Equilibrium Profiles of Tubular Reactor Nonlinear Models

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Abstract

The multiplicity of the equilibrium profiles is analyzed for axial dispersion nonisothermal tubular reactors described by Arrhenius type nonlinear models. It is reported that there is at least one steady state among the physically feasible states for such models. Moreover physically meaninful conditions which ensure the multiplicity of equilibrium profiles are given.

1 Introduction

The dynamics of non isothermal tubular reactors are described by nonlinear partial differential equations. The main source of nonlinearities in the dynamics is concentrated in the kinetics terms of the model equations. The existence of Arrhenius type nonlinearities in the kinetics can generate multiple steady states, either stable or unstable, and in practical applications, the unstable steady states may correspond to the operating points of interest. The study of the steady state multiplicity and stability has been the object of intensive research activity in the sixties and seventies: most of the results are gathered in the book chapter written by Varma and Aris [16]. Multiple steady states have been observed experimentally, e.g. in adiabatic tubular reactors [9, 17]. Whereas the steady state multiplicity and stability of stirred tank reactors is now well understood, the tubular reactor case is still the object of research works, see e.g. [16] ,[5].

We report the multiplicity of the equilibrium profiles for axial dispersion nonisothermal tubular reactors governed by nonlinear models, [10], [11]. Essentially, it is emphasized that there is at least one steady state profile in the physically feasible states, by using similar arguments as those considered in [5] and [15] (see also [19]), as well as compactness and nonlinear operator arguments.

Conditions to guarantee the multiplicity of equilibrium profiles are also given.

2 Problem Statement

The dynamics of tubular reactors are typically described by nonlinear partial differential equations (PDEs) derived from mass and energy balances [10, 1]. In particular, the dynamics of an axial dispersion reactor for one nonisothermal reaction are given by the following equations:

$$\left.\begin{array}{rcl} \dot{x}(t) & = & Ax(t) + N(x(t)) \\ x(0) & = & x_0 \in H \end{array}\right\} \tag{1}$$

where H is the separable real Hilbert space given by $H = L^2[0,1] \times L^2[0,1]$ and A is the linear (unbounded) operator defined on its domain

$$\begin{aligned} \mathcal{D}(A) = \Big\{ & x = (x_1, x_2)^T \in H : x, \frac{dx}{dz} \in H \text{ are absolutely continuous,} \\ & \frac{d^2x}{dz^2} \in H \text{ and } \beta_i \frac{dx_i}{dz}(0) - x_i(0) = \beta_i \frac{dx_i}{dz}(1) = 0 \quad i = 1,2 \Big\} \end{aligned} \tag{2}$$

by

$$\begin{aligned} Ax & = \begin{pmatrix} \beta_1 \frac{d^2x_1}{d^2z} - \frac{dx_1}{dz} - \gamma x_1 & 0 \\ 0 & \beta_2 \frac{d^2x_2}{d^2z} - \frac{dx_2}{dz} \end{pmatrix} \\ & = \begin{pmatrix} A_{\beta_1,\gamma} x_1 & 0 \\ 0 & A_{\beta_2,0} x_2 \end{pmatrix}, \end{aligned} \tag{3}$$

and the nonlinear operator N is defined on the set of all physically feasible state values [10]

$$\begin{aligned} D \;:=\; \Big\{ & (x_1, x_2)^T \in H : -1 \le x_1(z) \;\text{ and } 0 \le x_2(z) \le 1, \\ & \text{for almost all } z \in [0,1] \Big\} \end{aligned} \tag{4}$$

by

$$N(x) \;=\; \left(\alpha\delta(1 - x_2)\exp\!\left(\frac{\mu x_1}{1 + x_1}\right), \alpha(1 - x_2)\exp\!\left(\frac{\mu x_1}{1 + x_1}\right) \right)^T . \tag{5}$$

In this paper, we are interested in the existence of multiple equilibrium profiles for the axial dispersion nonisothermal tubular reactor described by the nonlinear model (1). More precisely, our purpose is to establish the existence of multiple solutions to the following functional equation:

$$Ax + N(x) = 0 \;, \quad x = (x_1, x_2)^T \in D \cap \mathcal{D}(A) \;, \tag{6}$$

or equivalently

$$\left\{ \begin{array}{l} \beta_1 \frac{d^2x_1}{d^2z} - \frac{dx_1}{dz} - \gamma x_1 + \alpha\delta(1 - x_2)\exp(\frac{\mu x_1}{1+x_1}) = 0 \;, \\ \\ \beta_2 \frac{d^2x_2}{d^2z} - \frac{dx_2}{dz} + \alpha(1 - x_2)\exp(\frac{\mu x_1}{1+x_1}) = 0 \;, \\ \\ x = (x_1, x_2)^T \in D \cap \mathcal{D}(A) \;. \end{array} \right. \tag{7}$$

Remark 2.1 In [8], the question above is considered for a different model, of the same kind than the model described above. By using arguments from topological degree theory (see e.g. [5, 19]) , the authors show that there are at least two positive solutions. In the same line, in [3], the existence of three solutions for the general problem is established by using the method of sub-super solutions. A model similar to (1)-(5), namely the *adiabatic* tubular reactor model (corresponding to the case $\gamma = 0$ in (1)-(5) above), is also studied in [5, p.248]; however the analysis developed there is based on unfeasible assumptions on the model parameters.

3 Existence of an Equilibrium Profile

In this section, it is reported that there exists at least one equilibrium profile for the general model (1)-(5) *without any additional assumption on its parameters.* Compactness is a main tool in order to establish this result. First recall the following result from C_0 (i.e. strongly continuous) - semigroup theory.

Theorem 3.1 *[13, Theorem 3.3, p. 48]* A C_0-semigroup $(T(t))_{t\geq 0}$ of bounded linear operators on a Banach space is compact (i.e $T(t)$ is a compact operator for all $t > 0$) if and only if $T(t)$ is continuous in the uniform operator topology for $t > 0$ and $R(\lambda, A)$ is compact for all $\lambda \in \rho(A)$.

In what follows, a closed linear operator A densily defined on a linear subspace $\mathcal{D}(\mathcal{A})$, i.e. its domain, will be denoted by the pair $(A, \mathcal{D}(A))$. Now observe that the operator $(A, \mathcal{D}(A))$ given by (2)-(3) generates an exponentially stable C_0- semigroup $(T(t))_{t\geq 0}$ on H (see [21]). Moreover, by the proofs of [10, lemmas 5.1 and 5.2], the resolvent operator $R(0, A)$ is a Hilbert-Schmidt integral operator. Therefore $R(0, A)$ is a compact operator. Consequently, by the resolvent identity, the resolvent operator $R(\lambda, A)$. By using Theorem 3.1, the following proposition can be proved.

Proposition 3.1 The semigroup $(T(t))_{t\geq 0}$ on H, whose infinitesimal generator is the operator $(A, \mathcal{D}(A))$ given by (2)-(3), is compact.

The following measure of noncompactness plays a fundamental role in the sequel. Let B be a bounded subset of a Banach space X. The nonnegative number $\chi(B)$ is defined by

$$\chi(B) \quad := \quad \inf\{r > 0 : B \text{ can be covered by finitely many balls of radius } r\} \ .$$

For a linear operator T on H, the nonnegative number $\chi(T)$ is defined by

$$\chi(T) \quad := \quad \chi(T[B(0, 1)])$$

where $B(0, R)$ denotes the closed ball centered at 0 whose radius is equal to R, viz. $B(0, R) := \{x \in X : \ \| x \| \leq R\}$.

Remark 3.1 (a) If B is a Hausdorff compact set, then $\chi(B) = 0$.
(b) If T is a compact linear operator, then $\chi(T) = 0$.

The semi-inner products $(\cdot, \cdot)_+$ and $(\cdot, \cdot)_-$ on X will also be needed in the analysis below. They are defined, for all $x, y \in X$, as follows:

$$\begin{aligned}
(x, y)_+ &= \max\{(x, y^*) : y^* \in X^*, \| y^* \| = \| y \|, (y, y^*) = \| y \|^2\} \ ,\\
(x, y)_- &= \min\{(x, y^*) : y^* \in X^*, \| y^* \| = \| y \|, (y, y^*) = \| y \|^2\},
\end{aligned}$$

where $(\cdot, \cdot)$ denotes the standard pairing between X and its dual space X^*.

The analysis leading to the main result of this next section, viz Theorem 3.3, is also based on the following two results.

Theorem 3.2 *[14, Theorem D.]* Let D be a closed convex bounded subset of a Banach space X, A be the generator of a C_0-semigroup $(T(t))_{t\geq 0}$ on X such that $\| T(t) \|\leq e^{\omega_0 t}$ for all $t \geq 0$, for some ω_0, and $N : D \to X$ be a locally Lipschitz and bounded function such that:

$$\lim_{h\to 0^+} h^{-1} d(T(h)x + hN(x), D) = 0 \ , \quad \text{for all } \ x \in D \ , \tag{8}$$

and

$$\chi(N(B)) \leq \kappa_1 \, \chi(B) \text{ for } B \subset D, \ \chi(T(t)) \leq e^{\omega_1 t} \text{ for } \ t \geq 0, \quad \omega_1 + \kappa_1 < 0 \ . \tag{9}$$

Then, the equation

$$A\phi + N(\phi) \;\; = \;\; 0$$

admits at least one solution $\phi \in D \cap \mathcal{D}(A)$.

Remark 3.2 If the C_0-semigroup $(T(t))_{t\geq 0}$ is compact, then, for all $t > 0$, $\chi(T(t)) = 0$, whence $\chi(T(t)) \leq e^{\omega_1 t}$ holds for all $t \geq 0$, for any $\omega_1 \in \mathbb{R}$, since $\chi(I) \leq 1$.

Lemma 3.1 *[14, Lemma C.]* Let D be a closed convex bounded subset of a Banach space X, A be the generator of a C_0-semigroup $(T(t))_{t\geq 0}$ on X such that $\| T(t) \|\leq e^{\omega_0 t}$ for all $t \geq 0$, for some ω_0, and $N : D \to X$ be a locally Lipschitz and bounded function. Assume that $D \cap \mathcal{D}(A)$ is dense in the closed set $D \subset X$, D is $T(t)$-invariant, i.e. for all $t \geq 0$, $T(t)[D] \subset D$, and

$$\lim_{h\to 0^+} h^{-1} d(x + hN(x), D) = 0 \ , \quad \text{for all } x \in D \ . \tag{10}$$

If, in addition, there exists $R > 0$ such that

$$(Ax + N(x), x)_- \leq 0 \quad \text{for all } \ x \in D \cap \mathcal{D}(A) \text{ such that } \ \| x \|\geq R \ .$$

then (8) holds with respect to the set $D \cap B(0, R)$.

It is shown in [10] (see also [21]) that the operator $(A, \mathcal{D}(A))$, defined by (2)-(3), is the infinitesimal generator of an exponentially stable semigroup $(T(t))_{t\geq 0}$ in H. Therefore, there exist $\omega_0 > 0$ and $M \geq 1$ such that $\| T(t) \|\leq M e^{-\omega_0 t}$, for all $t \geq 0$. Let us define a new norm which is equivalent to the given norm in H by:

$$| \, x \, | := \sup\{ e^{\omega_0 t} \, \| \, T(t) x \, \| : \ t \geq 0 \} \ , \quad \text{for all } x \in H \ . \tag{11}$$

In view of Proposition 3.1, $(T(t))_{t\geq 0}$ is still a compact semigroup on H, with respect to this norm, such that $| \, T(t) \, | \leq e^{-\omega_0 t}$ (see e.g. [13, p.19]). Recall the definition of the closed set D:

$$\begin{aligned} D \ \ := \ \ & \Big\{ (x_1, x_2)^T \in H : -1 \leq x_1(z) \ \text{ and } 0 \leq x_2(z) \leq 1, \\ & \text{for almost all } z \in [0,1] \Big\} \ . \end{aligned} \tag{12}$$

Lemma 3.2 $D \cap \mathcal{D}(A)$ is dense in D .

The following result follows from Lemmas 3.1 , 3.2, and [10, Proposition 5.2, Lemma 3.1].

Lemma 3.3 For all $R > 0$ sufficiently large, there holds

$$\lim_{h \to 0^+} h^{-1} d\left(T(h)x + hN(x), D \cap B(0,R)\right) = 0 \quad \text{for all } \; x \in D \cap B(0,R) \; .$$

Theorem 3.3 The axial dispersion nonisothermal tubular reactor with nonlinear model, given by (1)-(5), has at least one equilibrium profile in D, i.e. equation (6) admits at least one solution.

Proof:
By the compactness of the semigroup $(T(t))_{t\geq 0}$ (see Proposition 3.1), (9) is obviously satisfied (see Remark 3.2). In view of Lemma 3.3, condition (8) holds on $D \cap B(0,R)$, for some $R > 0$ sufficiently large. Then the conclusion follows by applying Theorem 3.2 to the set $D \cap B(0,R)$, i.e. there exists $\phi \in D \cap \mathcal{D}(A) \cap B(0,R)$ such that $A\phi + N(\phi) = 0$. ■

Remark 3.3 Theorem 3.3 holds independently of the fact that the reactions are endothermic ($\delta < 0$) or exothermic ($\delta > 0$).

4 Multiplicity of the Equilibrium Profiles

In this section, the question of the multiplicity of solutions to the following equation is considered:

$$Ax + N(x) = 0 \; , \quad x = (x_1, x_2)^T \in D \cap \mathcal{D}(A) \; , \tag{13}$$

where the operators $(A, \mathcal{D}(A))$ and N, and the set D are given by (1)-(5). Since 0 is not in the spectrum of A, equation (13) is equivalent to

$$x = R(0, A)N(x), \quad x \in D \; . \tag{14}$$

The following lemma, concerning the multiplicity of fixed points for nonlinear operators on ordered Banach spaces, is the main tool in the analysis of multiple solutions of equation (14), briefly described below.

Lemma 4.1 *[15, Lemma 2.1, p.442]* Let $(X, \leq)$ be an ordered Banach space such that the positive cone $X^+ := \{w \in X \; : \; 0 \leq w\}$ has a non-empty interior. Moreover let $\eta : X^+ \to [0, +\infty)$ be a continuous and concave functional and let G be a compact mapping of $X^+_\tau := \{w \in X^+ : \parallel w \parallel \leq \tau\}$ into X^+ for some constant $\tau > 0$ such that

(i) $\parallel G(w) \parallel < \tau$ for all $w \in X^+_\tau$ such that $\parallel w \parallel = \tau$.

Assume that there exist constants $\tau_1 \in (0, \tau)$ and $\tau_2 > 0$ such that

(ii) the set $W = \{w \in int(X^+_\tau) : \eta(w) > \tau_2\}$ is not empty ,

(iii) $\parallel G(w) \parallel < \tau_1$ for all $w \in X^+_{\tau_1}$ such that $\parallel w \parallel = \tau_1$,

(iv) $\eta(w) < \tau_2$ for all $w \in X^+_{\tau_1}$, and

(v) $\eta(Gw) > \tau_2$ for all $w \in X^+_\tau$ such that $\eta(w) = \tau_2$.

Then the mapping G has at least three distinct fixed points .

Remark 4.1 Lemma 4.1 above is a slightly modified version of [20, Lemma 4.4, p. 568]. The proof of that result is based on fundamental ideas developed in [12], which are related to the concept of fixed point index and Schauder's fixed point theorem, see e.g. [2]. The proof of the main result of this section, viz. Theorem 4.1 below, goes along the lines of the proof of [15, Theorem 1]. However it is a nontrivial adaptation of that analysis to the model considered here.

Il is clear that lemma 4.1 is not applicable to the hilbert space H because the interior of its positive cone H^+ is empty. To overcome this technical difficulty, we choose as state space, $X = C[0,1] \times C[0,1]$ endowed with the norm

$$\| x \|_\infty \;=\; \max\big(\| x_1 \|_\infty, \| x_2 \|_\infty \big) , \quad \text{for all } x = (x_1, x_2)^T \in X .$$

Observe that X is an ordered Banach space, with positive cone X^+ given by

$$X^+ = C^+[0,1] \times C^+[0,1] ,$$

where $C^+[0,1]$ is the set of nonnegative (real-valued) continuous functions on $[0,1]$, and that X^+ has a nonempty interior. The set D will be also replaced by

$$\mathcal{D} := \Big\{ (x_1, x_2)^T \in X : \; x_1(z) \geq -1 \;\text{ and }\; 0 \leq x_2(z) \leq 1, \;\text{for all } z \in [0,1] \Big\}. \tag{15}$$

It is clear that $R(0, A_i)\big(C[0,1]\big) \subset C[0,1]$ (i=1,2), and recall that is an integral operator with continuous kernel [10, see proof of Lemma 5.1] which implies that $R(0, A_i)$ is compact operator of $C[0,1]$[18, Example 1. p.277]. Therefore $R(0, A) =: diag(R(0, A_1), R(0, A_2))$ is a compact operator of $C[0,1] \times C[0,1]$.
Let $G : X^+ \to \mathcal{D}(A) \cap X^+$ be given by

$$G(x) \;:=\; R(0, A)N(P(x)), \qquad x \in X^+, \tag{16}$$

where P is the projection operator on the closed convex set

$$\mathcal{D}_{\alpha e^\mu} := \Big\{ (x_1, x_2)^T \in X : \; x_1(z) \geq -1 \;\text{ and }\; 0 \leq x_2(z) \leq \alpha e^\mu, \;\text{for all } z \in [0,1] \Big\}.$$

Lemma 4.2 If $\alpha e^\mu < 1$ and $\delta > 0$, then the fixed points of G are in $\mathcal{D}_{\alpha e^\mu} \subset \mathcal{D}$, and consequently, are solutions of (13).

Remark 4.2 (a) In view of Lemma 4.2, in order to prove the multiplicity of solutions of equation (13), it is sufficient to show the multiplicity of solutions of equation (16).
(b) The projection operator P is uniquely determined by the following expression

$$P(x) = (x_1, P_2 x_2) , \quad (x_1, x_2)^T \in X ,$$

where P_2 is the projection operator of $C[0,1]$ onto the set $\Lambda_{\alpha e^\mu} = \{x_2 \in C[0,1] : 0 \leq x_2(z) \leq \alpha e^\mu, \;\text{ for all }\; z \in [0,1] \}$.

Let us define the functions ϕ_i $i = 1, 2$, the functional η, and the constant ϖ as follows:

$$\phi_i \;=\; R(0, A_i)\, 1\!\!1, \qquad i = 1, 2, \tag{17}$$

$$\eta : X^+ \;\to\; [0, +\infty) \tag{18}$$

$$x \;\to\; \min_{z \in [0,1]} x_1(z),$$

$$0 < \varpi \;=\; \eta(\phi) = \inf_{z \in [0,1]} \phi_1(z) , \qquad \text{where } \phi = (\phi_1, \phi_2). \tag{19}$$

It is clear that η is a continuous concave functional. By a simple variational analysis, one can easily prove the following result:

Lemma 4.3 If $\mu > 4$, the function $v(t) = t\exp(\frac{-\mu t}{1+t})$, $t \geq 0$, has a local maximum at $\tau_1(\mu) = \frac{(\mu-2)-\sqrt{\mu^2-4\mu}}{2}$ and a local minimum at $\tau_2(\mu) = \frac{(\mu-2)+\sqrt{\mu^2-4\mu}}{2}$.

The preliminary results above lead to the fact that all the assumptions of Lemma 4.1 are satisfied, whence the following theorem holds.

Theorem 4.1 For positive parameters $\alpha, \beta_1, \beta_2, \delta, \gamma, \mu$ such that $\mu > 4$, $\alpha e^{\mu} < 1$ and $\delta > 0$, the axial dispersion nonisothermal tubular reactor nonlinear model, given by (1)-(5), has at least three equilibrium profiles provided that

$$\varpi^{-1} v(\tau_2(\mu)) \quad < \alpha\delta(1-\alpha e^{\mu}) < \alpha \max(\delta, 1) < \quad \| \phi \|_{\infty}^{-1} v(\tau_1(\mu)) \ , \tag{20}$$

where the function $v(\cdot)$ and the real numbers $\tau_1(\mu)$ and $\tau_2(\mu)$ are defined in Lemma 4.3, the functions ϕ_i $i = 1, 2$ are given by (17) and the constant ϖ is defined by (18)-(19).

5 Concluding remarks

(a) Observe that $v(\tau_1(\mu)) \sim \frac{1}{\mu}\exp(\frac{-\mu}{1+\mu})$ and $v(\tau_2(\mu)) \sim \mu\exp(-\mu)$ as $\mu \longrightarrow +\infty$. Hence condition (20) is meaningful with respect to the model considered in this paper.

(b) In tubular reactor theory, the dynamics of tubular reactors are typically described by nonlinear PDE's derived from mass and energy balances principles, see e.g. [6, 10, 1]. In the axial dispersion nonisothermal tubular reactor nonlinear model (1)-(5) studied in this paper, the parameters α, β_1, β_2, δ, γ and μ are given by the following expressions

$$\begin{aligned} \alpha &= \frac{k_0 L}{v}\exp(-\frac{E}{RT_{in}}), \ \beta_1 = \frac{D_1}{vL}, \ \beta_2 = \frac{D_2}{vL} \\ \gamma &= \frac{4hL}{\rho C_p d v}, \ \delta = -\frac{\Delta H}{\rho C_p}\frac{C_{in}}{T_{in}}, \ \mu = \frac{E}{RT_{in}} \ , \end{aligned}$$

where D_1, D_2, v, ΔH, ρ, C_p, k_0, E, R, h, d, T_c, T_{in} and C_{in} hold for the energy and mass dispersion coefficients, the superficial fluid velocity, the heat of reaction, the density, the specific heat, the kinetic constant, the activation energy, the ideal gas constant, the wall heat transfer coefficient, the reactor diameter, the coolant temperature, the inlet temperature, and the inlet reactant concentration, respectively. τ, ζ and L denote the time and space independent variables, and the length of the reactor, respectively, see [10]. Hence, in Theorem 4.1, the conditions $\mu > 4$ and $\alpha e^{\mu} < 1$ correspond to the following conditions

$$\frac{E}{RT_{in}} > 4 \quad \text{and} \quad \frac{k_0 L}{v} < 1 \ .$$

Moreover the positivity condition for the parameter δ means that the reactions are assumed to be exothermic, i.e. $\Delta H < 0$.

(c) A further important question is to study the stability of these equilibrium profiles when they exist. However, although there is a strong convergence of results in the literature in the direction of an alternance of stable and unstable steady states in presence of multiple equilibrium points, the stability of the multiple steady states in the general non adabiatic reactor with different diffusion coefficients remains an open question.

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