and NO_2 is transformed to innoxious nitrogen and water. The following figure 1 shows a scheme of such a combined system for exhaust gas treatment.



Figure 1: Scheme of a combined system for exhaust gas treatment on the basis of the SCR method [7]

The "VDI nachrichten" reported in September 2002 that the SCR method has proven its basic suitability in a field test sponsored by the Bavarian State Ministry for Development and Environmental Protection. In this project ten commercial trucks of Mercedes-Benz equipped with SCR systems have been successfully tested over a total of 3.2 million kilometers [8]. Other automobile manufacturers, such as MAN in Nuremberg, Renault in Lyon, Volvo Power Train Corp. in Gothenburg and Mack Truck Inc. in Hagerstown, USA, have investigated the suitability of the SCR method with urea solution as a reducing agent.

2 Urea/AdBlue: production and properties

2.1 Historical excursion

The chemical compound, urea, has played a very special role in the history of natural sciences. In 1828 the chemist Friedrich Wöhler succeeded in converting the inorganic compound ammonium cyanate into the organic substance urea. This reaction defeated the then known theory of "vis vitalis", which meant that a vital force is needed for the generation of organic matter. Wöhler's friend Justus Liebig rightly characterized this corner stone in the history of chemistry as "the beginning of the truly scientific organic chemistry" [9].

2.2 **Production and use of urea**

Urea, or the diamide of carbonic acid, with its chemical formular $(NH_2)_2CO$ is a colourless and odourless cristalline substance, which easily dissolves in water. It is the final product of the protein metabolism of all mammals and therefore also of man. Humans discharge 20 – 35 g of this substance daily with their urine [10].

Technically urea is produced from CO_2 and NH_3 , with ammonium carbamate as an intermediate, in closed units under pressure and elevated temperatures according to the following equation:

$$2 \text{ NH}_3 + \text{CO}_2 \iff H_2 \text{NCOONH}_4 \iff (\text{NH}_2)_2 \text{CO} + H_2 \text{O}$$

The worldwide production capacities for urea are annually published by various international associations and institutions in the area of the fertilizer industry; these include the European Fertilizer Manufacturers Association (efma), the International Fertilizer Industry Association (IFA) and the International Fertilizer Development Center (IFDC). In consideration of the capacities published by them and with regard to the planned extension of the EU as of 2004, one arrives at the following picture:

| Area | Production capacity in t |
|--|--------------------------|
| Western Europe (EU member states) | ca. 5,000,000 |
| - thereof Germany | ca. 2,000,000 |
| EU candidates (beginning 2004) | ca. 5,600,000 |
| Remaining Eastern Europe (without former Soviet Union) | ca. 600 |

Tab. 2. Current production capacity for urea [11]

According to the above data the current production capacity of Western Europe amounts to ca. 5 million t, Germany produces about 2 million t thereof. The main producers in this country are BASF AG in Ludwigshafen, Hydro Agri Brunsbüttel GmbH and SKW Stickstoffwerke Piesteritz GmbH in Lutherstadt Wittenberg. About 80 % of the synthetic urea are utilized in agriculture as fertilizer and feed additive; the remainder is used in the plastics, textile and pharmaceutical industry [10].

2.3 **Properties and environmental compatibility of urea**

Urea is of low acute toxicity and does not act as a skin-irritant; sensitizing, carcinogenic or mutagenic effects are unknown. Urea is not listed as a dangerous substance in annex VI of the Hazardous Substances Ordinance and therefore does not require any labelling according to the Chemicals Act. The Commission for the Evaluation of Substances Hazardous to Waters (KBwS) has classified urea in the Water hazard Class 1 (low hazard to waters). Urea is not subject to the Hazardous Incident Ordinance. This substance is listed in group III comprising products which represent a low endangering potential for the general population and the environment according to current knowledge [10]. Due to its chemical properties urea must be stored or transported separate from nitrites.

Urea and its decomposition products can get into surface waters by leaching out of agricultural areas. However, its excellent solubility in water prevents absorption in the ground or bioaccumulation in considerable amounts. Urea is biodegradable; on reversing the above mentioned synthesis it is decomposed into ammonia and carbon dioxide. Microorganisms transform ammonia to nitrate via nitrite (nitrification), which is then available to plants as a nitrogen resource. This explains the reason for urea being a fertilizer.

2.4 **Production and use of AdBlue**

For the application of urea in the SCR method it is intended to use a solution of technically pure urea in desalted water, which is easier to handle and to dosage. The solution contains 32.5 % by weight of urea, because the cristallisation point reaches a minimum of minus 11 °C (eutectical mixing ratio) at that concentration.

In the future this aqueous solution of urea will be distributed under the internationally applicable designation "AdBlue". After careful evaluation of all aspects with regard to patent rights and registered trade marks, this name was jointly chosen by representatives of the mineral oil business, the chemical industry, the automobile manufacturers and their suppliers [12]. AdBlue can be produced from urea and desalted water at any location, where urea is synthesized in plants of the chemical industry [AdBlue leaflet, attachment 2].

2.5 **Properties and environmental compatibility of AdBlue**

Like pure urea, its aqueous solution is not toxic, it is neither a dangerous substance nor a dangerous good and it is listed in the lowest water-endangering class 1. Storage stability tests of the aqueous urea solution have shown that no changes of concentration occur under regular storage conditions [13]. At higher storage temperatures (> 50 °C), however, AdBlue tends to decompose through the formation of biuret, free ammonia as well as ammonium carbonate and carbamate. This can easily be detected by the odour of ammonia. As already mentioned, the AdBlue solution starts to cristallize at temperatures below minus 11 °C.

Description of product properties as well as product safety data sheets are listed in attachment 1.

The urea solution is transported in insulated tank cars or in plastic bulk containers (IBC). The solution of urea is corrosive. Therefore only stable materials can be used for containers, piping and fittings, such as austenitic steel, aluminum and various plastic materials. Unalloyed and galvanized steels as well as copper and its alloys are not compatible with urea solutions [15].

As mentioned in chapter 2.4, the 32.5 % aqueous solution of urea starts to cristallize at a temperature of minus 11 °C. Therefore, storage tanks, pumps and nozzles need to be heated, if the ambient temperature falls below this level, in order to prevent cristallization. On the other hand AdBlue must be protected against higher temperatures (> 50 °C) to avoid its decomposition.

Due to its excellent solubility in water (1,080 g/l) urea will tend to stay in the aqueous phase in an oil separator, and from there it will get into the public sewage system and eventually into the public waste water treatment plants. There it will be completely catabolized within a few days. However, the question of permits for release of urea solutions into the public sewage systems still needs to be investigated.

2.6 Standardization of AdBlue

Product quality and properties of the 32.5 % aqueous solution of urea are defined by the standard DIN V 70070. This standard provides a quality description of the reducing agent, which is independent of any producer and available to the public. DIN V 70070 was published in August 2003 and shall form the basis for a later EN or ISO standard [15].

Maintaining quality could be a problem when handling urea solutions, especially during transportation. Transportation vessels are often cleaned with agents containing phosphorus, therefore traces of these agents could get into the AdBlue solution. However, this must be avoided, because the catalyst on board of the vehicles can otherwise be poisoned.

3 Demand forecast and profitability of AdBlue

3.1 Demand forecast for AdBlue

On the basis of a survey of all European builders of commercial vehicles, VDA has forecast the AdBlue demand for the purification of exhaust gases by the SCR method as follows:

| Year | 2003 | 2004 | 2005 | 2006 | 2007 |
|--------|---------|-----------|-----------|-----------|-------------|
| t/year | 500 | 6,600 | 60,000 | 350,000 | 640,000 |
| Year | 2008 | 2009 | 2010 | 2011 | 2012 |
| t/year | 950,000 | 1,350,000 | 1,750,000 | 2,150,000 | > 2,500,000 |

Table 3: Annual demand of 32.5 % urea solution (AdBlue) for HD commercial vehicles with SCR technology in Western Europe [14]

As shown in table 3, it is prognosticated that the European annual demand for AdBlue for the SCR technology in HD commercial vehicles will amount to approximately 500 t, initially. By the year 2012 this demand will increase to more than 2.5 million t, which corresponds to about 15 % of the current European production capacity of pure urea. This estimate is based on the assumption that the demand for urea solution amounts to approximately 5 % of the fuel consumption. As the VDA emphasizes, this prediction is a rather conservative estimate. Meanwhile, there is another forecast by ACEA, which goes beyond the VDA prognosis.

If financial incentives are added to the legal requirements of emission limits, e.g. an emissions dependent truck toll on motorways or a corresponding tax for commercial vehicles, then the demand for AdBlue could increase significantly faster than shown in table 3. The same applies, if not only heavy duty trucks, but also light trucks and especially diesel passenger cars will be equipped with SCR converters.

3.2 **Profitability of AdBlue**

The expense for the production of urea is primarily determined by the cost of energy and raw materials (crude oil, natural gas). The cost of production and distribution of AdBlue have to be added. The latter depend mainly on the mode of distribution and the investments that may be required at the service stations. Finally, the retail price will depend on demand, that is, how quickly and to what extent the SCR/urea technology will prevail on the market. Consumption of AdBlue amounts approximately to 5 % of fuel consumption, and the savings in diesel fuel, made possible by the urea solution, are in the same order of magnitude. Therefore the retail price for AdBlue should be lower than that of diesel fuel, thus acting as an incentive for application to the customer. Further motivation could be achieved through a tax reduction or an emission dependent truck toll, in order to provide at least a partial compensation for the additional investments at the vehicle.

Besides, it should be kept in mind that the transportation of AdBlue on board the vehicle will result in a considerable loss of its useful load.

4 National and international activities

A number of committees of the automobile and mineral oil industry as well as their associations, scientific societies, accessories industry, urea manufacturers, shipping agencies, environmental authorities and universities are engaged on a national or European level in the issue of "reducing NO_x emissions from diesel engines" and more specifically in the SCR/urea technology. The activities of the main committees are described in this chapter.

4.1 Association of the automobile industry (VDA)

On request of the manufacturers of commercial vehicles, VDA established an adhoc working group in the beginning of 2001, called "logistics of urea" (renamed later: "working group AdBlue"). In this group, the relevant European manufacturers of automobiles and engines, jointly with producers of urea and tank systems, with participation of the mineral oil business and the operators of commercial vehicles, dealt with the following issues:

- Standardization of the urea solution, quality requirements
- Tanking systems for urea solution
- Interface filling nozzle/filling neck
- Anti-freeze measures
- Urea: production and demand estimate; costs
- Establishment of a balanced distribution network for urea throughout Europe
- Symbol and trade name for the urea solution
- Alternative usage of ammonia as a reducing agent

On the occasion of the 59. International Automobile Exhibition in Hanover in September 2002, VDA has announced that the European manufacturers of HD commercial vehicles have agreed to apply the SCR method in order to meet the Euro 4 and 5 emission limits for commercial diesel engines [16].

The main motivation for introducing this technology instead of engine modifications is the fact that the SCR method is the only possible way, to reduce exhaust gas emissions, optimize engine performance and improve fuel economy (up to about 3 %) at the same time. This is of particular importance, since the motorway toll planned for commercial vehicles, will depend on the amount of emissions. Also, the introduction of a motor vehicle tax, depending on emissions, is currently under discussion. Finally, ACEA points out that SCR technology can be applied to all kinds of engines. At this point it is difficult to predict to what extent light duty commercial vehicles and passenger cars will be equipped with the SCR technique. VDA had already convened a meeting of the boards of the European manufacturers of commercial vehicles in Frankfurt/Main on September 3, 2001. An evaluation of ammonia versus urea with regard to vehicle technique and safety was then undertaken, which resulted in the unanimous decision for the use of urea as a reduction agent, to be distributed under the designation "AdBlue" [17]. According to VDA, the SCR technology combined with urea as a reduction agent decreases NO_x emissions by more than 80 %.

VDA has announced that the European manufacturers of HD commercial vehicles will start to offer vehicles equipped with the SCR/AdBlue system as of 2004/2005, in order to meet the requirements of Euro 4 and 5 of the years 2005 and 2008, respectively. Therefore, according to the conception of VDA, the setup of an infrastructure for the supply of these vehicles with AdBlue should be started no later than 2004. It is difficult to predict at this time, whether and when the SCR converter will be installed in light duty trucks and passenger cars with diesel engines.

4.2 Association of the European Automobile Manufacturers (ACEA)

The association of the European car manufacturers is also engaged in the issue of SCR technology and urea. According to an agreement with VDA, ACEA will focus on the following technical and political issues [18]:

- Exhaust gas regulations
- Systems of self-supervision
- Application in different vehicle categories from HD to light duty commercial vehicles and passenger cars
- Programs for the assistance and introduction of the SCR technique in combination with urea

The ACEA report "Selective Catalytic Reduction" is included as attachment 3 of this report.

Furthermore, ACEA and VDA have formed a joint working group, named "WG Interface Filling Nozzle/Filler Neck for AdBlue", which deals with the interface between service station and vehicle. This work consists mainly of the standardization of filler nozzle and filler neck and the prevention of faulty tanking, i.e. mixing AdBlue with diesel fuel erroneously. For this purpose ACEA has developed a standard for the filler neck on the vehicle, see attachment 5.

4.3 The European Petroleum Industry Association (Europia)

Europia had a number of open questions in connection with the introduction of the SCR technology and has asked ACEA for a response:

- What is the SCR system? What are the alternatives?
- How shall this system be implemented technically and logistically?
- What is the evidence that SCR is necessary, preferred and does work?
- Can the environmental requirements be fulfilled by SCR?
- Will all members of ACEA introduce the SCR/urea system?
- Is SCR/urea a transition solution or a long-term option?
- What does ACEA see as the timing of introduction?
- What are the control mechanisms envisaged to ensure use by drivers?
- Are there special solutions for any nation?
- Is the price of AdBlue below or above that of diesel fuel?

ACEA has answered these questions in September 2002 (see attachment 6).

Meanwhile Europia has declared, they will follow the progress of the work in the DGMK working group concerning the implementation of the SCR/urea technology, especially with regard of product supply and distribution. Furthermore, Europia will communicate those issues within the European mineral oil industry which require a Europe-wide consideration.

4.4 Development in the USA

The USA are also concerned with the issue of "reduction of NO_x emissions from diesel engines"; the US Department of Energy investigates different methods for after-treatment of exhaust gases. Arthur D. Little has written a report on behalf of the National Renewable Energy Laboratory, it is entitled "Selective Catalytic Reduction – Urea Infrastructure Study", and is available through the internet [19].

Representatives of the American Environmental Protection Agency (EPA), US manufacturers of commercial vehicles and organizations of commercial vehicle fleets have a more differentiated point of view vis-à-vis the SCR technology. This method is considered to be at least one option for meeting the new US exhaust gas limits of 2007 and improving fuel economy [20].

4.5 Development in Japan

Japan plans to introduce new exhaust gas regulations by 2007, which will correspond to the requirements of Euro 4/5. MITI, the Japanese "Ministry for International Trade and Industry", has formed a working group in the summer of 2003, which will investigate the various technical possibilities of exhaust gas treatment. Like the USA, Japan would prefer a solution, which provides for a decrease of NO_x emissions without the use of a reduction agent [21].

4.6 Manufacturers of filling nozzles

As a result of the discussion in the ACEA/VDA working group "Interface filling nozzle/filler neck for AdBlue", the firm Elaflex in Hamburg has developed an AdBlue filling nozzle (open system, venting of AdBlue tank through filler neck). It is available as standard ZVA-AdBlue with the following technical data, also see attachment 7:

| Pump delivery: | 40 l/min |
|----------------------------------|-----------|
| Pump pressure: | 2 – 3 bar |
| Outer diameter of the end spout: | 19 mm |
| Inner diameter of the end spout: | 17 mm |
| End spout length: | 70 mm |

The firm IDENTIC AB, Sweden, will develop an AdBlue filling nozzle (as a closed system with separate tank venting), which will make it possible to control the automatic tanking process electronically.

4.7 Tank container

Special smaller AdBlue filling stations have been developed as equipment for private gas stations and truck stops. They are also available in a frost-resistant outdoor version, which is suited for temperatures down to minus 40 °C. Further information can be obtained through the internet under <u>www.tankschutz-bott.de</u>.

5 Infrastructure for AdBlue – Requests of the automobile industry – consequences for the mineral oil industry and trade

5.1 Considerations and requests of the automobile industry

The considerations and requests of the automobile industry result from the following three starting points:

- 1. In 1999 the European Union issued new regulations with more stringent exhaust gas limits, which will require a considerable reduction of NO_x and particle emissions for HD commercial vehicles in 2005 and 2008, respectively.
- 2. The European automobile industry has investigated various technical options in order to meet these more stringent requirements. In 2002 they agreed on <u>one</u> technology: the **selective catalytic reduction** for the application in HD commercial vehicles, because this is the only method which makes it possible to meet the exhaust gas limits of the Euro directive without impairing the performance and the fuel economy of the engines at the same time.
- 3. The SCR method requires the use of a reduction agent for the nitrogen oxides. The automobile industry has agreed on <u>one</u> chemical substance: a 32.5 % **aqueous solution of urea**, which will be distributed under the trade name "AdBlue".

The European automobile industry plans to equip their HD commercial vehicles with the SCR/urea technology beginning in 2004. According to their ideas the distribution of the urea solution "AdBlue" will be accomplished in three phases:

Step 1: supply through filling stations of larger fleet owners Step 2: supply through filling stations along expressway routes in Europe Step 3:supply through public filling stations in a balanced distribution in Europe

At first only HD commercial vehicles with a permissible total weight of over 6 t will be equipped with the SCR technology, so they can meet the more stringent Euro norms of 2005 for NO_x emissions. According to the ideas of the automobile industry, trucks can be supplied initially with "AdBlue" at the filling stations of hauliers and other operators of larger fleets, later on expressway routes in Europe. As a final step the automobile industry requests a Europe-wide distribution of "AdBlue" in a balanced way. This shall be accomplished through the existing public filling stations of the mineral oil companies, which could eventually supply "AdBlue" to all diesel engines equipped with the SCR technique, including diesel passenger cars. For this purpose mineral oil industry and trade would have to equip their service stations with the necessary installations, such as tanks, pipes and filling systems. On September 15, 2002 VDA issued a press release titled: *IAA: Chief engineers for truck construction request a Europe-wide filling station network for additive distribution: "Parallel to the market introduction of vehicles equipped with SCR, the logistics for AdBlue need to be established quickly in Europe".*

According to the automobile industry the SCR/urea technique has proven ist suitability in successfully conducted field tests. The producers of urea can supply the necessary amounts of AdBlue in the required quality.

In view of the introduction of the first SCR vehicles scheduled for 2004, and the enactment of Euro 4 in the following year, the automobile industry expects the mineral oil business to begin the planning for the establishment of the corresponding infrastructure for AdBlue distribution without delay.

5.2 Consequences for the mineral oil business

The following consequences result from the above listed facts for the mineral oil industry and trade: They have to decide,

- 1. whether they are prepared to offer the reducing agent needed for the SCR technology at their filling stations and
- 2. whether they are prepared to install the additional tanks, piping and filling devices, which are needed for AdBlue or
- 3. whether they prefer to offer AdBlue in mobile containers, from which the truck drivers can fill up their AdBlue tanks.

6 Description of necessary developments

The request of the automobile industry to distribute AdBlue in the same way as fuels through service stations results in a number of unresolved questions regarding technical and logistic requirements. These mainly include:

- Standardization and maintaining quality of AdBlue (from product delivery at the producer to sale at the filling station)
- Delivery of AdBlue to the filling station
- Storage of AdBlue at the filling station
- Constructive and technical arrangement of AdBlue and diesel filling pumps
- Temperature effects (cristallisation, hydrolysis)
- Material compatibility
- Endangering of surface waters/oil separator
- Optimal access to the AdBlue filling pump
- Interface filler nozzle/filler neck for AdBlue tanking
- Durability of concrete pavement on driveways against AdBlue dripping losses
- Accounting with the customer at the service station
- Calibrated instrument for volume measuring with official permission

In order to resolve some of these questions, a field test is being conducted at a service station of OMV in Dingolfing/Bavaria since March 2003 under the coordination of DGMK. Proposed solutions will be checked for their suitability during this test, especially during winter time. Parallel to this field test a pilot test is conducted in a climate chamber at the Aral research center. Tanking operations are simulated at normal and very low ambient temperatures, dripping losses are determined and the behavior of AdBlue in oil separators and on concrete surfaces is investigated. In addition, the storage stability of the urea solution at various temperatures, ranging from minus 15 up to plus 70 °C is examined. Activities of VDA and the joint ACEA/VDA working group have been mentioned in chapter 4 already.

The results of the field tests initiated by DGMK will be reported in DGMK research report 616-2 (publication in the middle of 2004). This report will include among others:

- Field test at the OMV service station in Dingolfing, Bavaria
- Pilot and laboratory tests at the Aral research center in Bochum
- Winter tests at specially equipped service stations of Shell and Total
- Vehicle tests of the car manufacturers DaimlerChrysler and MAN during the winter and summer 2003

7 Conclusions

The technology of selective catalytic reduction for the decrease of nitrogen oxide emissions from exhaust gases, in combination with aqueous urea solution as a reducing agent, has proven its applicability in stationary installations, e.g. in production of electricity and in incineration of waste. This method can also be transferred to mobile systems, especially diesel engines of HD trucks. This has been proven by the manufacturers of automobiles through a series of successfully conducted field tests. Questions, which are still unresolved, are currently under investigation in additional tests.

In view of the imminent tightening of the EU exhaust gas regulations for HD trucks and buses, the European automobile industry plans the successive introduction of this technique in commercial vehicles. A stepwise introduction of the first standardtype SCR vehicles is already planned for the year 2004.

The automobile industry proceeds on the assumption that the mineral oil business will offer AdBlue at their public service stations through a separate tanking system, eventually in a balanced distribution. Each mineral oil company will have to decide, whether it will make the required investments at its service station network. In view of these expenses, automobile industry and mineral oil business should jointly investigate the question, whether the customer can be supplied alternatively with AdBlue through mobile containers.

8 Literature

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- [20] Diesel Fuel NEWS, Vol. 7, No. 11, June 23, 2003
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9 List of abbreviations

| ACEA | Association des Constructeures Européens d'Automobiles (Association of the European Automobile Manufacturers) |
|-----------|---|
| BUA | Beratergremium für umweltrelevante Altstoffe der Ges. Dt. Chemiker (Consulting committee for environmentally relevant, existing substances of the Society of German Chemists) |
| DIN | Deutsche Industrienorm (German Industrial Norm) |
| EN | Euro Norm |
| EPA | Environmental Protection Agency, USA |
| EUROPIA | European Petroleum Industry Association |
| GefStoffV | Gefahrstoffverordnung (Hazardous Substances Ordinance) |
| HC | hydrocarbons |
| IAA | Internationale Automobil-Ausstellung (international exhibition of automobiles) |
| IBC | International bulk container |
| ISO | International Standardization Organization |
| MITI | Ministry for International Trade and Industry, Japan |
| PAH | polycyclic aromatic hydrocarbons |
| SCR | selective catalytic reduction |
| SiDBlatt | Sicherheitsdatenblatt (safety data sheet) |
| VDA | Verband der Automobilindustrie (Association of the Automobile Industry) |
| VDI | Verein Deutscher Ingenieure (Society of German Engineers) |
| | |

10 Attachments

Attachment 1 (Anhang 1)

- 1 a) Product specification, AdBlue safety data sheet provided by AgroLinz Melamin GmbH
- 1 b) Product specification, AdBlue safety data sheet provided by Hydro Gas and Chemicals GmbH
- 1 c) Product specification, AdBlue safety data sheet provided by SKW Stickstoffwerke Piesteritz GmbH
- 1 d) AdBlue safety data sheet provided by BASF AG

Attachment 2 (Anhang 2)

AdBlue:

Leaflet of the AdBlue producers: "Reducing agent for SCR exhaust gas aftertreatment of diesel engines"

Attachment 3 (Anhang 3)

ACEA-Report "Selective Catalytic Reduction" The most promising technology to comply with the imminent Euro IV and Euro V emission standards for HD engines, 23 June 2003

Attachment 4 (Anhang 4)

ACEA Statement on the Adoption of SCR-Technology to Reduce Emissions Levels of Heavy-Duty Vehicles

Attachment 5 (Anhang 5)

Standard for Urea filler neck, ACEA information of 18.07.2003

Attachment 6 (Anhang 6)

ACEA's answers to EUROPIA questions EUROPIA/ACEA meeting of 09.09.2002, IAA, exhibition of commercial vehicles, Hanover

Attachment 7 (Anhang 7)

ZVA-AdBlue