STABILITY OF A CLASS OF NONLINEAR STIRRED TANK REACTOR

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Abstract

We consider a continuous stirred tank reactor containing n chemical or biological species X_i ; we suppose that this reactor is isothermal (it can be also the zero dynamics of a thermal reactor if we are able to control the heat transfer and measure the temperature T in the reactor) ; the basic reactions are of the form $X_i \to X_j$ and the kinetics law of this reaction depends only of X_i, but is nonlinear ; we give some sufficient conditions giving the global asymptotic stability of the equilibrium. The kinetics laws can depend on the temperature, on the time, and be partially unknown ; they are not necessarily increasing with respect to X_i, but are rather subject to "positivity" with respect to the equilibrium.

1 Introduction

A continuous stirred tank reactor (CSTR) is a reactor, of constant volume, containing an homogeneous mixture of chemical or biochemical species, with equal rates of inflow and outflow [1, 2, 3] . Frequently, the state variables are the concentrations of the components in the reactor, and the temperature ([1, 7]).

Of course, the study of the dynamics and stability of the mathematical system modelling the CSTR is of fundamental importance ; it is well known ([15, 7]) that a CSTR can have complicated dynamics (multiple equilibria, limit cycles or more complex attractors).

In this paper, we will study isothermal CSTR, e.g. the temperature T is constant. This approximation is often very good in some reactors (bioreactors) where the heat produced by the reactions is negligible. The study of the dynamics of isothermal CSTR is also very important in the framework of input-output linearization methods, where the control is the heat tranfer and the output is the temperature ([12]). The zero dynamics of an open thermal stirred tank reactor is the dynamics of the isothermal CSTR, and has to be asymptotically stable to apply input-output methods.

This study has also close links with the mathematical theory of compartmental systems ([6]) where there are flows of material between compartments, and input and output flows.

More precisely, we consider here a CSTR, with a constant temperature T, and n components $X_1, ...X_n$ of concentrations $x_1, ...x_n$. We suppose that the reactions are of the form:

$$X_i \to X_j$$

and we call x_i^{in} the constant inflow for the i-th component. We denote by $f_{j,i}$ the kinetics law (the rate) of the reaction $X_i \to X_j$, and we remark that, in this reaction, the rate of consumption of X_i is the same as the production rate of X_j. If D is the constant dilution rate, we can write the ordinary differential equation :

$$\dot{x}_i = \sum_{j \neq i} (-f_{ji} + f_{ij}) + D(x_i^{in} - x_i) \ \ (i = 1, ...n) \qquad (1)$$

Finally we suppose that the rate of reaction f_{ji} depends only on the component consumed ("donor-controlled", [6]), i.e.

$$f_{ji} = f_{ji}(t, T, x_i)$$

where t is the time and T the temperature, which is supposed constant but may be not precisely known. The fact that the reaction depends only on the reactant and not on the product is a natural hypothesis related to causality. Of course, if there is no reaction, then $f_{ji} \equiv 0$. We suppose also that the functions are regular enough (say C^1 with respect to the variables), so that the differential system admits one unique solution.

We have the following constraints, that are due to the physical meaning of the system :

- the state variables x_i and the inflows x_i^{in} are nonnegative
- the reaction rates f_{ji} are nonnegative
- at least one of the x_i^{in} is positive, and D is positive.

Now we are interested by the stability of the equilibria of this system. Usually, one studies the existence of the equilibria (section 2), then, under the hypothesis of increasingness of the functions f_{ji} with respect to x_i, one proves the global asymptotic stability of the equilibrium (section3). We prove here the stability under a weaker hypothesis (section 4).

2 The equilibria

With the above hypotheses, the system is $(i = 1, ..., n)$:

$$\dot{x}_i = \sum_{j \neq i} (-f_{ji}(t, T, x_i) + f_{ij}(t, T, x_j)) + D(x_i^{in} - x_i) \quad (2)$$

We recall now some well known facts concerning the positivity and boundedness of variables ([6, 7]) ; to ensure the positivity of the variables, we make, in all the paper, the (realistic) assumption that ;

$$f_{ji}(t, T, 0) = 0$$

that means that if there is no reactant, there is no reaction. Then we can verify that the vector field is repulsive at the boundary $x_i = 0$, and therefore the nonnegative closed orthant $x_i \geq 0$ $(i = 1, ...n)$ is invariant. Moreover the following relation is obtained by addition of the equations :

$$\sum_{i=1}^{n} \dot{x}_i = D(\sum_{i=1}^{n} x_i^{in} - \sum_{i=1}^{n} x_i)$$

This linear equation shows that the variables converge towards the set H (intersection of an hyperplane and of the nonnegative space) :

$$\sum_{i=1}^{n} x_i = \sum_{i=1}^{n} x_i^{in}$$

therefore the variables x_i are ultimately bounded above by $\sum_{i=1}^{n} x_i^{in}$.

Because H is invariant, convex and compact, then, by the Brouwer theorem, there exists at least one nonnegative equilibrium in H. The problem is to study the dynamics and stability of these equilibria on H ; more precisely, the want to find conditions ensuring the global asymptotic stability of the equilibrium in the nonnegative space.

3 Known results

If the kinetics of the reaction follow the mass action law, then $f_{ji}(t, T, x_i) = k_{ji} x_i$ (k_{ji} being a positive constant) , and the differential system is linear. The theory of Feinberg, Horn, and Jackson ([5]) give the global asymptotic stability of the equilibrium, but it is not necessary to invocate such a theory ; simple properties of (-M)-matrices (matrices that are positive outside the main diagonal), intimately linked with the Perron-Froebenius theorem, give also the result ([4]) by proving the stability of the matrix.

When the kinetics is not linear, several results are available. Rouchon ([12]) has shown, using Rosenbrock's theorem([9]), that the system is globally asymptotically stable (GAS) if the functions f_{ij} are such that:

$$f_{ji}(t, T, x_i) = k_{ji}(T) \phi_{ji}(x_i)$$

where ϕ is an increasing function such that $\phi(0) = 0$.

It is also possible to use the results of the theory on cooperative systems ([14]); indeed, if the system is autonomous $(f_{ji}(t, T, x_i) = f_{ji}(T, x_i))$ and if the functions f_{ij} are increasing with respect to x_i, then the Jacobian matrix has nonnegative off-diagonal elements, and the system is cooperative. Moreover, H is invariant ; to study the system on H is therefore equivalent to study, on H, the following system having a first integral $\sum_{i=1}^{n} x_i = c$:

$$\begin{aligned} \dot{x}_i &= \sum_{j \neq i} (-f_{ji} + f_{ij}) + D(x_i^{in} - x_i) \quad (i = 1, ...(n-1)) \\ \dot{x}_n &= \sum_{j \neq n} (-f_{jn} + f_{nj}) - D \sum_{j=1}^{n-1} (x_j^{in} - x_j) \end{aligned}$$

We can apply a result of [8] concerning the behaviour of cooperative systems with a first integral, that gives the global convergence on H towards the equilibrium, under the hypothesis that the Jacobian matrix is everywhere off-diagonal positive.

4 Global asymptotic stability for positive functions

The above results use the hypothesis that the functions f_{ij} are monotone increasing. We will impose a weaker assumption on these functions, related to positivity. Let us recall that there exists (at least) one nonnegative equilibrium x^* ; in fact, this equilibrium can depend on T, but we keep this notation.

Assumption 1 :

$$\forall (t, T, x_i) \ \ (f_{ji}(t, T, x_i) - f_{ji}(t, T, x_i^*))(x_i - x_i^*) \geq 0$$

i.e., the functions $f_{ji}(t, T, x_i)$ are such that their graph is in the first and third quadrant with respect to $f_{ji}(t, T, x_i^*)$.

This assumption is commented at the end of this section.

Theorem 1 *Under the above assumption, the equilibrium x^* is GAS in the nonnegative space.*

Proof : We denote

$$\frac{(f_{ji}(t,T,x_i) - f_{ji}(t,T,x_i^*))}{(x_i - x_i^*)} = k_{ji}(t,T,x_i,x_i^*)$$

which is nonnegative by assumption. This is nothing but the "mean slope with respect to x^*". Of course, if $x_i = x_i^*$, we write by continuity $k_{ji}(t,T,x_i^*,x_i^*) = f'_{ji}(t,T,x_i^*)$ where f' is the derivative of f.

Because x^* is an equilibrium, we can write the system (2) :

$$\begin{aligned} \dot{x_i} &= \sum_{j\neq i}(-f_{ji}(t,T,x_i) + f_{ji}(t,T,x_i^*)) \\ &\quad + \sum_{j\neq i}(f_{ij}(t,T,x_j) - f_{ij}(t,T,x_j^*)) - Dx_i + Dx_i^* \end{aligned}$$

for $i = 1, ..., n$. If we factorize the $(x_i - x_i^*)$, we obtain

$$\dot{x} = A(t,T,x,x^*)(x - x^*) \tag{3}$$

where the square matrix A with elements a_{ij} is such that:

$$a_{ii} = -\sum_{j\neq i} k_{ji} - D$$

and for $i \neq j$

$$a_{ij} = k_{ij}$$

Because of the assumption, we remark that $a_{ij} \geq 0$.

We take now the following Lyapunov function, which is the absolute-value norm ; such a function has been used in, for example, [9, 10, 13]:

$$V(x) = \sum_{i=1}^{n} |x_i - x_i^*|$$

Of course, this function is not derivable for $x_i = x_i^*$, and, following [13] we use the right-hand derivative with respect to time $\frac{d^+}{dt}V(x(t))$ and define the following operator :

$$\sigma_i = \begin{cases} 1 \text{ if } x_i(t) > x_i^* \text{ or if } x_i(t) = x_i^* \text{ and } \dot{x}_i(t) > 0 \\ 0 \text{ if } x_i(t) = x_i^* \text{ and } \dot{x}_i(t) = 0 \\ -1 \text{ if } x_i(t) < x_i^* \text{ or if } x_i(t) = x_i^* \text{ and } \dot{x}_i(t) < 0 \end{cases}$$

Now we can take the right derivative of $V(x(t)) = \sum_{i=1}^{n} \sigma_i(x_i - x_i^*)$:

$$\begin{aligned} \frac{d^+}{dt}V(x(t)) &= \sum_{i=1}^{n} \sigma_i \dot{x}_i \\ &= \sum_{i=1}^{n} \sigma_i \sum_{j=1}^{n} a_{ij}(x_j - x_j^*) \\ &= \sum_{j=1}^{n} \sigma_j a_{jj}(x_j - x_j^*) + \sum_{j=1}^{n}\sum_{i\neq j} \sigma_i a_{ij}(x_j - x_j^*) \\ &\leq \sum_{j=1}^{n}(-D - \sum_{i\neq j} k_{ij})|x_j - x_j^*| + \sum_{j=1}^{n}\sum_{i\neq j} k_{ij}|x_j - x_j^*| \\ &\leq -D\sum_{j=1}^{n}|x_j - x_j^*| \\ &\leq -D\, V(x(t)) \end{aligned}$$

Using differential inequalities ([11]), we deduce easily that V is a Lyapunov function and that x^* is GAS for the nonnegative space.

We can comment now the assumption 1. We remark that this assumption says in fact that the functions $f_{ji}(t,T,x_i)$ are "positive" with respect to the point $(x_i^*, f_{ji}(t,T,x_i))$.. We illustrate this point with the figure 1, where the positivity hypothesis is fulfilled for the two points x^* and x^{**}. The functions f can be not increasing : this situation can appear in some chemical or biochemical situations. For example, for enzymatic reactions, there can be substrate inhibition [2][p. 115], where the function is decreasing after some concentration. Moreover, these reaction rates are often partially unknown, at least for some region. Therefore it is interesting to have a qualitative criterion of stability as weak as possible. The rates can also depend on the time and temperature, giving a rather broad class of admissible reaction kinetics leading to stability. One remark yet is that one has to know a priori the (approximate) value of the equilibrium.

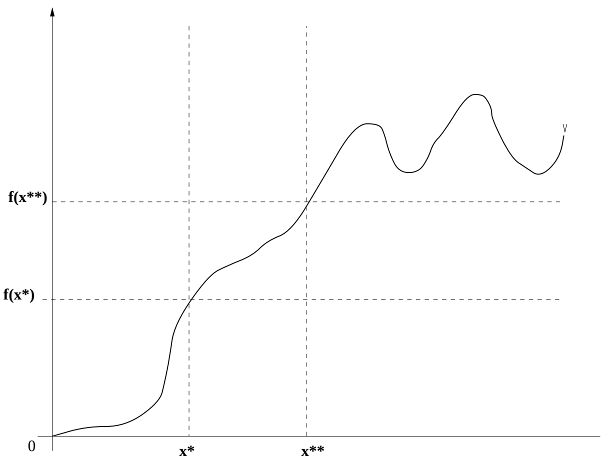

Figure 1: Condition of positivity for $f(x)$ with respect to $(x^*, f(x^*))$.

5 Conclusions

Our main assumption is more related to positivity than to monotony; of course, it is trivially verified if the function are monotone increasing, as it is usually assumed.

The fact that the total inflow and outflow is positive (the system is open) is also very important. If the system were closed, there could be, for example, non isolated equilibria.

The hypothesis concerning the reactions $X_i \to X_j$ is very strong ; in many chemical reactors, one can find reactions $X_i + X_j \to ....$, where the kinetics laws depend on x_i and x_j ([1, 3]). It is possible that the tools developped here apply to this more complex case.

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