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Use of experimental design in development of a catalyst system

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Abstract

NO_x storage and reduction experiments have been performed with stationary operation of a heavy-duty diesel engine rig. An optimization of the NO_x reduction performance has been done using experimental design. The adjustable parameters in this study were cycle time, injection time, injection rate and bypass time (period of reduced flow through catalysts). NO_x was reduced by 50–60% (3.3–4.1 g/kWh) with a fuel penalty below 5%. It was shown that experimental design was efficient for optimizing the NO_x reduction and this systematic approach enabled important conclusions to be drawn about the system performance.

Keywords: Experimental design; Design of experiments; DoE; NO_x storage and reduction; NSR; Injection parameters; Fuel penalty; Bypass; System optimization

1. Introduction

1.1. Background

The legislative limits for permitted NO_x emissions in exhaust gas from heavy-duty diesel engines will be decreased by 60% (3.0 g/kWh) until 2008 in the European Union [1]. In the USA and Japan a similar trend can be seen. Thus, the development of a NO_x after treatment system for heavy-duty trucks seems unavoidable. The aim of this work is to develop a fully operational after treatment system that reduces NO_x from the exhaust of a heavy-duty diesel engine. This after treatment system is based on the NO_x storage and reduction approach: the diesel engine runs in a continuous lean mode, i.e., there is a high concentration of oxygen in the exhaust gas. Under these conditions NO_x can be stored as $Ba(NO_3)_2$ on a $BaCO_3$ surface on the catalyst [2]. After a period of 1 or 2 min, diesel fuel is injected into the exhaust stream creating rich conditions. Then, the stored NO_x is desorbed and reduced to nitrogen.

1.2. The project

The engine rig consists of an 11 dm^3 Scania diesel engine, oxidation catalysts and NO_x storage and reduction catalysts. A bypass system has been installed to reduce the catalyst flow under the regeneration periods. This helps to avoid a high fuel consumption to obtain rich conditions [3]. Also without the bypass flow, there is a risk that the heat created from hydrocarbon oxidation could destroy the catalyst. It is the aim of this project to develop an after treatment system able to control the NO_x reduction performance under transient conditions.

1.3. Project sub task: optimizing parameters

The degree of NO_x reduction is dependent on a number of uncontrolled parameters, e.g., temperature which is governed by engine speed and torque as well as several other controllable parameters (described in Section 3.2). This study is focused on steady-state engine experiments (constant speed and torque, i.e. load points) and was performed to optimize the controllable parameters for maximum NO_x reduction with low fuel penalty at each given load point. The results will be used when implementing a control strategy for a European transient cycle (ETC).

1.4. Design of experiments (DoE)

Experimental design or design of experiments (DoE) has been used for many years within the field of catalysis.

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The applications cover, for example, catalytic reactors [4], catalyst formulation and preparation [5–7], catalytic kinetic modeling [8,9] as well as NO_x reduction experiments [3]. DoE is an important tool and in this article, we have focused on the use of small designs in the application for NO_x reduction in a full-scale engine rig, where the design parameters are of a practical nature (i.e. the parameters that are easily modified). The objective of DoE is to gain information with as few experiments as possible. Without the use of DoE in this project, the time and labor needed for the investigation would increase significantly using other methodologies such as variation of one parameter at a time. The experiments define an experimental space (i.e. range of parameter values) where a simple linear model may be fitted to estimate the effects of the parameters under investigation. In this study we used so called screening designs. Their purpose is to model main factors and to capture general trends. Screening designs are characterized by a limited number of experiments to save time, so called center points to estimate the reproducibility and the ability to model the responses (in this case NO_x reduction and fuel penalty) with linear terms. In this article we have also included one interaction term and some experimental constraints. This article is intended to be a methodological description of how to deal with complex experimental systems. The methodology is shown to be very efficient in this project but is applicable for every NO_x storage and reduction system as well as any experimental optimization task.

2. Experimental

2.1. Experimental setup

The experimental setup used in the NO_x storage and reduction experiments is shown in Fig. 1. The setup contains a bypass line to bypass most of the exhaust gas under the regeneration periods. Thus a portion of the exhaust bypasses the catalyst and is not subject to treatment. However, the NO_x emissions which bypass the catalyst are not regarded as a major contributor to the total NO_x emissions. The catalysts consist of NO_x traps with a total volume of 18.9 dm^3 and oxidation catalysts of 9.4 dm^3 to pre-oxidize NO and the injected hydrocarbons. The NO_x traps contained Pt/BaO among other components on an Al_2O_3 support, the oxidation catalyst was Pt on Al_2O_3. The system has been described previously in detail [10,11]. Gas sampling is made up- and downstream of the bypass line so that the whole exhaust gas flow is subject to gas analysis.

2.2. Parameter definition

There are many parameters that influence the NO_x reduction performance. The catalyst temperature is one main parameter for NO_x conversion because of its strong influence on reaction kinetics and equilibrium. The temperature is, however, governed by the engine speed and torque, which are uncontrollable under normal operation of the engine in a vehicle. Thus, the temperature is not regarded as a controllable parameter. The parameters for injecting the reducing agent are, however, controllable and are used for optimizing the NO_x reduction performance. Fig. 2 illustrates the injection parameters. A cycle consists of two phases, one longer lean period (NO_x storage) and one shorter rich period with reduced flow (NO_x release and reduction, bypass time). The rich phase or ''bypass time'' starts with an injection of reducing agent (here: Swedish MK1 diesel) into the exhaust stream possibly followed by a time period with no injection and just the reduced flow. The bypass time can thus be equal to or longer than the injection time. The design parameters chosen were:

1. Cycle time (ct) (s).
2. Injection time (it) (s).
3. Injection rate (ir) (mg/s).
4. Bypass time (bt) (s).

Other parameters are just functions of the ones above, making them impossible to vary independently (i.e., storage time = cycle time − bypass time, injected amount = injection time × injection rate). For every design experiment, 5 cycles were run. The responses, namely NO_x reduction and fuel penalty were calculated as an average of the three last cycles. The fuel penalty is the ratio between the injected amount and the engine fuel consumption given in percent.

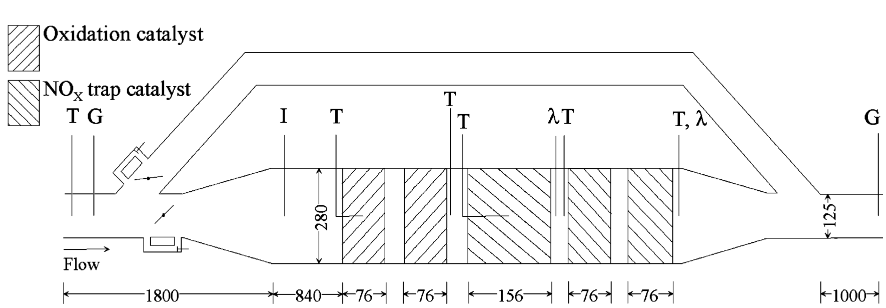

Fig. 1. Catalyst setup. G = gas sampling point, T = thermocouple, λ = broad band λ-sensor, I = injector.