Conversion in Chemical Reactors using Hollow Cylindrical Catalyst Pellet

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Abstract—Heterogeneous catalysis is vital for a number of chemical, refinery and pollution control processes. The use of catalyst pellets of hollow cylindrical shape provide several distinct advantages over other common shapes, and can therefore help to enhance conversion levels in reactors. A better utilization of the catalytic material is probably most notable of these features due to the absence of the pellet core, which helps to significantly lower the effect of the internal transport resistance. This is reflected in the enhancement of the effectiveness factor. For the case of the first order irreversible kinetics, a substantial increase in the effectiveness factor can be obtained by varying shape parameters. Important shape parameters of a finite hollow cylinder are the ratio of the inside to the outside radii (κ) and the height to the diameter ratio (γ). A high value of κ the generally helps to enhance the effectiveness factor. On the other hand, lower values of the effectiveness factors are obtained when the dimension of the height and the diameter are comparable. Thus, the departure of parameter γ from the unity favors higher effectiveness factor. Since a higher effectiveness factor is a measure of a greater utilization of the catalytic material, higher conversion levels can be achieved using the hollow cylindrical pellets possessing optimized shape parameters.

Keywords—Finite hollow cylinder, Catalyst pellet, Effectiveness factor, Thiele Modulus, Conversion

I. INTRODUCTION

HETEROGENEOUS catalysis is used widely in chemical, refinery and pollution control processes, and is therefore critically important for the economic and environmental welfare of any society. For an optimal heterogeneous catalytic reactor design, many variables must be accounted for in order to maximize reactor performance in terms of yield and selectivity. To this end, an integrated approach should begin at the micro-level by an optimal design of the catalyst itself by considering features such as catalyst size, shape and non-uniform distribution of the active material.

For effective utilization of the expensive catalyst, the active material is mostly dispersed on permeable porous supports with large surface area. The transport of reactant inside the porous network of the catalyst pellet occurs by the diffusion phenomenon. The reaction takes place at the active catalyst site, and the product diffuses out. Due to the transport resistance, the concentration of the reactant in the interior of the catalyst pellet is lower as compared as compared to the bulk fluid. The consequence of concentration gradients is that reactions occur at different rates, depending on position of the catalyst site within the porous support.

It is therefore proposed here to design the shape of the catalyst pellet itself in such a way so that the mass-transport effects are minimized, thereby enhancing the overall effectiveness of the catalyst pellet.

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The present work seeks to address the issue of the shape optimization of the catalyst pellet in order to enhance its effectiveness. This will lead to an improved efficiency of the heterogeneous reactor configuration, and will ultimately help the overall economics of the process. The case study here is based on the hollow cylindrical geometry with a uniform distribution of active component throughout the porous structure. The present study will examine the enhancement of the effectiveness factor by using the hollow cylindrical shape as compared to that of a spherical shape for a fixed catalyst volume for the case of a first order irreversible reaction.

II. LITERATURE REVIEW

The main issues to be considered in the optimal design of the catalyst are mainly its size, shape and the distribution whether uniform or non-uniform of the active component inside the porous support. The effect of deliberate non uniform distribution of the catalytic material within the support started receiving attention in the 1960s. Early publications which demonstrated the superiority of non uniform catalysts include those of Mars and Gorgels [1], Michalko [2,3] and Kasaoka and Sakata [4]. Mars and Gorgels [1] showed that catalyst pellets with an inert core can offer superior selectivity during selective hydrogenation of acetylene in the presence of large excess of ethylene. Michalko [2,3] used sub-surface impregnated Pt/Al2O3 catalyst pellets for automotive exhaust gas treatment and found that they exhibited better long term stability than surface impregnated pellets. Kosaoka and Sakata [4] derived analytical expression for the effectiveness factor for an isothermal, first order reaction with various catalyst activity distribution and showed that those declining towards the slab centre gave higher effectiveness factor. Pellets with larger catalyst activity in the interior than on the surface can result in higher effectiveness factors in the case of reactions which behave as negative order at larger reactant concentrations, such as those with bimolecular Langmuir Hinshelwood kinetics (Villadsen [5]; Becker and Wei [6]). Non uniform catalyst distribution can also improve catalyst performance for reactions following complex kinetics (Juang and Weng [7]; Johnsen and Verykios [8,9]).

An important index of evaluating the performance of the catalyst is the effectiveness factor, which relates primarily to the reactant conversion that can be achieved by a certain amount of catalyst. Studies discussed before demonstrated that non uniform catalysts can offer superior conversion, selectivity, durability and thermal sensitivity characteristics to those where in activity is uniform. This was done by comparing the performance of the catalysts with selected types of activity profiles, which led to the best profile within the class considered, but not to the optimal one. Morbidelli *et al.* [10] first showed that under the constraint of fixed total

amount of active material, the optimal catalyst distribution is an appropriately chosen Dirac-delta function, i.e., all the active catalyst should be located at specific position within pellet. Following this assumption, many research investigations have been carried out on the application of Dirac-delta catalyst for various reactions (Morbidelli *et al.* [11]). This distribution remains optimal even for the most general case of an arbitrary number of reactions with arbitrary kinetics, occurring in a non-isothermal pellet with finite heat and mass transfer resistances (Wu *et al.* [12]). Also, Baratti *et al.* [13] optimized a non-isothermal and non-adiabatic fixed-bed reactor with Dirac-delta type catalyst in a fixed bed reactor

Whilst it has been demonstrated extensively that Dirac-delta type catalysts can be optimized to give the best reactor performance for un-degraded catalyst, when degradation are considered, other profiles might offer advantages. Considering the case of ethylene oxidation, it has been recently demonstrated by Hwang *et al.* [14] that non-uniform catalyst profiles other than Dirac-delta, such as egg-shell, egg-yolk etc, can be optimized to provide a performance approaching that of Dirac-delta under clean conditions. In their work, issues including multiple-beds, different arrangements of non-uniform catalyst pellets, shape and size were all optimized simultaneously in order to achieve a better reactor performance with optimum temperature profiles inside a reactor.

It has been recently pointed out by Armor [15] that the choice of a better catalyst is, in fact, governed by several factors before its actual commercialization can be considered. Although substantial work has been reported in the literature highlighting the usefulness of the Dirac-delta distribution, it is important to note the following points. First of all, it is practically more difficult and economically more expensive to use catalyst with Dirac-delta distribution. Second, it is a wellknown fact that the optimal location of the Dirac-delta distribution inside the spherical catalyst pellet is extremely sensitive, which in turn depends upon the transport and kinetic parameters. Should there be even a small error in the estimation of these parameters, which is quite likely in an actual heterogeneous reactor environment, the resulting performance could substantially deviate from the optimal operation of the heterogeneous reactor. Third, the concentration profile of the reactant always changes along the length inside the reactor, so should the optimal location of the Dirac-delta distribution inside the catalyst pellet along the length the length of the reactor (Khanaev et al. [16]). Implementing such a catalyst distribution in an industrial reactor is quite challenging, if not formidable.

Fourth, the reactor performance could moreover alter significantly, should there occur catalyst degradation.

It becomes abundantly clear at this stage that while the main focus of the research in connection with the optimal design of the catalyst has been the non-uniform distribution of the catalytic activity inside a spherical catalyst pellet, not much work has been reported dealing with the optimization of the shape of the catalyst pellet itself in a heterogeneous reactor

environment. In this connection, it should be pointed out that for any catalytic and non-catalytic contacting of process fluid, it is often desirable to increase the surface area in order to enhance the rate of the heat and the mass transfer. This leads to a straightforward conclusion that a non-spherical catalytic support is preferable over a spherical one since the former provides a greater surface area than the latter for any given volume. In this connection, it is worthwhile to mention a recent study dealing with the simulation of an isothermal hydro-desulfurizaion reactor using different catalyst particle shape (Macias and Ancheyta [17]). Comparing several geometrical shapes of the catalyst with uniform catalytic activity, such as sphere, short-cylinder, long-cylinder, 2-lobe, 3-lobe and 4-lobe shapes, the highest effectiveness factor was reported using 4-lobe shape. Note that that a 4-lobular structure, as seen in their work, provides greater surface area than any other shape, and is about 50% higher than that of a spherical shape for a fixed catalyst volume.

Of non-spherical particles, the hollow shapes hold greater potential (Buffham [18]). Enhancement of effectiveness factor using a hollow particle is, in fact, a straight forward conclusion. When the reaction is controlled by the diffusion, the reactant cannot access the interior of the catalyst pellet. This is reflected in a high value of the Thiele modulus and low effectiveness factor. Removing the interior of the pellet which is not accessible to reactant will naturally lead to an increase in the effectiveness factor. The fabrication of a hollow spherical pellet is of course not as feasible as a hollow cylindrical pellet. There are already several types of hollow non-spherical packing material which are in common industrial use. Note that an additional surface is generated in the case of a hollow cylinder that helps augment the transport of the reactant inside the catalyst. In view of the above discussion, the main goal is to optimize the shape of nonspherical catalyst pellet with a uniform catalytic activity. The objective is to maximize the effectiveness factor in order to enhance the performance of the heterogeneous reactor. The main case study will involve the hollow cylindrical catalyst pellet.

III. MATHEMATICAL MODEL

It is important to mention key features and assumptions of the present heterogeneous isothermal reactor model.

 $These\ are\ following:$

- 1. In the fluid phase, the concentration gradients are only in the axial direction. No gradient exist in the radial direction. This reduces to the one-dimensional problem for the fluid phase.
- 2. In the fluid phase, the mass transport mechanism comprises of both the convection as well as the dispersion.
- 3. In the solid phase, the catalyst pellets are of finite hollow cylindrical shape. Thus, the solid phase is two-dimensional depending upon both the radial as well as the axial directions.
- 4. In the solid phase, the reaction kinetics could be linear or nonlinear. The internal mass transport resistance is important. The effective diffusivities are same in both the radial as well as the axial directions.

5. At the interface of the solid and liquid phase, the film or the external mass transport resistance are important.

A. Solid phase model equations for general kinetics

The steady-state concentration distribution of a reactant inside a permeable finite hollow cylindrical pellet undergoing diffusion-impeded reaction is given by

$$D_{r} \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial C_{As}}{\partial r} \right) + D_{z} \frac{\partial^{2} C_{As}}{\partial z^{2}} - R_{x} \left(C_{As} \right) = 0$$
 (1)

The boundary conditions are

At
$$r = R_i$$
, $D_r \left(\frac{\partial C_{As}}{\partial r} \right) = k_m \left(C_{As} - C_{Af} \right)$ (2a)

At
$$r = R_o$$
, $-D_r \left(\frac{\partial C_{As}}{\partial r} \right) = k_m \left(C_{As} - C_{Af} \right)$ (2b)

At
$$z = 0$$
, $\left(\frac{\partial C_{As}}{\partial z}\right) = 0$ (2c)

At
$$z = (H/2)$$
, $-D_z \left(\frac{\partial C_{As}}{\partial z}\right) = k_m \left(C_{As} - C_{Af}\right)$ (2d)

where, C_{As} and C_{Af} are the reactant concentration in the solid phase and fluid phase, respectively. D_r and D_z , are effective diffusivities in r and z directions, respectively. $R_x(C_{As})$ is the generalized rate of the reaction For the hollow cylindrical catalyst pellet, H is the height, R_i and R_o are inner and outer radii, respectively. k_m is the mass transfer coefficient. Note that the boundary conditions imposed account for the film mass transfer resistance.

Introducing the following dimensionless variables,

$$u = \frac{C_{As}}{C_{Afo}}; \quad \rho = \frac{r}{R_o}; \quad \xi = \frac{z}{(H/2)}; \quad \gamma_o = \frac{(H/2)}{R_o};$$

$$\kappa = \frac{R_i}{R_o}; \quad \gamma^2 = \frac{D_r}{D_z} \gamma_o^2; \quad B_m = \frac{k_m R_o}{D_e}$$
(3)

Rewriting differential equation and boundary conditions in the dimensionless form:

$$\frac{1}{\rho} \frac{\partial}{\partial \rho} \left(\rho \frac{\partial u}{\partial \rho} \right) + \frac{1}{\gamma^2} \left(\frac{\partial^2 u}{\partial \xi^2} \right) - \phi_x^2 f_x(u) = 0 \tag{4}$$

At
$$\rho = \kappa$$
; $-\frac{1}{B_m} \left(\frac{\partial u}{\partial \rho} \right) + u = v$ for all ξ (5a)

At
$$\rho = 1$$
; $\frac{1}{B_m} \left(\frac{\partial u}{\partial \rho} \right) + u = v$ for all ξ (5b)

At
$$\xi = 0$$
; $\frac{\partial u}{\partial \xi} = 0$ for all ρ (5c)

At
$$\xi = 1$$
; $\frac{\gamma_0}{\gamma^2 B_m} \left(\frac{\partial u}{\partial \xi} \right) + u = v$ for all ρ (5d)

where

$$\phi_x^2 = \left(\frac{R_x(C_{Afo})R_o^2}{C_{Afo}D_r}\right); \quad f_x(u) = \frac{R_x(C_{As})}{R_x(C_{Afo})}$$
(6)

Note that a solid cylinder is a special of a hollow cylinder for which the internal diameter is set to zero. Therefore, substituting $\kappa=0$ in the forgoing equations leads to the case of a solid finite cylindrical catalyst pellet.

1. Fluid phase model equations

The differential equation governing the axial distribution of the concentration of the reactant A in the fluid phase (C_{Af}) is given by,

$$\varepsilon \left[\frac{\partial C_{Af}}{\partial t} \right] = - \left[\frac{\partial}{\partial z} \left(-D_{Az} \frac{\partial C_{Af}}{\partial z} + UC_{Af} \right) \right] + \frac{(1 - \varepsilon)}{V_p} W_A$$
 (7)

where, ε is the fraction of solid phase per unit bed volume, V_p is the volume of the catalyst pellet, U is the local fluid superficial velocity, D_{Az} is the fluid phase dispersion coefficient. Here, the molar flow (W_A) of the reactant to the solid catalyst pellet, in terms of the rate of the reaction (R_x) , is give by,

$$W_{A} = \iiint R_{x} \left(C_{As} \right) dV = \int_{0}^{2\pi} \int_{-H/2}^{H/2} \int_{R}^{R_{o}} R_{x} \left(C_{As} \right) r dr dz d\theta \tag{8}$$

where, C_{As} is the concentration distribution of the reactant A in the solid phase comprising of the hollow cylindrical catalyst pellet of height H, and R_i and R_o being the internal and outer radii, respectively.

Introducing the dimensionless variable,

$$\tau = \frac{U_o t}{\varepsilon L}; \quad Pe = \frac{U_o L}{D_{Az}}; \quad v = \frac{C_{Af}}{C_{Afo}}; \quad x = \frac{z}{L};$$

$$y = \frac{P}{P_o}; \quad v = \frac{1 - X}{1 + \sigma X}y;$$
(9)

and rearrangement leads to,

$$\frac{\partial v}{\partial \tau} = \frac{1}{Pe} \frac{\partial^2 v}{\partial x^2} - \frac{1 + \sigma X}{y + \sigma v} \left[\frac{\partial v}{\partial x} - \frac{v}{y} \frac{\partial y}{\partial x} \right] + (1 - \varepsilon) \frac{L}{U_o} \left(\frac{W_A}{C_{Afo}V_p} \right)$$

where, P is the local pressure and P_0 is the pressure at the feed conditions. L is the reactor length, X is the conversion, and σ is the change in total number of moles for complete conversion divided by total moles fed. In the above equation, the Peclet number (Pe) is a measure of the degree of the backmixing prevailing in the reactor. For long reactors, Pe is large and the effect of the fluid back-mixing is negligible. Recalling that the molar flow to the catalyst pellet depends upon the average rate of the reaction inside the catalyst pellet, the fluid-solid interaction term can be written as,

$$\left(\frac{W_A}{V_p}\right) = \frac{2}{1 - \kappa^2} \int_0^1 \int_{\kappa}^1 R_x(C_{As}) \rho d\rho d\xi$$

The steady-state counterpart of (7) can now be rewritten as,

$$\frac{1}{Pe} \left[\frac{d^2 v}{dx^2} \right] - \frac{\left(1 + \sigma\right)}{\left(y + \sigma v\right)^2} \left[y \frac{dv}{dx} - v \frac{dy}{dx} \right]
+ \left(1 - \varepsilon\right) \frac{L}{U_o} \frac{R_{x0}}{C_{Afo}} \frac{2}{1 - \kappa^2} \int_{0}^{1} \frac{R_x \left(C_{As}\right)}{R_{x0}} \rho d\rho d\xi = 0$$
(10)

where

$$R_{x0} = R_x \left(C_{Afo} \right)$$

is the rate of reaction evaluated at the feed conditions.

Defining the effectiveness factor (η) and the Damkohler number (Da) as follows:

$$Da = \frac{LR_{x0}}{U_{o}C_{Afo}}; \quad \eta = \frac{2}{1 - \kappa^{2}} \int_{0}^{1} \int_{\kappa}^{1} \frac{R_{x}(C_{As})}{R_{x0}} \rho d\rho d\xi$$
 (11)

Therefore, Eq. (10) can be written as,

$$\frac{1}{Pe} \left[\frac{d^2 v}{dx^2} \right] - \frac{(1+\sigma)}{(y+\sigma v)^2} \left[y \frac{dv}{dx} - v \frac{dy}{dx} \right] + (1-\varepsilon) Da \eta = 0$$
 (12)

The widely used boundary condition can be rewritten as,

At
$$x = 0$$
; $-\frac{1}{Pe} \left(\frac{dv}{dx} \right) + v = 1$ (13a)

At
$$x = 1$$
; $\frac{dv}{dx} = 0$ (13b)

B. Pressure-drop evaluation in packed bed reactors

For the pressure drop in packed-bed reactors, the well-known Ergun equation can be written

$$-\frac{dP}{dz} = \frac{G(1-\varepsilon)}{\rho \varepsilon^3 D_p} \left[\frac{150(1-\varepsilon)\mu}{D_p} + 1.75G \right]$$
(14)

where, D_p is the catalyst pellet diameter, $G=\rho U$ is the mass velocity. From the mass-conservation principle in flow systems.

$$\rho = \rho_0 \frac{P}{P_0} \left(\frac{T_0}{T}\right) \frac{F_{T_0}}{F_T}$$
(15)

where F_T is the total molar flow, and subscript '0' stands for feed conditions. For isothermal systems, one can therefore write.

$$-\frac{dy}{dx} = \left[\frac{G(1-\varepsilon)L}{\rho_0 D_p \varepsilon^3} \left(\frac{150(1-\varepsilon)\mu}{D_p} + 1.75G\right) \frac{1}{P_0} \frac{1}{y} (1+\sigma X)\right]$$
$$= \left[\beta_0 \left(\frac{1+\sigma}{y+\sigma v}\right)\right]$$
(16)

For the case, $\sigma = 0$, the above equation simplifies to,

$$y = (1 - 2\beta_0 x)^{0.5} \tag{17}$$

IV. RESULTS AND DISCUSSION

The key objective of the present work is to carry out an assessment of the efficacy of the proposed strategy of enhancing the conversion level of a chemical reactor by varying the shape parameters of the hollow cylindrical catalyst pellet. Towards this end, the effectiveness factor is obtained for the case of the linear kinetics first for the catalyst pellet of hollow cylindrical geometry is compared with that of the spherical geometry.

The dependence of the effectiveness factor on the Thiele modulus is shown in Fig. 1a for different γ and a fixed value of the $\kappa=0$, which corresponds to a solid cylinder. Note that the Thiele modulus used is based on the equivalent volume diameter, which is defined as follows:

$$\phi_{V} = \sqrt{\frac{k_{1}}{D_{r}}} R_{V} \tag{18}$$

where, R_V is the equivalent volume diameter defined by,

$$R_V = \left[1.5(1 - \kappa^2)\gamma_0\right]^{\frac{1}{3}}R_0 \tag{19}$$
 The effectiveness factor of the sphere is also presented here

The effectiveness factor of the sphere is also presented here for comparison It is clear from the figure that a nonspherical cylindrical shape leads to a substantial rise in the effectiveness factor of the catalyst pellet. The shape parameter γ plays an

important role in this connection. The greater the departure of γ from unity, the higher is the effectiveness factor. The quantitative improvement obtained by using the nonspherical catalyst pellet is shown in Fig. 1b. The ordinate is the percent difference obtained using the nonspherical particle as compared to the case of the sphere for a fixed volume of the catalyst pellet. An improvement as high as 200% is noted a short cylinder with a height to diameter ratio of 0.050.

The dependence of the effectiveness factor on the γ is shown in Fig. 2 for different Thiele modulus and a fixed value of the $\kappa=0.5.$ It is seen here that as the Thiele modulus increases, the effectiveness factor decreases. This is a clear indication that the internal mass transfer resistance increases with the increase in the Thiele modulus thereby leading to a decrease in the effectiveness factor. This issue is of great interest for the design of heterogeneous reactor owing to the fact that the decrease in the effectiveness is a measure of decrease in the effective utilization of the catalyst being used for the conversion. This consequently leads to a decrease in the conversion level of the reactor.

The key feature of the Fig. 2 is the effect of the γ on the effectiveness factor. It is important to point out here that the total volume of the catalyst pellet is held constant for a fixed value of the Thiele modulus. Therefore, the change in the effectiveness factor seen here is solely due to the variation in γ . It is interesting to note that all the curves show a minimum. The minimum tends to occur when the γ ratio is close to unity. A close look on figure reveals another interesting aspect of these curves. These curves are not symmetrical around the unity. For $\gamma = 0.1$, the effectiveness factor is substantially higher than $\gamma = 10$. Note that smaller values of γ represent hollow ring while larger values of the same represent long hollow cylinder. It can be concluded from Fig. 2 that the greater the departure of the γ from the unity, the higher is the effectiveness factor. For the design of the catalyst shape, such a geometrical configuration is therefore recommended for achieving a higher degree of conversion.

Fig. 3 depicts the dependence of the effectiveness factor on the γ for different values of κ when the Thiele modulus is fixed at 5. As seen before, all the curves show a minimum. This minimum tends to progressively shifts towards a lower y as the value of κ is increased. Note that κ is in fact a measure of the thickness of the hollow cylinder. The higher the κ , the thinner is the hollow cylinder. Smaller values of κ , on the other hand, represents a hollow cylinder for which the inner diameter is very small as compared to the outer one, the shape is thus close to the one of a solid cylinder. This means that the lowest value of the effectiveness factor will occur for the solid cylinder. As the k is increased, the effectiveness factor always shows an increase for all values of the γ . The difference is however more pronounced for tall hollow cylinders. For short hollow cylinders, the change in the κ does not make a significant difference unless it happens to be very thin.

V. CONCLUSIONS

The results presented here conclusively prove that the shape plays an important role in improving the effectiveness of a

catalyst pellet. By varying shape parameters, a significant enhancement in the catalyst effectiveness is evident for the case of the linear kinetics. For example, a shift of the parameter γ from unity is important for better performance. As far as solid cylinders are concerned, a short cylinder with a small γ is better. On the other hand, better performance is obtained for a long hollow cylindrical catalyst pellet as compared to short ones. On the other hand, larger values of the parameter κ are always more useful for the conversion enhancement. For industrial application however, the value of κ will be limited by the strength considerations. In view of this issue, a value of κ (=0.5) is rather more practical. It is seen here that the variation in the shape parameters become increasing important for higher values of the Thiele modulus when the internal mass transport resistance is important.

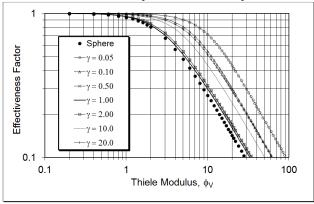


Fig. 1a Dependence of effectiveness factor on the Thiele modulus for $\kappa = 0$

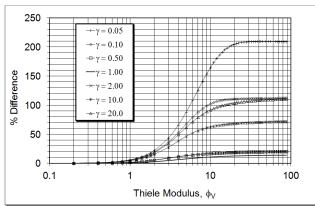


Fig. 1b Percent improvement in effectiveness factor as function of Thiele modulus and γ for $\kappa=0$

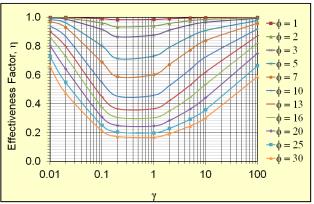


Fig. 2 Dependence of the effectiveness factor on γ for different values of the Thiele modulus and fixed κ =0.5

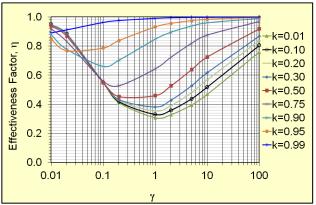


Fig. 3 Dependence of the effectiveness factor on the γ for different values of κ =0.5 and a fixed Thiele modulus (ϕ_{ν} =5)

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