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Transient effectiveness factors in the dynamic analysis of heterogeneous reactors with porous catalyst particles



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HIGHLIGHTS

- A model for the transient effectiveness factor TEF is shown in gradientless reactors.
- The TEF depends not only on the Thiele modulus but also on operational variables.
- The TEF is larger than the steady state effectiveness factor SSEF in pulse injections.
- The SSEF can be corrected by means of two correction factors to yield the TEF.
- This approach is general and embraces the case of the usual steady state assumptions.

ARTICLE INFO

Article history:

Received 6 April 2015

Received in revised form

1 June 2015

Accepted 18 June 2015

Available online 2 July 2015

Keywords:

Transient effectiveness factor

Diffusion–adsorption–reaction

Porous catalysts

Reactor dynamics

ABSTRACT

A model to calculate the actual transient effectiveness factor in spherical porous catalyst particles in gradientless reactors, where a first order reaction takes place under isothermal conditions, linear equilibrium adsorption and intraparticle diffusion control, was developed. After a certain time has elapsed following a change in the feed's reactant concentration, the transient effectiveness factor can be approximated as a linear combination of the steady state effectiveness factor and the relative rate of change of the concentration in the fluid phase. Oppositely to the well-known steady state effectiveness factor, which depends only on the Thiele modulus ϕ , related to intrinsic properties of the catalysts, the transient effectiveness factor also depends on other two dimensionless numbers: α , the relationship between the capacity of accumulation of reactant in the fluid and solid phases, and ϕ_f , a relationship between the convective flow and the intraparticle diffusion rate. In this way, the catalyst load, the reactor volume and the volumetric flow do impact on the effectiveness factor. The coefficients defined as I_a and I_f are the Diffusion–Adsorption–Reaction Factor and the Flow Factor, respectively, which are complex functions of the Thiele modulus, the convective modulus ϕ_f and the system's adsorption capacity α , and which can be used to correct the steady state effectiveness factor to yield the approximated transient effectiveness factor, which is more exact. In the case of a pulse perturbation in the reactant's concentration, the transient effectiveness factor reaches a constant value which is larger than that from the steady state effectiveness factor and, the larger the catalyst mass and the flow, the larger the difference. Results show the existence of a pseudo-equilibrium state in gradientless reactors.

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1. Introduction

In order to simplify the analysis of chemical reaction systems in heterogeneous reactors with porous catalyst particles, the linearization of adsorption isotherms, the lumping of chemical species and the assumption of steady state for the concentration profiles

in the inner surface of the solid catalyst particles are assumed (Aris, 1975). The immediate consequence is that the steady state effectiveness factor η^{ss} , which is defined as the relationship between the actual chemical reaction rate taking place at the inner pore surface and that at the external particle surface, depends only on the dimensionless Thiele modulus (Thiele, 1939). Moreover, the need of solving the partial differential equations (PDEs) of the mass balances in the particle, which are coupled to the corresponding mass balances in the fluid phase, can be avoided since pseudo homogeneous systems outcomes. An important number of works exists where the effectiveness factor is

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Notation			
<i>Symbols</i>		ϕ	Thiele modulus (dimensionless)
C	reactant concentration (gmol/m ³)	ϕ_f	convective modulus (dimensionless)
D	diffusivity (m ² /s)	η	effectiveness factor (dimensionless)
Da	Damköhler number for first order reaction in a flow reactor (dimensionless)	λ	eigenvalues in Eq. (22), defined in Eq. (23) (dimensionless)
F	volumetric flow (m ³ /s)	ρ	dimensionless radial distance
f	perturbation at the inlet concentration in a well stirred flow reactor (gmol/m ³)	τ	dimensionless time
I	correction factors in Eq. (27) (dimensionless)	ξ	dimensionless concentration in solid phase
K	Henry's constant (dimensionless).	<i>Subscripts</i>	
k	reaction rate constant (s ⁻¹)	a	adsorption
Q	concentration in the solid phase, adsorbed compounds (gmol/m ³)	e	equivalent or effective
R	catalyst particle radius (m)	f	fluid phase, Flow
r	radial distance (m)	i	order of coefficients S_i in Eq. (23)
s	coefficients of expansion in Eq. (22), defined in Eq. (23) (dimensionless)	k	refers to the second term in the right hand side in Eq. (28)
t	time (s)	n	order of the eigenvalues in Eq. (22)
V	volume (m ³)	obs	observed
<i>Greek symbols</i>		p	particle
α	relationship between fluid and solid phase capacities (dimensionless)	s	solid phase
χ	dimensionless concentration in the fluid phase	<i>Superscripts</i>	
δ	Dirac delta function	-	volume averaged variable
ε	porosity (dimensionless)	in	inlet
		o	initial
		ss	steady state
		ts	transient state

studied and approximate expressions are developed to obtain fast estimations, or to extend the approach to different particle geometries or chemical reaction kinetics (Bischoff, 1965; Gonzo et al., 1988; Gottifredi et al., 1981; Gottifredi and Gonzo, 2005; Haynes, 1986; Lee and Kim, 2006; Szukiewicz and Petrus, 2004).

Another option to simplify the mathematical treatment is to apply the concept of linear driving force (LDF), an approximation which transforms the partial differential equations (PDEs) in the particle into ordinary differential equations (ODEs) (Álvarez-Ramírez et al., 2005; Glueckauf, 1955; Goto and Hirose, 1993; Kim, 1989, 2008, 2009; Kim and Lee, 2012; Lee and Kim, 2011, 2013; Szukiewicz, 2000, 2002). Even though the LDF concept produces simpler calculations, the total number of dynamic equations does not reduce and, then, most of the studies devoted to non-linear or complex kinetics, even in cases where the balances are simple (e.g. case of gradientless reactors) use the steady state assumption for the reactants' concentrations in the particle (Al-Sabawi and de Lasa, 2010; Gomez Carrasco et al., 2011; Kim et al., 2013; Wang et al., 2011). It is to be mentioned that the simultaneous solution of the coupled transient mass balances for a reactant in the catalyst particles and the fluid phase has been solved by some authors following different aims (e.g. Frost, 1981; Marroquin de la Rosa et al., 2002; Miró et al., 1986; Oberoi et al., 1980; Schobert and Ma, 1981a, 1981b).

Besides the widespread use of steady state effectiveness factors, and even though evidences exist about their limitations, the confirmation of the applicability of the steady state assumption in the catalyst particles is very unusual. However, it is true that the concentration profiles inside the particles can be similar to the steady state ones only in cases of moderate changes of the concentration in the fluid phase. Nevertheless, in batch systems with high adsorption capacity, the changes in the concentration of

the reactant are significant and the exact solution of the equations which describe mass transport and chemical reaction simultaneously shows that the steady state assumption for the particle is not always appropriate (Bidabehere and Sedran, 2006). Similar conclusions can be obtained through the observation of the changes of reactant concentration in the fluid phase and averaged in the particle when they are calculated for the transient operation of continuous reactors under high flow conditions (Marroquin de la Rosa et al., 2002).

Since the substitution of the reaction term in the reactor's mass balance by the product between the actual effectiveness factor and the chemical reaction evaluated at the external catalyst surface is attractive and easy to use, it is the objective of this work to provide a simple view of this transient effectiveness factor, aimed at maintaining the classic approach. Moreover, it is shown that it is possible to develop an approximate solution in order to represent such a effectiveness factor directly in terms of physical system parameters and truly measurable macroscopic variables, such as reactor volume, catalyst load, volumetric flow and fluid phase concentration, as applied to gradientless reactors with porous particles. The influence of these parameters, as well as the relationship between the steady state and the transient effectiveness factors, is analyzed in batch and continuous operations.

2. Theoretical model

The system under study is a heterogeneous stirred chemical reactor where solid, porous catalyst particles are used. Assumptions in the model are that an isothermal regime occurs, that an adsorption equilibrium of the reactant is reached in the pores, which is linear ($Q = KC$), that the chemical reaction is first order

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