Influences of exhaust gas temperature and flow rate on optimal catalyst activity profiles

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ABSTRACT

This paper presents an optimization of an axial catalyst distribution for the two types of optimal axial catalyst distributions: continuous exponential and discrete two-zone, to enhance the light-off of a catalytic converter with a fixed amount of a catalyst. The influences of exhaust gas temperature and mass flow rate on the two types of optimal active component distribution profiles are identified and hence an optimum value for each of the operating conditions is quantitatively established. The catalyst surface area of the optimal distribution in the upstream section is greater, and in the downstream is lower than that of the uniform distribution. As an inlet exhaust gas temperature becomes lower than the catalyst light-off temperature, i.e., about 600 K, the catalyst surface area of the optimal distribution at the monolith inlet increases greatly to attain high reaction rates at the monolith entrance. In contrast, at temperatures above 600 K, it decreases slightly and hence its variation becomes negligibly small, as compared to the variation of the inlet gas temperature. The catalyst surface area at the monolith entrance decreases gradually with the increasing mass flow rate at gas temperatures above 550 K.

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1. Introduction

While catalytic converters have been widely used to reduce harmful emissions, the converters operate inefficiently until they are heated up to optimum operating temperature range, namely a light-off temperature of typically 600 K or higher, which is generally taking several minutes of operation after a cold start of the internal combustion engines [1]. Consequently, combustion engines emit about 60–80% of their total mass emissions during the first 200 s of operation when tested under the New European Driving Cycle (NEDC) and the US FTP-75 cycle [1–3]. For this reason, the achievement of intensified emission standards for combustion engines will be based on the reduction of cold-start emissions by achieving faster catalyst light-off [1].

There have been numerous prior attempts to either provide alternative emissions control or to further enhance the performance of catalytic converters. One of the more prevalent methods of expediting catalyst temperature rise is to electrically heat the catalyst substrate. However, this is inefficient and yet more equipment is essentially required, such as an auxiliary battery, further increasing the price penalty associated with the incorporation of such systems. Another way is to improve the light-off of the

catalytic converter using a non-uniform catalyst distribution along the monolith length, and the effect of the non-uniform axial distribution on the catalytic converter performance has been studied for a simple chemical reaction and a specific operating condition [4–12]. Oh and Cavendish [4] studied the light-off behavior of three different catalyst distributions, such as uniform, linear increasing and linear decreasing. Psyllos and Philippopoulos [5] showed the CO oxidation performance of catalysts with various parabolic axial catalyst distributions. Non-uniform axial catalyst distribution within a pellet also provided a better performance, as compared to uniform catalysts [6]. For a methane oxidation, it was presented that non-uniform axial catalyst distributions have a high potential to attain lower thermal stresses, particularly during steady-state operation, but only small changes in the methane reduction were observed for all of the distributions considered [7]. Also, in terms of economic aspects and conversion performance, non-uniform catalyst distribution is more effective than the uniform distribution [8]. The aforementioned studies demonstrated that non-uniform catalyst distribution can enhance the conversion performance of the converter. However, an optimal axial catalyst profile has not been identified. In comparison, the optimal catalyst distribution profiles were determined by using different mathematical models and optimization methods [9–12]. They showed that a high catalyst loading in the monolith upstream reduces significantly the pollutant emissions under certain operating conditions. Furthermore,

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Nomenclature

a_f, a_r	dimensionless catalytic surface area per unit monolith volume in the first and second zones of the monolith, respectively
$a(x)$	catalyst distribution function (m^{-1})
a_0	total catalytic surface area over total monolith volume (m^{-1})
A	open frontal area of the monolith (m^2)
c_i	concentration of species i (mole fraction)
c_p	specific heat capacity $(J\,kg^{-1}\,K^{-1})$
CE	CO emissions per unit time in the exhaust gas $(kg\,s^{-1})$
d_h	hydraulic diameter of a channel of the monolith (m)
D_i	binary diffusion coefficient of species i in the mixture $(m^2\,s^{-1})$
$E_{a,k}$	activation energy of reaction k $(J\,mol^{-1})$
$F(X)$	objective function
G	inhibition factor (K)
GSA	geometrical surface area per unit monolith volume (m^{-1})
h_t	heat transfer coefficient $(W\,m^{-2}\,K^{-1})$
$k_{a,j}$	pre-exponential factor of the adsorption equilibrium constant
$k_{m,i}$	mass transfer coefficient of species i in the mixture $(m\,s^{-1})$
$k_{0,i}$	pre-exponential factor of rate constant i $(mol\,K\,m^{-2}\,s^{-1})$
K_j	adsorption equilibrium constant
L	channel length (m)
m	mass flow rate $(kg\,s^{-1})$
N_R	number of reactions
Nu_∞	limiting Nusselt number $(Nu = h_t d_h \lambda_g^{-1})$
P_t	total pressure (Pa)
r_i	rate of reaction i $(mol\,m^{-2}\,s^{-1})$
R_g	universal gas constant $(J\,mol^{-1}\,K^{-1})$

Sh_∞	limiting Sherwood number $(Sh = k_{m,i} d_h D_i^{-1})$
t	time (s)
t_f	time required to reach steady-state (s)
T	temperature (K)
v_{ik}	stoichiometric coefficient of species i in the reaction k
v	superficial gas phase velocity $(m\rho_g^{in-1}A^{-1}$, m s$^{-1})$
V_m	monolith volume (m^3)
x	axial coordinate (m)
X	design variables vector

Greek letters

α	coefficient in the catalyst distribution function $a(x)$
β	coefficient in the catalyst distribution function $a(x)$
$\Delta H_{a,j}$	adsorption heat $(J\,mol^{-1})$
ΔH_k	enthalpy of reaction k $(J\,mol^{-1})$
ε	void fraction of the monolith
λ	thermal conductivity $(W\,m^{-1}\,K^{-1})$
ξ	dimensionless axial coordinate
ρ	density $(kg\,m^{-3})$
$\Phi(T_s)$	$(1-\varepsilon)\rho_s[c_{p,s} + T_s(dc_{p,s}/dT_s)]$

Subscripts

1	length of the first zone in discrete two-zone distribution
g	gas
i	species index
k	reaction or objective function index
s	solid

Superscripts

0	initial
in	inlet

Kim et al. [1] performed the optimization of an axial catalyst distribution with a fixed amount of catalyst to achieve the best conversion performance of a dual-monolith catalyst system during the first 200 s of the FTP-75 cycle. Among the previous studies regarding the optimal design of the catalyst loading pattern, most have been performed for a specific operating condition and hence the effect of various operating conditions, such as mass flow rate and gas temperature, on the optimal catalyst distribution was not considered.

In the present work, based on the optimization algorithm first suggested by Kim and Kim [12], the optimization of an axial catalyst distribution with a fixed amount of an active metal for the minimization of cold-start pollutant emissions over a wide range of operating conditions, i.e., 525–900 K and 20–80 g/s, has been performed for the two types of optimal axial catalyst distributions: continuous exponential and discrete two-zone. The effect of the exhaust gas temperature and mass flow rate on the optimal axial catalyst distribution is extensively examined, and an optimum value for each of the operating conditions is quantitatively established.

2. Model description

2.1. Physical–mathematical model

In a one-dimensional (1D) heterogeneous plug-flow reactor (PFR) model, the axial temperature and concentration gradients should be considered in a full-scale monolithic catalytic converter [13,14]. Here, the dispersion in the axial direction is negligible due to the short residence times, the laminar flow in the channels and the high values of the Peclet number. The accumulation of heat and mass in the gas phase and mass in the solid phase can also be neglected for a quasi-steady PFR model since their time constants are much smaller than those of the solid thermal response [15]. The conduction and heat accumulation in the solid phase, on the other hand, must be taken into consideration along with the heat and mass transfer between gas and solid phase. An 1D PFR model solves the mass and energy conservation equations for both gas and solid phases. The two phases are coupled with the transport coefficients for convective heat and mass transfer. The energy balance in the gas phase involving convection and external heat transfer can be described as:

$$v\rho_g c_{p,g} \frac{\partial T_g}{\partial x} = -h_t GSA(T_g - T_s) \tag{1}$$

By considering convection and external mass transfer, the species mass balance in the gas phase is given by

$$v\frac{\partial c_{i,g}}{\partial x} = -k_{m,i} GSA(c_{i,g} - c_{i,s}), \quad i = CO, C_3H_6, CH_4, H_2, O_2 \tag{2}$$

The energy balance equation in the solid phase takes account of accumulation, axial heat conduction, external convective heat transfer and heat source from the chemical reactions, which can be expressed as

$$\Phi(T_s)\frac{\partial T_s}{\partial t} = (1-\varepsilon)\lambda_s \frac{\partial^2 T_s}{\partial x^2} - h_t GSA(T_s - T_g)$$
$$+ a(x)\sum_{k=1}^{N_R}(-\Delta H_k)r_k \tag{3}$$