

# HEAT AND MASS TRANSFER LIMITATIONS IN MONOLITH REACTOR SIMULATION WITH NON UNIFORM WASHCOAT THICKNESS

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**Abstract**— In this contribution, effectiveness factor ( $\eta$ ) calculations are performed by a perturbation and matching technique developed by the authors which takes into account both the intrinsic kinetic expression and external heat and mass transfer resistances. The simplified method of Papadias *et al.* (2000) was used to consider the non-uniform washcoat thickness usually present in monolith channels. As a result a global effectiveness factor ( $\eta_0$ ) is calculated at each point on the grid to simulate monolith reactor performance. The procedure was tested to predict experimental findings taking into account the actual kinetic expression to describe CO oxidation on Pt catalyst. Agreement among theoretical predictions and Ullah *et al.* (1992) and Holmgren and Andersson (1998) experimental results are fairly good.

**Keywords**— Monolith reactor, Diffusion, Reactor engineering, Effectiveness factor.

## I. INTRODUCTION

Catalytic monolith reactors are widely used to reduce emissions of undesired products in automotive exhaust gases; the abatement of NO<sub>x</sub> emitted in the stack gases from power stations by the selective catalytic reduction processes, and the catalytic combustion of volatile organic compounds (VOCs) (Cybulski and Moulijn, 1994; Heck *et al.*, 2001). Monoliths are increasingly under development and evaluation for many new reactor applications. As a consequence, the studies with monolithic catalysts are now more and more relevant in areas outside traditional chemical engineering and catalysis, e.g., energy fields such as production, car manufacturing and pollution control.

A monolith reactor is an array of channels with honeycomb like structure. The monolith catalyst is a structured substrate (e.g., cordierite, a material having a low thermal expansion coefficient) which is covered with a layer (washcoat) of material that serves as a catalyst. The low pressure drop of the monolith, compared to a packed catalyst bed, is a great and important advantage. When a monolith reactor operates the reactants must be transported from the bulk fluid phase to the solid interface. Then, they must diffuse and react into the catalytic washcoat in a simultaneous process. Due to the importance assigned to external mass transfer in monolith reactors, there have been several theoretical (Balakotaiah and West, 2002) and experimental (Holmgren and

Andersson, 1998; Uberoi and Pereira, 1996) studies on this subject. Therefore, various external mass transport correlations are available for typical monolith reactor applications and for different channel geometry.

Several researchers, recognizing that washcoat layers are thin (e.g. 10 – 100  $\mu\text{m}$ ), have assumed that the effect of the internal mass transport is negligible (i.e. effectiveness factor of one) (Ullah *et al.*, 1992; Uberoi and Pereira, 1996). Other studies have considered first or pseudo-first order kinetic expressions for the intrinsic chemical reaction (Groppi *et al.*, 1995), oversimplified kinetic expression (Holmgren and Andersson, 1998) or global kinetics that included pore diffusion. Notwithstanding the small washcoat thickness, the influence of internal diffusion is significant for many reactions under operation conditions usually found in practice (Leung *et al.*, 1996; Hayes *et al.*, 2004).

Diffusion and reaction inside the washcoat are characterized by the intrinsic effectiveness factor  $\eta$ . Meanwhile the global effectiveness factor  $\eta_0$  is used to quantify the combination of limitations, the internal resistance (washcoat) and the external mass transfer. The use of the full expression of the intrinsic rate of reaction is necessary to account for the effect of the internal washcoat mass transport resistance. However, for complex non linear kinetic expressions the effectiveness factor calculation becomes computationally expensive. The solution of the heat and mass balance differential equations is quite difficult. Besides these difficulties, when a honeycomb like catalyst is prepared, there is a tendency for the coating to accumulate in the corners of monolith channel. Therefore, the varying thickness of the catalytic washcoat should be considered to estimate effectiveness factors. Papadias *et al.* (2000) reported a simplified method to calculate effectiveness factor in non uniform washcoat shapes. They proposed dividing the washcoat into a series of slices (or particles side by side on the monolith wall). A variable effectiveness factor  $\eta_i$  in each slice is calculated using a 1D analysis, assuming a characteristic length for each slice ( $L_{ci}$ ) as the ratio between its area ( $A_i$ ) and the length of the fluid solid interface ( $L_i$ ):

$$L_{ci} = \frac{A_i}{L_i} \quad (1)$$

Finally, by considering an approximate average surface concentration all over the washcoat perimeter, the aver-