

EFFECTIVENESS FACTOR CALCULATION AND MONOLITH REACTOR SIMULATION WITH NON-UNIFORM WASHCOAT AND ARBITRARY CATALYTIC ACTIVITY DISTRIBUTION

M. PARENTIS, N. BONINI[†] and E. E. GONZO[‡]

INIQUI-Instituto de Investigaciones para la Industria Química, Univ. Nac. de Salta, Facultad de Ingeniería,

[†]Facultad de Ciencias Exactas, Buenos Aires 177, A4402FDC Salta, Argentina.

[‡]gonzo@unsa.edu.ar

Abstract— A simple, precise and fast procedure to simulate monolith reactors is presented. The method allows the estimation of effectiveness factors (η) in monolith with washcoat of irregular geometries and arbitrary catalytic activity distribution. Catalytic washcoat with the same quantity of active material, deposited in different manners, are compared in their influence on monolith reactor performance. Intrinsic effectiveness factor estimations, with the approximate method, for first order reaction gave results very close to the rigorous 2D calculation. It is shown that differences between η values can be as much as 54% when non uniform catalytic activity distribution is considered. It is also shown the influence of different catalyst distribution on the behavior of a monolith reactor where the isothermal NO decomposition on Cu/ZSM-5 washcoat with complex Langmuir-Hinshelwood kinetic expression, is carried out. Estimated results are in close agreement with experimental findings. The influence of different catalytic activity distribution on η can be as much as 66%, while exit NO conversion changes more than 42%.

Keywords— Effectiveness factor, Monolith reactor simulation, non-uniform washcoat, non-uniform catalytic distribution.

I. INTRODUCTION

Today, due to their advantages respect to packed-bed or slurry reactors, monolithic reactors are widely used in reducing air pollution by automobile exhaust gases, selective catalytic reduction of nitrogen oxides, catalytic abatement of volatile organic compounds, catalytic fuel combustion and hydrogen production (Irandoost *et al.* 1998; Heck and Ferrauto, 2001; Williams, 2001; Machado *et al.*, 2005; Gonzo, 2008; Chauhan and Srivastava, 2008).

Ceramic and metallic monoliths are produced commercially, although ceramic monoliths have dominated the field thus far (Valentini *et al.*, 2001). The structure of the ceramic monolith can be of low or high surface area. In general, low surface area and metallic supports have been developed for emission control. In these cases the catalytic compound is present in a thin layer on the monolith wall. Usually, it is distinguished a primary and a secondary support. The primary support is the monolith itself and the secondary support is the material

where the catalytic active compound is applied. Normal procedures used involves the application of the pure secondary support and then loading it with the active component or first loading the secondary support with the catalytic active material and then coating the wall of the monolith with the loaded secondary support. These two procedures are known as “wash-coating” and “dip-coating”, respectively (Geus and van Giezen, 1999). In the case of “wash-coating”, the precursor of the active component is applied by impregnation of the washcoat with a solution of a precursor of the active compound and then drying. Also a deposition-precipitation procedure can be used. In this case the catalytic active material precursor is precipitated from the solution.

Dip-coating procedure gives, generally, washcoat with uniform catalytic activity distribution. While wash-coating gives non-uniform distributions. The use of a primer to improve the adherence of the washcoat layer is a common procedure (Valentini *et al.*, 2001). In this case, if the washcoat is applied using dip-coating, the catalyst active material distribution will be of the external egg-shell configuration type.

Earlier investigations assumed that the washcoat was so thin that diffusion resistance is not important. However, recent studies have shown this is not true (Hayes *et al.*, 2004; Gonzo, 2008). Also, a frequent approximation in modeling monolithic reactors is to consider a uniform and flat coating of washcoat in the monolith channel. However, as was demonstrated in several works (Hayes *et al.*, 2005; Papadiaz *et al.*, 2000), in the corner of the channel due to non-uniform coating the reaction rate is markedly influenced by diffusion limitations.

Recently, Gonzo and Gottifredi (2010) have published a simple, accurate and fast procedure to predict monolith reactor performance by taking into account realistic kinetics, external and internal mass and heat transfer resistances and geometrical parameters to describe non uniform washcoat thickness along reactor section perimeter. In this contribution the approximate procedure is extended to take into account different catalytic activity distributions in the washcoat. To takes into consideration the irregular geometry of the washcoat, the approximate method, rather than solve the 2D washcoat problem, divides the washcoat into a series of (N) 1D slice. Thus an effectiveness factor (η) is calculated

for each slice and an average intrinsic effectiveness factor for the whole washcoat is estimated according to:

$$\eta = \sum_1^N \omega_i \eta_i, \quad (1)$$

The weight (ω_i) for each slice's effectiveness factor would be the fraction of the total washcoat in the slice (A_i/A). Assuming a characteristic length for each slice (L_{ci}) as the ratio of its cross section area (A_i) to the arc length of the fluid-washcoat interface (L_i). Figure 1 shows (1/4) of a square monolith channel of side (L) covered by a non uniform washcoat with minimum thickness (δ) and a radius in the corner (R_c).

The estimation of the effectiveness factor for each slice, accounting for the non-uniform catalytic activity distribution were carried out through an approximate technique developed by Gottifredi *et al.* (1986), which avoid the numerical integration within the slice. This technique allows the estimation of the effectiveness factor for any form of the kinetic expression, as was successfully applied to the methanol-steam reforming on a monolith reactor (Gonzo, 2008).

II. RESULTS AND DISCUSSION

A. Effectiveness factor estimation

To estimate the effectiveness factor, the dimensionless continuity equation for the key component (B), assuming constant effective diffusivities (D_{Beff}) within each slice (slab geometry), where the catalyst is distributed according to $f(x)$, is given as:

$$\frac{d^2C}{dx^2} = h^2 f(x) R(C), \quad (2)$$

where C and $R(C)$ are the dimensionless concentration and rate of reaction, related to its washcoat-fluid interface value (C'_{Bs} and r_s), and $f(x)$ is the catalytic activity distribution function.

$$C = \frac{C'_B}{C'_{Bs}} \quad R(C) = \frac{r}{r_s}, \quad (3)$$

where C'_B and r_s are the dimensional concentration of

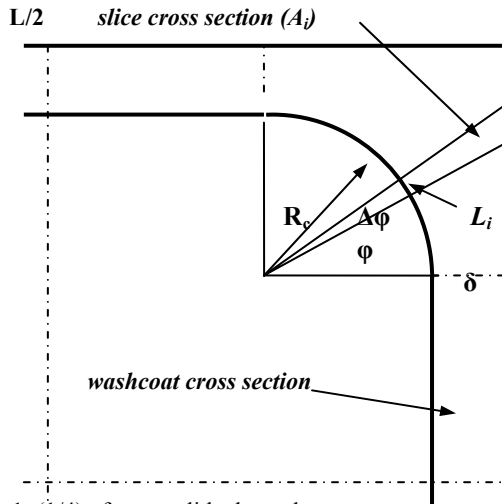


Figure 1: (1/4) of a monolith channel

species B and the reaction rate, respectively. The Thiele modulus h is:

$$h^2 = \frac{L_{ci}^2 r_s}{D_{Beff} C'_{Bs}}, \quad (4)$$

where L_{ci} is the characteristic length for each slice and x is the dimensionless coordinate.

Equation (2) must be solved subject to the following boundary conditions:

$$\frac{dC}{dx} = 0 \quad \text{at} \quad x = 0 \quad (5)$$

and

$$\frac{dC}{dx} = \frac{B_{im}(1-C_s^*)}{C_s^*} \quad \text{at} \quad x = 1.$$

where C_s^* is the dimensionless surface concentration with respect to its bulk fluid value at each point of the monolith reactor, and B_{im} denotes the Biot number for mass fluid film transfer.

$$B_{im} = \frac{k_{gB} L_g}{D_{Beff}}, \quad (6)$$

where k_{gB} is the mass transfer coefficient and L_g is the global characteristic length defined as the ratio between the washcoat total cross section (A) and the fluid-solid interface perimeter.

The catalytic activity distribution function $f(x)$ is also normalized to ensure comparison based on fixed amount of catalyst:

$$\int_0^1 f(x) dx = 1, \quad (7)$$

Consequently η_i is calculated from:

$$\eta_i = \int_0^1 R(C) f(x) dx, \quad (8)$$

Usually, Eq. (2) is solved by applying an orthogonal collocation technique (Villadsen and Michelsen, 1978) or shooting procedures (Kubicek and Hlavacek, 1983; Tischer and Deutschmann, 2005). Instead, in this contribution, η_i is calculated by an approximate procedure (Gottifredi *et al.*, 1986). According to this method, η_i is estimated through the matching expression:

$$\eta_i = [\phi^2 + \exp(-a \phi^2)]^{-\frac{1}{2}}, \quad (9)$$

where

$$\phi = \frac{h}{\rho}; \quad \rho = \left[2 f(1) \int_0^1 R(C) dC \right]^{\frac{1}{2}}, \quad (10)$$

$$a = 1 - 2 \sigma, \quad \sigma = \alpha R'(1) \rho^2,$$

$$\alpha = \int_0^1 \left[\int_0^x f(x) dx \right]^2 dx. \quad (11)$$

In the case that $a \leq 0$, then it must be taken: $a=0$. $R'(1)$ is the first derivative of $R(C)$ with respect to C evaluate at $C=1$.

More details on the procedure to calculate the weight factor (ω_i) and the characteristic length of each slice can be seen in Gonzo and Gottifredi (2010).

The global effectiveness factor and bulk Thiele modulus η_o and ϕ_o are given, respectively, by:

$$\eta_0 = \eta \left(\frac{r_0}{r_s} \right), \quad \phi_0^2 = \phi^2 \left(\frac{r_0}{r_s} \right) \left(\frac{C_s}{C_0} \right), \quad (12)$$

where the subscript “0” indicates parameters calculated at the bulk fluid phase conditions.

B. Catalytic activity distribution function, $f(x)$

Taking into account previous contributions of Li *et al.* (1994) and Au *et al.* (1995), in which realistic $f(x)$ are found from experimental basis and considering the “dip-coating”, “wash-coating” or primer plus “dip-coating” procedure to prepare the catalytic monolith; the following functions for the catalytic activity distribution will be analyzed:

- a) Uniform $f(x) = 1 \quad 0 \leq x \leq 1 \quad f(1) = 1 \quad \alpha = 1/3$
- b) Linear $f(x) = 2x \quad 0 \leq x \leq 1 \quad f(1) = 2 \quad \alpha = 1/5$
- c) Parabolic $f(x) = 3x^2 \quad 0 \leq x \leq 1 \quad f(1) = 3 \quad \alpha = 1/7$
- d) Uniform in a shell $f(x) = 3.333 \quad 0.7 \leq x \leq 1 \quad f(1) = 3.333 \quad \alpha = 1/10$
- e) Linear Negative $f(x) = 1.5 - x \quad 0 \leq x \leq 1 \quad f(1) = 0.5 \quad \alpha = 0.425$

To have a better insight on the distribution functions utilized in this study, Fig. 2 gives a graphic representation of the different $f(x)$ considered.

To analyze the effect of the different catalytic activity distribution on the estimated intrinsic effectiveness factor for a non-uniform washcoat, the propane oxidation first order reaction expression of Hayes *et al.* (2005), was used:

$$r_p = 5.0 \times 10^9 \exp\left(-\frac{10800}{T}\right) C_p \left(\frac{\text{mol}}{\text{m}^3 \text{ s}} \right), \quad (13)$$

where C_p is the propane molar concentration.

The calculation were performed over the temperature range 500 – 900 K, which covered the extreme cases of small to large effect of diffusion. In all calculations a constant effective propane diffusion coefficient in the washcoat of $1 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$, was assumed. A “filled in square” washcoat shape (Hayes *et al.*, 2005), in a square monolith of 1 mm^2 cross section, 0.145 mm^2

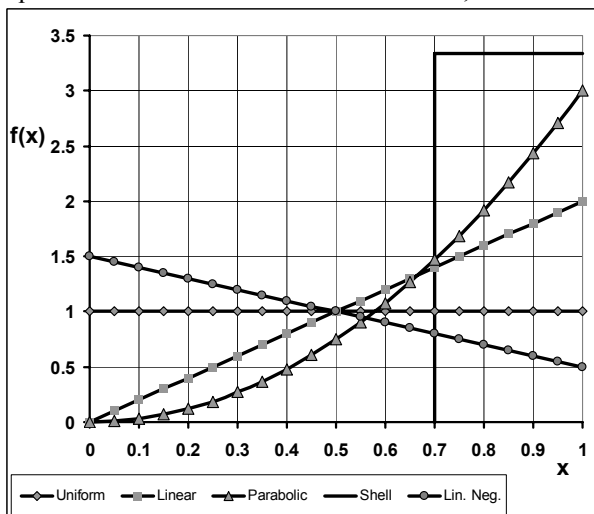


Figure 2: Different catalytic activity distribution considered in this work. Monolith-washcoat interface ($x = 0$). Washcoat-fluid interface ($x = 1$).

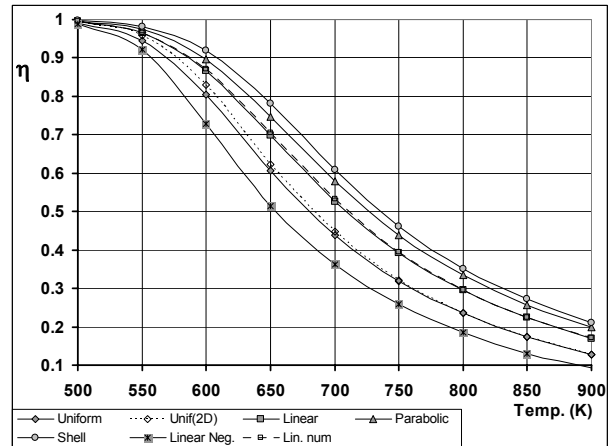


Figure 3: Effectiveness factors for different catalyst distribution. Fillet in square geometry with first-order reaction. Points: numerical data for each case.

washcoat area, an interface length of 3.32 mm and a minimum washcoat thickness of $10 \mu\text{m}$, was considered in this study.

Figure 3 shows the comparison of average intrinsic effectiveness factor calculated with the procedure here presented for different catalytic activity distribution, including the η profile for uniform distribution obtained by 2D simulation for Hayes *et al.* (2005). As can be seen, for uniform activity distribution, maximum difference between approximate and 2D calculated η values is around 3.85%. In this figure it is also depicted the η profile, for linear distribution, calculated using a very time consuming but robust numerical procedure (Kim and Lee, 2004). A maximum difference of 0.8% was found between estimated and calculated η values. In both cases maximum differences are found in the η range 0.7 – 0.8. It is interesting to point out that effectiveness factor estimation for “parabolic” and “uniform in a shell” distribution can be considered undistinguishable since differences are in the order of that observed between numerically calculated and estimated values. Nevertheless, the effect of non-uniform catalytic activity distribution on η can be as high as 100% by comparison with η values considering uniform distribution. Under high Thiele modulus regime the influence of a non-uniform activity distribution can be extremely large.

With the aim to show the influence of different catalyst distribution on the behavior of a monolith reactor, the isothermal decomposition of NO in a square zeolite-based (Cu/ZSM-5 on cordierite) monolith reactor, was studied. Experimental results obtained by Tomašić *et al.* (2004) and Tomašić and Gomzi (2004) in a square monolith reactor, $L_z = 75 \text{ mm}$ long and hydraulic diameter $d_h = 1.537 \text{ mm}$ (monolith MZ1), was used for comparison in this work. The non-uniform washcoat is characterized by a washcoat radius in the corner of 0.346 mm and a minimum washcoat thickness of $153 \mu\text{m}$. The global characteristic length of the catalytic layer was 0.217 mm . Copper ion exchange step of Na/ZSM-5 had been performed before coating the monolith (dip-coating). Tomašić *et al.* (2004) carried out the NO de-

composition at atmospheric pressure at temperature of 673K and at various space times. The temperature of the reactant gas (4% NO/He) was monitored by a thermocouple placed at the exit of the monolith. The following intrinsic kinetic equation is given for the reaction:

$$2 \text{NO} \rightarrow \text{N}_2 + \text{O}_2$$

$$r = \frac{k C_{\text{NO}}^2}{\left(1 + \sqrt{K_D C_{\text{O}_2}}\right)^2} \quad \frac{\text{mol of NO}}{\text{m}^3 \text{ s}}. \quad (14)$$

The value of the reaction rate constants, for monolith MZ1 and MZ2, are, respectively:

$$k = 100 \frac{\text{m}^3}{\text{mol s}} \quad \text{and} \quad k = 360 \frac{\text{m}^3}{\text{mol s}}. \quad (15)$$

The estimated value for the equilibrium adsorption constant was found to be:

$$K_D = 0.02 \frac{\text{m}^3}{\text{mol}}. \quad (16)$$

The main assumptions used to develop the model were: steady-state and equal conditions within each monolith channel, plug flow and negligible pressure drop along the reactor. Due to the low concentration of the reactant a small amount of heat is generated by the reaction, therefore an isothermal system was considered.

Fluid-solid film mass transfer coefficient was calculated using the Holmgren and Andersson (1998) correlation for the average Sh number:

$$\text{Sh} = 3.53 \exp[0.0298 \text{Re} \text{Sc} d_h / L_z], \quad (17)$$

where

$$\text{Re} = \frac{G d_h}{\mu}, \quad \text{Sh} = \frac{k_g d_h}{D_A}, \quad \text{Sc} = \frac{\mu}{\rho D_A}, \quad (18)$$

D_A , μ and G are the bulk diffusion coefficient, fluid viscosity and mass flow density, respectively.

C. Effectiveness factor estimation

Considering the kinetic expression (14), the dimensionless rate of reaction $R(C)$ and $R'(1)$, using C for the dimensionless NO concentration, are:

$$R(C) = \frac{C^2 (1+K)^2}{\left[1 + K(1 + \Gamma_O (C-1)^{1/2})\right]^2}, \quad (19)$$

$$\text{with } K = \sqrt{K_D C_{\text{O}_2}}, \quad \Gamma_O = -\frac{D_{\text{efNO}} C'_{\text{NO}_2}}{2 D_{\text{efO}_2} C'_{\text{O}_2}}, \quad (20)$$

$$\text{and} \quad R'(1) = 2 - \frac{K \Gamma_O}{(1+K)}, \quad (21)$$

where C'_{NO_2} and C'_{O_2} are dimensional concentrations.

D. Reactor simulation

For the simulation the pseudo heterogeneous reactor model (Bischoff and Froment, 1980), accounting for interface and intraphase mass transport limitation was used. According to this model the conversion (X) of the key reactant (NO) along the reactor is given by:

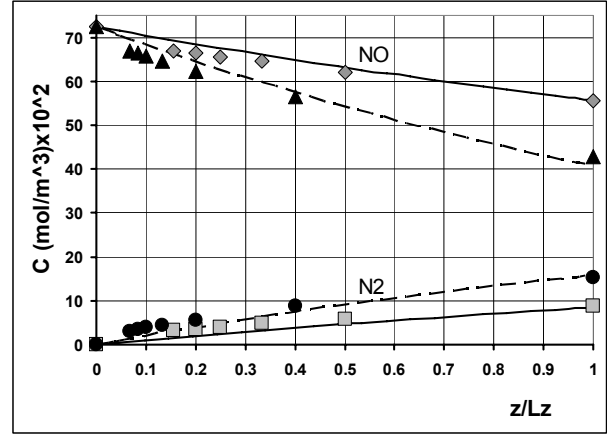


Figure 4: Comparison between experimental data (points) and values predicted by our method (lines). Grey points: sample MZ1. Black points: sample MZ2. Uniform catalytic activity distribution in non uniform washcoat.

$$\frac{dX}{dz} = \Omega \left(\frac{R_V}{F_{\text{NO}}^0} \right) \eta_0 r_0, \quad (22)$$

where Ω is the monolith cross section, η_0 the global effectiveness factor, R_V is the washcoat to monolith channel volume ratio and F_{NO}^0 the NO molar flow rate at the reactor entrance. It must be noticed that the appropriate definition of η_0 allows dealing with a plug flow homogeneous reactor. Since in η_0 calculation the fluid dynamic of the system was taken into account through the interphase mass transfer coefficient (B_{im}).

E. Simulation results

Estimated predictions by our approximate procedure considering uniform catalyst distribution are shown in Fig. 4, where the experimental results obtained by Tomašić *et al.* (2004) are also shown. In this figure results corresponding to the monolith sample MZ2 (global characteristic length $L_g = 0.527$ mm) are also included. The agreement between estimated and experimental data is as good as those obtained by a 2D complete model to describe both gas and solid phases as used by Tomašić *et al.* (2004) for simulating the monolith reactor.

To show the effect of different catalyst distribution in the washcoat on the reactor behavior, the comparison was performed with MZ1 monolith (since gives η values higher than MZ2 monolith) thus expecting a more pronounced effect of catalyst non uniform distribution. The effectiveness factor for sample MZ2 is small ($\eta \approx 0.15$) and consequently η calculated with the global characteristic length gives essentially the same value as η estimated considering non uniform washcoat.

In Figures 5 and 6, NO conversion and intrinsic effectiveness factor along the reactor for the five catalyst distribution function tested, are shown. It is clear that η increases as the catalyst is more concentrated in the outer part of the washcoat, reducing the internal washcoat diffusion effect. The effect of non uniform catalyst distribution on η , compared with uniform distribution, can be as large as 66%. These also explain the increasing exit NO conversion as the distribution goes from “linear

negative” to “uniform”, “linear”, “parabolic” and “uniform in an outer shell”. As can be seen, the non uniform catalyst distribution can affect the exit conversion by approximately 42%. However, for this particular intrinsic kinetic expression, the effect of considering the

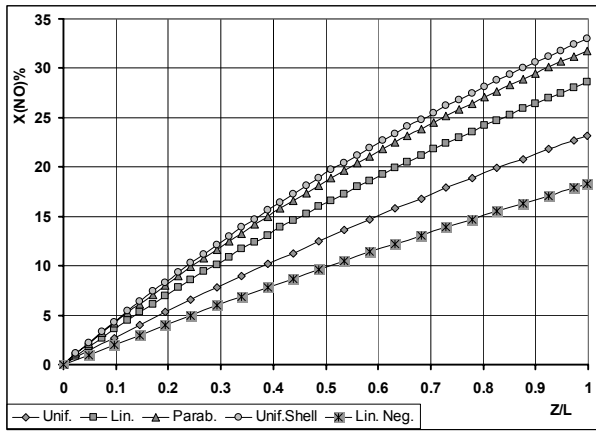


Figure 5: NO conversion along the reactor for different catalyst distribution in the washcoat. MZ1 monolith. Points: numerical data for each case.

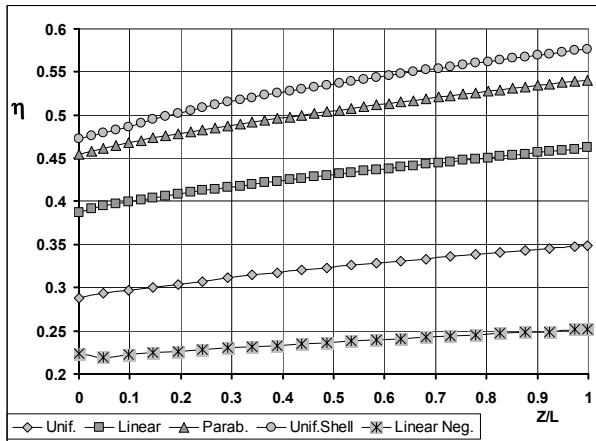


Figure 6: Intrinsic effectiveness factor along the reactor for different catalyst distributions in the washcoat. Points: numerical data for each case.

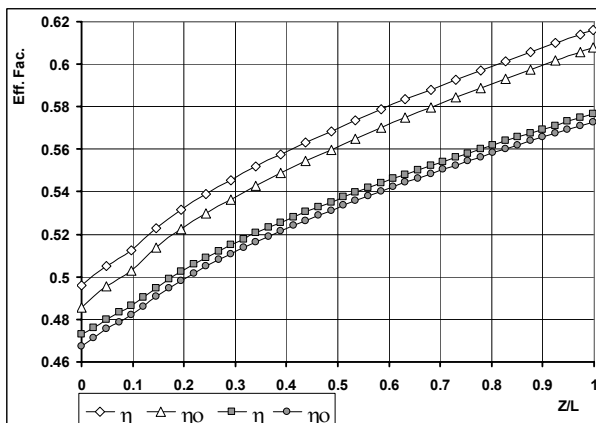


Figure 7: Intrinsic and global average effectiveness factor along the reactor. Monolith MZ1 with egg-shell catalyst distribution. Open symbols: η calculated with the global characteristic length (uniform washcoat). Close symbols: η calculated considering non uniform washcoat. Points: numerical data for each case.

non uniformity of the coating compared with results using global characterization length for the washcoat is only about 6%. This result can be seen in Fig. 7 where global characteristic length and non uniform washcoat thickness finding are compared for the particular case of uniform shell catalyst distribution.

External mass transport limitation is quite small; relative percent concentration difference is less than 0.6%. It must be pointed out that a constant mass transfer coefficient was obtained according to the Holmgren and Andersson (1998) correlation.

For system MZ1 the values of the different parameters are:

$$Re = 24 \quad Sc = 1.281 \quad Sh = 3.577$$

$$kg_{NO} = 0.915 \frac{m}{s}$$

It is important to note that the procedure presented in this work is applicable to reacting system with mild reaction rate. In such processes, the chemical reaction is carried out in the complete catalytic washcoat volume. Therefore, the internal diffusion-reaction process as well as the external heat and mass transport must be considered. The non-uniform thickness of the washcoat and the catalytic activity distribution play an important role in the effectiveness factor calculation. Gonzo and Gottifredi (2010) have shown that under these conditions, the effect of considering external heat and mass transport coefficients values differing in more than 40% only slightly modified the overall effectiveness factor and the effect on conversion along the reactor becomes negligible. Compensation between external and internal resistances is produced, as a whole, does not modify η_0 neither conversion.

However, for very fast exothermic reactions, like those found in catalytic combustors, the reactants are completely consumed at the washcoat-fluid interphase. Consequently, the process is totally controlled by the external heat and mass transport. The washcoat thickness neither the catalytic activity distribution has any importance. Therefore, in these cases, it is necessary to have precise and accurate values of the heat and mass transport coefficients, as was clearly demonstrated by Gupta and Balakotaiah (2001), Donsi *et al.* (2006) and Sari *et al.* (2008).

III. CONCLUSIONS

A simplified method to estimate effectiveness factors in irregular geometries of washcoat, with non uniform catalytic activity distribution, was presented. It can be applied to system with arbitrary intrinsic kinetic expressions. For first order reaction the approximated method gave results very close to the rigorous calculation with finite elements method. For the specific case of NO decomposition on a Cu/ZSM-5 washcoat with complex Langmuir-Hinshelwood kinetic expression (bimolecular surface reaction as the rate determining step), estimated results are in close agreement with experimental findings (Tomašić *et al.*, 2004), when uniform catalyst distribution is considered. It was also shown, in this case, that the differences between effectiveness factor values

calculated considering the non uniform washcoat with values of η estimated using the global characteristic length for the washcoat are of the order of 6%. However, when non uniform catalytic activity distribution is taken into account the differences between η values can be as large as 66%, while exit NO conversions change more than 42%.

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