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A NOTE ON NON-ISOTHERMAL DIFFUSION-REACTION PROCESSES

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Abstract: In this paper, a typical non-linear boundary value problem (NBVP) which arises as a mathematical descriptive model of several cases regarding catalytic chemical reactor design was considered. For such NBVP, a well known result on existence and uniqueness of solution was analyzed from a point of view of parametric dependence. In order to provide sufficient conditions for existence and uniqueness of solution, some results on parametric restriction were obtained. A number of NBVP were solved as illustrative examples.

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1. Introduction

Heterogeneous catalytic non-isothermal chemical reactors are very frequently involved in unit operations concerning a variety of chemical processes. Usually, in such devices a chemical process takes place when a gas or liquid component reacts on a metallic catalyst supported in a porous particle. Such phenomenon has been extensively studied concerning for example, the corresponding effectiveness factor η (See [1], for an overview); and numerical treatment looking for an approximate solution to the corresponding mathematical descriptive model (See e.g. [2], [3], [4], [5], and [6]).

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In [7] an analytical solution using a power series method, was obtained for a non-linear boundary value problems modeling diffusion reaction phenomena with a high non-linear kinetic.

Some results concerning the use of contraction mapping, energy and singularity methods, maximum principles, catastrophe and bifurcation, looking for to obtain sufficient conditions for uniqueness of solution in two-point non-linear boundary value problems as descriptive model of diffusion-reaction processes, were reported in [7], [8] and [9].

In the present study, we shall consider a porous slab of half thickness L, in which a reactive component A undergoes an irreversible non isothermal chemical reaction of order $(m,p) : mA + pB \to P$.

We assume steady state conditions and that all parameters are no dependent on the temperature T, mass concentration u and spatial coordinate x. Furthermore, we assume that external mass and heat transfer resistances are negligible.

Then, the following Boundary Value Problem (BVP) can be considered as a mathematical descriptive model for the process which concerns us:

$$\frac{d^2u}{dx^2} - \phi^2 f(u) = 0 \qquad \text{for} \qquad 0 < x < L \tag{1}$$

$$u = 1 \quad \text{at} \quad x = L, \quad \frac{du}{dx} = 0 \quad \text{at} \quad x = 0 \tag{2}$$

where the function u is defined as

$$u = \frac{C_A}{C_{A_S}} \tag{3}$$

being C_A the volumetric molar concentration of the key component A; and C_{A_S} its respective surface value. The function $f = f(u)$ represents a dimensionless reaction rate.

The parameter ϕ denotes the corresponding Thiele Modulus defined by

$$\phi = \sqrt{\frac{L^2 k_S C_{C_{A_S}}^{m-1} C_{B_S}^{P}}{D_A}} \tag{4}$$

where k_S, D_A and C_{B_S} denote specific kinetic constant, effective diffusivity coefficient and dimensional concentration of component B, respectively.

We remark that in our case, the temperature T and the concentration profile u are no longer independent. In fact, they satisfy the following relation (See [1]):

$$T = d(1-u) + 1 \tag{5}$$

where the parameter d is the thermicity of the reaction, defined by

$$d = \frac{D_A C_{A_S}(-\Delta H)}{kT_S} \tag{6}$$

in which $-\Delta H$ is the reaction heat; k, the effective thermal conductivity inside the porous slab; and T_S is the dimensional temperature at the external pellet surface.

The parameter d represents the deviation from isothermal conditions, being $d < 0$ and $d > 0$ for endothermic and exothermic reactions respectively. Now, we shall focus on the diffusionreaction process which is included in (1)(2) if we consider a non-linear dimensionless reaction rate given by the expression:

$$f(u) = u^{m+p}\exp\left[\frac{c(1-u)}{d(1-u)+1}\right] \tag{7}$$

where the parameter c is defined by

$$c = \gamma \cdot d \tag{8}$$

being γ the Arrhenius group given as

$$\gamma = \frac{E}{RT_S} \tag{9}$$

in which E is the activation energy; and R, the universal gas constant. Therefore, the corresponding Non-linear Boundary Value Problem (NBVP) to be analyzed is the following one:

$$\frac{d^2u}{dx^2} - \phi^2 u^{m+p}\exp\left[\frac{c(1-u)}{d(1-u)+1}\right] = 0 \qquad \text{for} \qquad 0 < x < L \tag{10}$$

$$u = 1 \quad \text{for} \quad x = L, \quad \frac{du}{dx} = 0 \quad \text{for} \quad x = 0 \tag{11}$$

As it can be seen in the literature on the subject (See e.g. [4]), the existence of at least one analytical solution, no solution, unique solution or a multiplicity of solutions for the mathematical model provided by equations (10) and (11) is strongly dependent on the domain of the governing parameters where the problem is analyzed. In such sense, in the cited reference, it can also be seen that the corresponding conclusions were obtained a posteriori, i.e., once the concentration profiles were evaluated.

The main aim of the present study was to develop an analysis from a point of view concerning parametric dependence on a well known basic result (See [7]), regarding the existence and uniqueness of solution of the model (10)-(11).

The more substantial outcome of this contribution consist in to provide a priori, some results given as specific restrictions on the governing parameters in order to obtain sufficient conditions to assure existence of a unique solution to the model (10)-(11). Clearly such results are potentially very useful as complementary information in order to help and orientate the final numerical treatment inherent to the obtention of approximations to the solution of the model.

In what follows it is convenient to treat a dimensionless model which is obtained from (10)-(11) using the following dimensionless variables z and y defined by

$$z = 1 - \frac{x}{L}, \quad y = 1 - u \tag{12}$$

The following equivalent dimensionless model was obtained

$$\frac{d^2y}{dz^2} + \phi^2(1-y)^{m+p}\exp\left[\frac{cy}{dy+1}\right] = 0 \qquad \text{for} \qquad 0 < z < 1 \tag{13}$$

$$y = 0 \quad \text{for} \quad z = 0, \quad \frac{dy}{dz} = 0 \quad \text{for} \quad z = 1. \tag{14}$$

2. Results and Analysis

To begin with, let us introduce the function $F = F(y)$ defined by

$$F(y) = \phi^2(1-y)^{m+p}\exp\left[\frac{cy}{dy+1}\right], \quad 0 \le y < 1 \tag{15}$$

Let F be a function so that it verifies the Lipschitz condition expressed as

$$|F(w) - F(v)| \le \overline{L}|w - v| \tag{16}$$

for all w, v belonging to the real numeric interval $[0, 1)$.Then, according to [7], whenever the following inequality is verified

$$\frac{\pi}{2\sqrt{\overline{L}}} > 1 \tag{17}$$

the NBVP (13)-(14) has one and only one solution.

2.1. Parametric Dependence

Whenever F is a continuous differentiable function on $0 \leq y < 1$, the Lipschitz constant $\overline{L}$ in the condition given by inequality (17) can be estimated as

$$\overline{L} = \sup_{0 \leq y < 1} \left| \frac{dF}{dy} \right| \tag{18}$$

Taking into account (15), it is clear that $\overline{L}$ can be obtained by using (18).

Conclusions of a systematic analysis on $\sup\limits_{0 \leq y < 1} |\frac{dF}{dy}|$, taking into account the following common general restriction on the parameters in model (13)-(14):

$$\begin{aligned} m = 0, 1, \ldots; \quad p \geq 0; \\ -1 < d < 1 \quad (d \neq 0), \\ 2 \leq \gamma \leq 30; \; 0.1 \leq \phi \leq 4 \end{aligned} \tag{19}$$

are illustrated in Table 1.

In order to estimate the Lipschitz constant $\overline{L}$ using equation (18), the corresponding information provided in Table 1 is used.

Then, introducing such estimation for $\overline{L}$ in inequality (17), specific restrictions defining respective regions for the governing parameters were obtained. Such restrictions represent sufficient conditions for the existence and uniqueness of the solution of the model (13)-(14), and they are ordered in Table 2.

2.2. Critical Point on Existence and Uniqueness Regions: Parametric Analysis

Concerning Table 2, let us denote by M the left hand side in inequalities (1)-(5), which provides the corresponding regions for existence and uniqueness of solution.

By critical point for M, we mean a set of values for parameters, which must result in

$$M = 1 \tag{20}$$

As a matter of practical application, the following specific cases are illustrated

$\underline{d < 0 \quad \text{(endothermic processes)}}$

$$m_0 = 0, \; p_0 = 0, \; \phi_0 = \frac{\pi}{2}, \; \gamma_0 = 2, \; d_0 = -\frac{1}{2} \tag{21}$$

$$M = \frac{\pi}{2\phi}\frac{1}{\sqrt{\gamma\cdot(-d)}},\ M(\phi_0,\gamma_0,d_0) = 1 \tag{22}$$

Since $\frac{\partial M}{\partial \phi} < 0, \frac{\partial M}{\partial \gamma} < 0, \frac{\partial M}{\partial d} > 0$, for all ϕ, γ, d (which verify the restrictions given by (19)) then, it follows that for any set of parameters belonging to Ω^0 given by

$$\Omega^0 \equiv \{m_0 = 0,\ p_0 = 0,\ 2 < \gamma \leq 30,\ d_0 = -\frac{1}{2},\ \frac{\pi}{2} < \phi \leq 4\} \tag{23}$$

as M results to be less than 1; when looking for the numerical treatment of the model (13)-(14) in Ω^0, a non unique solution can be expected.

While that, being $\frac{\partial M}{\partial d} > 0$, it follows that solving (13)-(14) in the region Ω^1 given by

$$\Omega^1 \equiv \{m_0 = 0,\ p_0 = 0,\ \phi_0 = \frac{\pi}{2},\ \gamma_0 = 2,\ -\frac{1}{2} < d < 0\} \tag{24}$$

since M results to be greater than 1, a unique solution must be obtained.

$\underline{d > 0 \quad \text{(exothermic processes)}}$

$$m_0 = 0,\ p_0 = 0,\ \phi_0 = \frac{\pi}{2},\ \gamma_0 = 10,\ d_0 = 0.1 \tag{25}$$

$$M = \frac{\pi}{2\phi}\frac{1}{\sqrt{\gamma\cdot d}},\ M(\phi_0,\gamma_0,d_0) = 1 \tag{26}$$

In this case, in view that $\frac{\partial M}{\partial d} < 0, \frac{\partial M}{\partial \phi} < 0$ it follows that solving (13)-(14) for Ω^2 given by

$$\Omega^2 \equiv \{m_0 = 0,\ p_0 = 0,\ \frac{\pi}{2} < \phi \leq 4,\ \gamma_0 = 10,\ 0.1 < d < 1\}$$

a non unique solution can be expected.

In view of industrial applications (See [4]) it is interesting to find a critical point for M within the following region for the parameters m_0, p_0, ϕ, d and γ given by

$$m_0 = 1,\ p_0 = 0,\ \phi < 0.16,\ 0 < d \leq 0.7,\ 2 \leq \gamma < 20 \tag{27}$$

Let the Thiele Modulus ϕ and thermicity d given as $\phi = 0.1$, $d_0 = 0.7$. In such case, from (3) of Table 2 it follows that M is given as

$$M = \frac{\pi}{2\phi}\frac{1}{\sqrt{\exp\left(\frac{\gamma\cdot d}{1+d}\right)}} = \frac{5\pi}{\sqrt{\exp\left(\frac{0.7\gamma}{1.7}\right)}} \tag{28}$$

Direct calculation on (28) leads to conclude that there exists a unique γ_0 with $10 < \gamma_0 < 15$, so that

$$M(\phi_0, d_0, \gamma_0) = M(0.1, 0.7, \gamma_0) = 1.$$

Then, in view that $\frac{\partial M}{\partial \phi} < 0,\ \frac{\partial M}{\partial \gamma} < 0,\ \frac{\partial M}{\partial d} < 0$, it follows that solving (13)-(14) in the region Ω^3 given by

$$\Omega^3 \equiv \{m_0 = 1,\ p_0 = 0,\ 0.1 < \phi \leq 4,\ 0.1 < d < 1,\ \gamma_0 \leq \gamma \leq 30\} \qquad (29)$$

a non unique solution can be expected. In fact, as an interesting illustrative case, we refer to (See [4]), in which three concentration profiles were founded, corresponding to the following set of values for parameters m, p, ϕ, d and γ

$$m = 1,\ m = 0,\ \phi = 0.16,\ d = 0.7,\ \gamma = 20 \qquad (30)$$

2.3. Examples

The NBVP (13)-(14) was solved by using some parametric sets belonging in each case to the respective existence and uniqueness regions consigned by the inequalities in Table 2. In such table, each parametric set was identified as (1),(2),...,(5). Also, the information on existence and uniqueness of solution provided in the preceding section, was taken into acount. The resulting concentration profiles and the effectiveness factor η, corresponding to the inequalities (1),(2),(3),(4) and (5) in table 2, are illustrated in tables 3,4,5,6 and table 7 respectively.

3. Conclusion

In conclusion, a conceptually simple procedure in order to provide valuable information a priori, to orientate the corresponding numerical treatment when looking for approximations to the solution of the NVBP in analysis was developed. In fact, the consigned procedure leads to deduce mathematical inequalities which provide the corresponding regions for the parameters of the process in order to have existence and uniqueness of solution.

Furthermore, since the critical point defined for the function M introduced at the beginning of Section 3.2, does not verify a condition of local extremum; then, for each existence and uniqueness region, a corresponding region can be deduced, where non uniqueness of solution can probably be expected.

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Appendix A

Assumed parametric sets	$\sup\limits_{0\le y<1} \lvert \frac{dF}{dy} \rvert$
$m = 0,\ p = 0,\ d < 0$	$-\gamma \cdot d \cdot \phi^2$
$m = 1,\ p = 0,\ -\frac{1}{2} < d < 0$	$\phi^2(1 - \gamma d^2)$
$m > 1,\ p = 0,\ d > 0$	$\gamma \cdot d \cdot \phi^2$
$m = 1,\ p = 1,\ d > 0,\ 0 < \gamma \cdot d < 1$	$\phi^2 \exp(\frac{\gamma \cdot d}{1+d})$
$m = 1,\ p = 0,\ d > 0$	$\phi^2 \exp(\frac{\gamma \cdot d}{1+d})$

Table 1: Parametric dependence of $\sup\limits_{0\le y<1} \lvert \frac{dF}{dy} \rvert$

Assumed parametric sets	**Regions for existence and uniqueness of solution**
$m = 0,\ p = 0,\ d < 0,$ $\gamma \cdot d = c < 0$	$\frac{\pi}{2\phi} \frac{1}{\sqrt{\gamma(-d)}} > 1,\ (1)$
$m = 0,\ p = 0,\ d > 0,$ $\gamma \cdot d = c > 0$	$\frac{\pi}{2\phi} \frac{1}{\sqrt{\gamma d}} > 1,\ (2)$
$m = 1,\ p = 0,\ d > 0$	$\frac{\pi}{2\phi} \frac{1}{\sqrt{\exp(\frac{\gamma \cdot d}{1+d})}} > 1,\ (3)$
$m = 1,\ p = 0,\ -\frac{1}{2} < d < 0,$ $\gamma \cdot d = c < 0$	$\frac{\pi}{2\phi} \frac{1}{\sqrt{1-\gamma d}} > 1,\ (4)$
$m = 1,\ p = 1,\ d > 0,$ $0 < \gamma \cdot d < 1$	$\frac{\pi}{2\phi} \frac{1}{\sqrt{\exp(\frac{\gamma \cdot d}{1+d})}} > 1,\ (5)$

Table 2: Parametric dependence of existence and uniqueness of solution for (13)-(14)

					x			
c	d		0.0	0.25	0.5	0.75	1.00	η
		$y(x)$	0.000000	0.045077	0.076180	0.094425	0.100440	
-2.5	-0.5							0.8412
		$y'(x)$	0.210309	0.151436	0.098128	0.048247	0.000000	
		$y(x)$	0.000000	0.039354	0.065829	0.081121	0.086125	
-5	-0.5							0.7461
		$y'(x)$	0.186524	0.130197	0.082705	0.040216	0.000000	
		$y(x)$	0.000000	0.035385	0.058706	0.072018	0.076351	
-7.5	-0.5							0.6797
		$y'(x)$	0.169935	0.115592	0.072300	0.034867	0.000000	
		$y(x)$	0.000000	0.041559	0.069825	0.086265	0.091662	
-4	-0.2							0.7827
		$y'(x)$	0.195676	0.138396	0.088689	0.043343	0.000000	
		$y(x)$	0.000000	0.039551	0.066203	0.081619	0.086668	
-5	-0.2							0.7493
		$y'(x)$	0.187313	0.130967	0.083330	0.040565	0.000000	

Table 3: Concentration profiles and effectiveness factors corresponding to case (1) in table 2, for $m = 0,\ p = 0,\ \phi = 0.5$

					x			
c	d		0.0	0.25	0.5	0.75	1.00	η
		$u(x)$	0.000000	0.074132	0.129628	0.164013	0.175663	
2.5	0.5							1.3195
		$u'(x)$	0.329878	0.261135	0.181163	0.092819	0.000000	
		$u(x)$	0.000000	0.068637	0.119455	0.150699	0.161243	
2.0	0.4							1.2294
		$u'(x)$	0.307348	0.240258	0.165102	0.084091	0.000000	
		$u(x)$	0.000000	0.060524	0.104481	0.131152	0.140093	
1.0	0.2							1.0961
		$u'(x)$	0.274027	0.209527	0.141647	0.071423	0.000000	
		$u(x)$	0.000000	0.057394	0.098722	0.123652	0.131985	
0.5	0.1							1.0446
		$u'(x)$	0.261149	0.197705	0.132693	0.066616	0.000000	
		$u(x)$	0.000000	0.055994	0.096149	0.120306	0.128369	
0.25	0.05							1.0215
		$u'(x)$	0.255384	0.192425	0.128708	0.064482	0.000000	

Table 4: Concentration profiles and effectiveness factors corresponding to case (2) in table 2, for $m = 0,\ p = 0,\ \phi = 0.5$

					x			
c	d		0.0	0.25	0.5	0.75	1.00	η
		$y(x)$	0.000000	0.059654	0.102825	0.128938	0.137675	
2.0	0.4							1.0820
		$y'(x)$	0.270511	0.20613	0.138858	0.069836	0.000000	
		$y(x)$	0.000000	0.054316	0.093042	0.116241	0.123967	
1.0	0.2							0.9940
		$y'(x)$	0.248500	0.186047	0.123807	0.061822	0.000000	
		$y(x)$	0.000000	0.052090	0.088971	0.110966	0.118275	
0.5	0.1							0.9572
		$y'(x)$	0.239311	0.177691	0.117576	0.058516	0.000000	
		$y(x)$	0.000000	0.051065	0.087099	0.108542	0.11566	
0.25	0.05							0.9403
		$y'(x)$	0.235078	0.173848	0.114717	0.057002	0.000000	
		$y(x)$	0.000000	0.050476	0.086023	0.107149	0.114157	
0.1	0.02							0.9306
		$y'(x)$	0.232642	0.171638	0.113076	0.056134	0.000000	
		$y(x)$	0.000000	0.050130	0.085393	0.106333	0.113278	
0.01	0.002							0.9249
		$y'(x)$	0.231215	0.170345	0.112116	0.055626	0.000000	
		$y(x)$	0.000000	0.050096	0.085330	0.106253	0.113191	
0.001	0.0002							0.9243
		$y'(x)$	0.231074	0.170217	0.112021	0.055576	0.000000	

Table 5: Concentration profiles and effectiveness factors corresponding to case (3) in table 2, for $m = 1,\ p = 0,\ \phi = 0.5$

					x			
c	d		0.0	0.25	0.5	0.75	1.00	η
		$u(x)$	0.000000	0.048985	0.083302	0.103629	0.110361	
-0.3	-0.1							0.9059
		$u'(x)$	0.226478	0.166054	0.108934	0.053945	0.000000	
		$u(x)$	0.000000	0.048288	0.082031	0.101987	0.108591	
-0.5	-0.1							0.8944
		$u'(x)$	0.223595	0.163446	0.107005	0.052927	0.000000	
		$u(x)$	0.000000	0.046673	0.079091	0.098191	0.104500	
-1.0	-0.1							0.8676
		$u'(x)$	0.216908	0.157412	0.102557	0.050587	0.000000	
		$u(x)$	0.000000	0.045214	0.076442	0.094775	0.100821	
-1.5	-0.1							0.8435
		$u'(x)$	0.210863	0.151975	0.098568	0.048494	0.000000	
		$u(x)$	0.000000	0.043888	0.074037	0.091678	0.097487	
-2.0	-0.1							0.8214
		$u'(x)$	0.205359	0.147040	0.094963	0.046609	0.000000	

Table 6: Concentration profiles and effectiveness factors corresponding to case (4) in table 2, for $m = 1,\ p = 0,\ \phi = 0.5$

Appendix B

Nomenclature

A, B : Chemical reactive components in a chemical reaction.
C_A, C_{A_S} : Volumetric molar concentration of the key component A and its respective surface value $(mol \cdot m^{-3})$.
C_{B_S} : Surface value for volumetric molar concentration C_B of chemical reactive component B.
c : Parameter defined by equation (8).
d : Parameter defined by equation (6). It represents the deviation from isothermal conditions.
D_A : Effective diffusivity coefficient of chemical component A $(m^2 s^{-1})$.
E : Activation energy $(J \cdot mol^{-1})$.
f : Scalar real function defined by equation (7). It represents a non-linear dimensionless reaction rate.
F : Scalar real function defined by equation (15).
k : Effective thermal conductivity inside the porous slab $(Wm^{-1}\,{}^{\circ}K^{-1})$

					x			
c	d		0.0	0.25	0.5	0.75	1.00	η
		$y(x)$	0.000000	0.013657	0.023410	0.029260	0.031210	
2.0	0.4							0.9991
		$y'(x)$	0.062441	0.046818	0.031203	0.015599	0.000000	
		$y(x)$	0.000000	0.013358	0.022862	0.028549	0.030442	
1.0	0.2							0.9793
		$y'(x)$	0.061209	0.045694	0.030361	0.015150	0.000000	
		$y(x)$	0.000000	0.013216	0.022601	0.028210	0.030076	
0.5	0.1							0.9699
		$y'(x)$	0.060620	0.045157	0.029959	0.014936	0.000000	
		$y(x)$	0.000000	0.013146	0.022473	0.028044	0.029897	
0.25	0.05							0.9653
		$y'(x)$	0.060332	0.044895	0.029763	0.014831	0.000000	
		$y(x)$	0.000000	0.013104	0.022397	0.027946	0.029791	
0.1	0.02							0.9626
		$y'(x)$	0.060161	0.044739	0.029646	0.014770	0.000000	
		$y(x)$	0.000000	0.013080	0.022352	0.027887	0.029727	
0.01	0.002							0.9609
		$y'(x)$	0.060059	0.044646	0.029577	0.014733	0.000000	
		$y(x)$	0.000000	0.013077	0.022347	0.027881	0.029721	
0.001	0.0002							0.9608
		$y'(x)$	0.060049	0.044637	0.02957	0.014729	0.000000	

Table 7: Concentration profiles and effectiveness factors corresponding to case (5) in table 2, for $m = 1,\ p = 1,\ \phi = 0.5$

k_S Specific kinetic constant.
L : Half thickness of porous slab (m).
$\overline{L}$: Lipschitz constant introduced by ineq. (16).
M : Composite parameter introduced in Section 3.2
m, p : Natural numbers which denote reaction order.
P : Product chemical component in a chemical reaction.
R : Universal gas constant.
T : Function which represents temperature profile in the porous slab ($^\circ K$).
$u = u(x)$: Dimensionless mass concentration of reactive component A.
v, w : Dimensionless functions.
x : Spatial coordinate (m).
z : Dimensionless spatial coordinate defined by eq. (12).

Sub index

$_0$: Specific value for parameters.

Greek letters

γ : Arrhenius group defined by eq. (9).

η : Effectiveness factor for the heterogeneous chemical reaction under analysis.

ϕ : Thiele Modulus defined by equation (4).