

Catalisis, cinética y reactores



DYNAMIC STABILITY IN AN ENDOREVERSIBLE CHEMICAL REACTOR ESTABILIDAD DINÁMICA DE UN REACTOR QUÍMICO ENDOREVERSIBLE

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Abstract

This article presents a local stability study for an endoreversible Chemical Reactor (ECR). The model consists of two particle reservoirs, one at high chemical potential μ_1 and the other at low chemical potential μ_2 (the terms “high” and “low” refer to the situation in which $\mu_1 > \mu_2$). There are two particle resistors (in this work for simplicity we take $h_1 = h_2 = h$), that restrict the particle flows N_1 and N_2 . The net result is a particle flow drop from N_1 to an intermediate particle reservoir N_3 and from N_4 to N_2 . From the local stability analysis it was concluded that the ECR is stable for every value of h , C_1 and $\tau = n_x/n_y$. After a small perturbation, the system declines to the steady state with two different relaxation times both being proportional to C , h and τ . Finally, when the power output in the steady state versus τ is plotted, it demonstrates how an increment of τ can improve the system stability. This suggests a compromise between the stability and the engine energetic properties driven by τ .

Keywords: endoreversible, chemical reactor, stability, steady state, power output.

Resumen

En este trabajo se presenta un análisis de estabilidad local de un Reactor Químico. El modelo consiste de dos almacenes de partículas uno a un potencial químico alto μ_1 y el otro a un potencial químico bajo μ_2 (los términos “alto” y “bajo” se refieren a la situación en la que $\mu_1 > \mu_2$). Adicionalmente, existen dos resistencias (en este trabajo por simplicidad tomamos $h_1 = h_2 = h$), que restringen los flujos de las partículas N_1 y N_2 . El resultado neto es una caída en el flujo de partículas desde N_1 hacia un almacén intermedio de partículas N_3 y de N_4 a N_2 . A partir del análisis de estabilidad local, se puede concluir que el ECR es estable para cada valor de h , C_1 y $\tau = n_x/n_y$. Después de una pequeña perturbación, el sistema decae hacia un estado estacionario con cualquiera dos tiempos de relajación diferentes ambos proporcionales a C , h y τ . Finalmente, cuando graficamos la potencia de salida en el estado estacionario contra τ , encontramos que un incremento de τ puede mejorar la estabilidad del sistema. Esto sugiere un compromiso entre la estabilidad del sistema y las propiedades energéticas del mismo manejados por τ .

Palabras clave: endoreversibilidad, reactor químico, estabilidad, estado estacionario, potencia de Salida.

1 Introduction

Recent studies on heat engines and heat pumps, Santillan *et al.* (2001), Guzman-Vargas *et al.* (2005), Chimal-Eguía *et al.* (2006), Páez-Hernández *et al.* (2006), Huang *et al.* (2007), Chimal-Eguía *et al.* (2007) Nie *et al.* (2008), Huang *et al.* (2008), Huang *et al.* (2009), Páez-Hernández (2008), Santillán *et al.* (2008), Kitano (2004), Angulo-Brown *et al.* (1995), have demonstrated that the systems studied by means of finite time thermo-dynamics should fulfill at least two characteristics in order to be considered well

designed. These two different sorts of good design principles are Díaz-Hernández *et al.* (2010):

- a) The Optimal Thermodynamic Properties.
- b) The Dynamical Robustness of the system.

The second shows the important fact that the system stability should be allowed to maintain all its operation in some internal or external regime despite the perturbations that could occur. On the

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other hand, the optimal thermodynamic properties, namely, high power output, high efficiency, low entropy production, etc., guarantees the robustness of the energy converting system. Moreover, it has been shown by Santillan *et al.* (2001), Guzman-Vargas *et al.* (2005), Chimal-Eguía *et al.* (2006), Páez-Hernández *et al.* (2006), Huang *et al.* (2007), Chimal-Eguía *et al.* (2007), Nie *et al.* (2008), Huang *et al.* (2008), Huang *et al.* (2009), He J. *et al.* (2010), Nie W. *et al.* (2007), Páez-Hernández *et al.* (2011), Páez-Hernández *et al.* (2012), Wu, X.H. *et al.* (2011), Wu, X.H. *et al.* (2012), Wu, X.H. *et al.* (2015), Páez-Hernández *et al.* (2008), Santillán *et al.* (2008), Kitano (2004), Angulo-Brown *et al.* (1995), Villanueva-Marroquin *et al.* (2009) that these two physical properties inherently contain a certain kind of trade-off due to the fact that both are often governed by the same parameters Díaz-Hernández, (2010) *et al.* The former is evident such that while the stability becomes stronger the thermodynamic properties become weaker, Santillan *et al.* (2001), Guzman-Vargas *et al.* (2005), Chimal-Eguía *et al.* (2006), Páez-Hernández *et al.* (2006), Huang *et al.* (2007), Chimal-Eguía *et al.* (2007), Páez-Hernández *et al.* (2011), thus, it is clear that both properties must be adjusted in order to have the best possible system design.

In specialized literature similar studies sharing similar properties which are not physical models can be found. In agreement with the notion of well designed systems one can find both artificial and biological systems, for instance, stretch-reflex regulatory pathway Páez-Hernández *et al.* (2008), Santillán *et al.* (2008) or the simple Brownian motor confirm all previous hypotheses.

One more idea in the same sense was found by Díaz-Hernández *et al.* (2010), analysing the stationary thermodynamic properties and the stability for an enzymatic reaction. The model couples an endothermic with an exothermic reaction via an enzyme. Díaz-Hernández, (2010) *et al.* concludes that the thermodynamic properties improve as the chemical potential gap increases, while the steady state-relaxation rate decreases, so the chemical potential gap acts, in this case, as the parameter that represents a trade-off between the thermodynamic and dynamic properties of an enzyme reaction.

This present work is a study of a chemical endoreversible engine. As has been noted, this type of analysis is made for physical systems (heat engines), biological (enzymatic reactions), but not for chemical engines. Thus, as the first step to demonstrate if the aforementioned system shows the properties of good

design shared by other systems, the dynamic stability in an isothermal molecular machine operating along a one-dimensional reaction coordinate is analyzed.

The paper is organized as follows: Section 2 introduces a model for an endoreversible chemical reactor. Section 3 presents the stability analysis of the endoreversible chemical reactor. Section 4 discusses some numerical results and finally, Section 5 gives some concluding remarks.

2 Endoreversible models for chemical reactors

When one thinks about heat engines, using the classical thermo-dynamics approach, our thinking usually is related with the Reversible Carnot Cycle. However, in the thermo-dynamical world a reversible heat engine is utopia. With this in mind Rubin, (1979) defined a class of irreversible heat engines called “endoreversible heat engines”. He thought that all irreversibilities have to be restricted from the engine couplings to the external world, while the working fluid of the cited engine undergoes only reversible transformations.

The simplest endoreversible process was described by Curzon *et al.* (1975) in his famous article, in which the engine called the “Curzon and Alhborn engine”, made a transformation of heat into work. For this engine all reversibilities are restricted to the heat exchange between the engine and the external world, moreover, it has a particular efficiency limited by the rate at which the heat is being exchanged between the working fluid and the heat reservoirs. So, one interesting conclusion obtained from these studies is that in the endoreversible process all the irreversibilities are located in the heat transport.

With the former in mind and the works of Ondrechen *et al.* (1980) emerged the first endoreversible models for chemical reactors in which there were not only heat flows but also a particle transport. Afterwards, in 1990 De Vos, De Vos (1991), generalized the concept by locating the irreversibilities in the transport of heat and /or matter from the heat and/or sources to the engine and from the engine to the heat and/or matter sinks.

To illustrate the ideas of De Vos (1991), the following reproduced its main results:

2.1 The Curzon and Alhborn engine

As previously mentioned, the Curzon and Alhborn model was the first which described an endoreversible process, and since then the model is termed as the Curzon and Alhborn engine, Curzon (1975) *et al.* Figure 1 shows the basic features of this engine. The model consists of two heat reservoirs, one at high temperature T_1 and the other at low temperature T_2 . There are two thermal resistors that restrict the heat flows Q_H and Q_L . The net result is a drop in temperature from T_1 to an intermediate temperature T_3 and from T_4 to T_2 . It should be noticed that between the intermediate temperatures T_3 and T_4 there exists a reversible Carnot engine. To determine the thermodynamical properties of the CA engine only the implementation of two important principles are needed, De Vos (1991):

a) Conservation of energy:

$$\sum Q + \sum W = 0, \quad (1)$$

where Q is the heat flow, W the work flow, and \sum denotes a summation over an arbitrary closed surface.

b) Conservation of entropy

$$\sum \frac{Q}{T} = 0 \quad (2)$$

where \sum represents the summation over the surface enclosing only the Carnot reversible part. From equations (1) and (2) and from the definition of thermodynamic efficiency ($W = \eta Q_1$), obtains,

$$\eta = 1 - \frac{T_4}{T_3} \quad (3)$$

Suppose that the heat transport follows a Fourier law, this gives,

$$\begin{aligned} Q_1 &= g_1(T_1 - T_3) \\ Q_2 &= g_2(T_4 - T_2) \end{aligned} \quad (4)$$

Equations (3) and (4) yield,

$$Q_1 = g \frac{T_1 - T_2 - T_1 \eta}{1 - \eta} \quad (5)$$

where $g = g_1 g_2 / (g_1 + g_2)$, and W could be obtained as,

$$W = g \eta \frac{T_1 - T_2 - T_1 \eta}{1 - \eta} \quad (6)$$

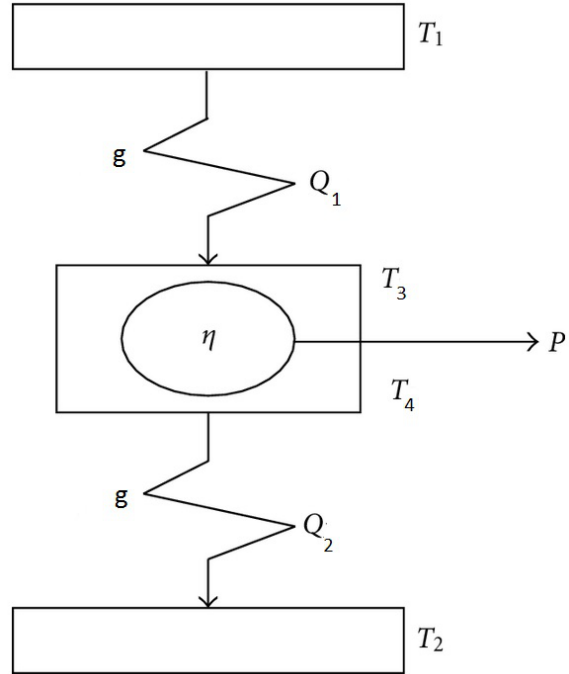


Figure 1. Basic features of the Curzon and Alhborn engine (CA-engine).

Finally, the entropy creation rate follows the expression, De Vos (1991):

$$\dot{S} = g \frac{(T_1 - T_2 - T_1 \eta)^2}{T_1 T_2 (1 - \eta)} \quad (7)$$

It is worthwhile to mention that the produced work displays a maximum value when:

$$\eta = 1 - \left(\frac{T_2}{T_1}\right)^{1/2} \quad (8)$$

this last expression is termed the well known Curzon and Alhborn Formula.

2.2 The endoreversible chemical reactor

The first model described an endoreversible process related to chemical reactions which was proposed by Ondrechen *et al.* (1980). Later De Vos (1991) demonstrated how the endoreversible process allows modeling a class of chemical reactors in the same fashion as was made for endoreversible heat engines (in particular for the Curzon and Alhborn engine).

Following these ideas let us consider a very simple chemical reactor. Figure 2 shows the basic features of the aforementioned Endoreversible Chemical Reactor (ECR). The model consists of two particle reservoirs, one at high chemical potential μ_1 and the other at

low chemical potential μ_2 (the terms “high” and “low” refer to the situation in which $\mu_1 > \mu_2$). There are two particle resistors that restrict the particle flows N_1 and N_2 . The net result is a drop in the particle flow from N_1 to an intermediate particle reservoir N_3 and from N_4 to N_2 . It should be noticed that between the intermediate particle reservoirs N_3 and N_4 there is a kind of reversible Carnot-like engine.

Figure 2 deliberately substitutes the chemical potentials μ for the more common concentrations n . Both variables can be interchanged because of a relationship known as the Nerst equation, De Vos (1991):

$$\mu = \mu_0 + kT \log(n/n_0) \quad (9)$$

where μ_0 denotes the chemical potential at normalized concentration n_0 .

Let us consider the following simple chemical reaction:



where, A_1 denotes the reactant in the reaction and A_2 is the product of the reaction (the calculation can be adapted for an arbitrary reaction with arbitrary stoichiometric coefficients, De Vos (1991)).

As in the CA engine in order to determine the properties of the ECR, only the implementation of two important principles is needed, De Vos (1991):

a) Conservation of matter:

$$\sum N = 0 \quad (11)$$

where N is the matter flow, and \sum denotes a summation over an arbitrary closed surface.

b) Conservation of energy

$$\sum \mu N + \sum W = 0 \quad (12)$$

where W is the work flow; for both principles the whole reactor has the same temperature T .

Then, taking into account the Fick’s particle diffusion law ($N = -d\nabla n$), the transport equations of matter could be written as,

$$\begin{aligned} N &= h_1(n_1 - n_x) \\ N &= h_2(n_y - n_2) \end{aligned} \quad (13)$$

From equations (11) and (12) and from the definition of conversion efficiency ($W = \zeta N$), obtains,

$$\zeta = \zeta_0 + kt \log(n_x/n_y) \quad (14)$$

Proceeding in the same fashion as in the endoreversible thermal engines described in many

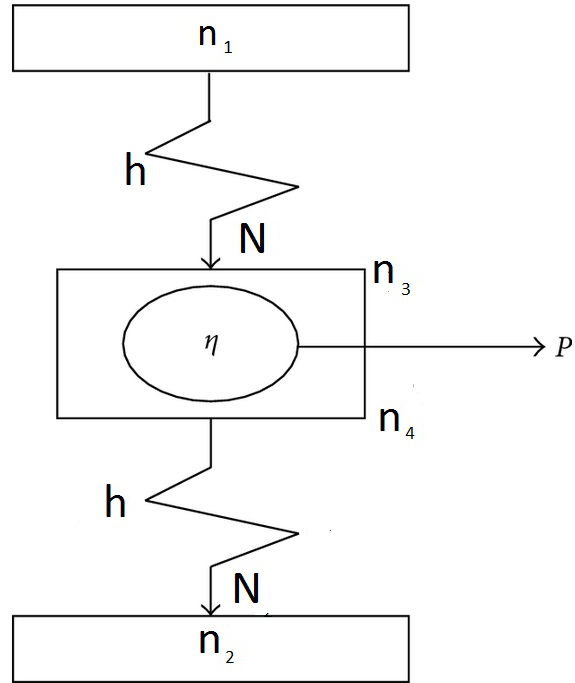


Figure 2. Basic features of the endoreversible chemical reactor (ECR).

papers, Santillan *et al.* (2001), Guzman-Vargas *et al.* (2005), Chimal-Eguia *et al.* (2006), Páez-Hernández *et al.* (2006), Huang *et al.* (2007), Chimal-Eguia *et al.* (2007) Nie *et al.* (2008), Huang *et al.* (2008), Huang *et al.* (2009), Páez-Hernández (2008), Santillán *et al.* (2008), from equations (13) and (14) it is possible to obtain the unknown concentrations n_x and n_y as,

$$n_x = \frac{h_1 n_1 + h_2 n_2}{h_1 + h_2 e^{-(\zeta - \zeta_0)/kT}} \quad (15)$$

and

$$n_y = \frac{h_1 n_1 + h_2 n_2}{h_1 e^{(\zeta - \zeta_0)/kT} + h_2} \quad (16)$$

Consider $h_1 = h_2 = h$, $x = \zeta/kT$ and $b = e^{-\zeta_0/kT}$ equations (15) and (16) can be re-written as;

$$n_x = \frac{n_1 + n_2}{1 + be^{-x}} \quad (17)$$

and

$$n_y = \frac{n_1 + n_2}{1 + be^x} \quad (18)$$

Now, taking the definition of conversion efficiency ($W = \zeta N$), it is possible to obtain a relation for W as follows,

$$W = \frac{h\zeta n_x (1 + be^{-x})(1 - Abe^x)}{(1 + A)(1 + be^x)} \quad (19)$$

where x and b are defined as before, ζ is defined by equation (14) and $A = e^{(\zeta_0 - \zeta_C)/kT}$ with $\zeta_C = \zeta_0 - kT \log(n_2/n_1)$ is obtained from the diffusion-limited reaction rate which is well-known in chemistry and electrochemistry, for example see Bejan (1988).

3 Dynamics for the endoreversible chemical reactor

The previous sections have considered that the chemical reactor works between two particle reservoirs μ_1 and μ_2 and there are particle flows from μ_1 to μ_x and from μ_y to μ_2 through particle resistances governed by a linear particle transfer law. It is important to notice that the particle reservoirs n_x and n_y (see equation (9)) are concentrations in steady state. Now let us consider that these concentrations are not in steady state, and they can change over time as follows,

$$\frac{dn_x}{dt} = C_1[h_1(n_1 - n_x) - N_1] \quad (20)$$

and

$$\frac{dn_y}{dt} = C_1[N_2 - h_2(n_y - n_2)] \quad (21)$$

Both of these derivatives must cancel when n_x , n_y , N_1 and N_2 take their steady-state values. Taking into account that $N_1 = N_2 = N$ and from the conservation of energy (equation 12), obtains,

$$n_y N + W - n_x N = 0 \quad (22)$$

which implies that,

$$N = \frac{W}{n_x - n_y} \quad (23)$$

Substituting equation (22) into equation (20) yields,

$$\frac{dn_x}{dt} = C_1 \left[h_1(n_1 - n_x) - \frac{W}{n_x - n_y} \right] \quad (24)$$

and

$$\frac{dn_y}{dt} = C_1 \left[\frac{W}{n_x - n_y} - h_2(n_y - n_2) \right] \quad (25)$$

then substituting equation (19) into equations (24) and (25) leads to the following set of differential equations for the concentrations n_x and n_y of the endoreversible chemical reactor,

$$\frac{dn_x}{dt} = C_1 \left[h_1(n_1 - n_x) - \frac{h\zeta n_x(1+be^{-x})(1-Abe^x)}{(1+A)(1+be^x)} \right]$$

and

$$\frac{dn_y}{dt} = C_1 \left[\frac{h\zeta n_x(1+be^{-x})(1-Abe^x)}{(1+A)(1+be^x)} - h_2(n_y - n_2) \right]$$

Let $f(x,y)$ and $g(x,y)$ be defined as,

$$\frac{dn_x}{dt} = f(n_x, n_y) \quad (26)$$

$$\frac{dn_y}{dt} = g(n_x, n_y) \quad (27)$$

with,

$$f(n_x, n_y) = C_1 \left[h_1(n_1 - n_x) - \frac{h\zeta n_x(1+be^{-x})(1-Abe^x)}{(1+A)(1+be^x)} \right] \quad (28)$$

$$g(n_x, n_y) = C_1 \left[\frac{h\zeta n_x(1+be^{-x})(1-Abe^x)}{(1+A)(1+be^x)} - h_2(n_y - n_2) \right] \quad (29)$$

The steady-state solutions \bar{n}_x and \bar{n}_y are couples (n_x, n_y) that simultaneously satisfy $f(n_x, n_y) = 0$ and $g(n_x, n_y) = 0$. Following the ideas of Strogatz (1994) the local stability of the steady state can be determined by the Jacobian matrix:

$$J = \begin{pmatrix} f_x & f_y \\ g_x & g_y \end{pmatrix} \quad (30)$$

where, f_x , f_y , g_x , g_y are the partial derivatives with respect to concentrations n_x and n_y for the functions f and g , respectively. Let λ_1 and λ_2 be the respective eigenvalues of the Jacobian matrix, and \vec{v}_1 and \vec{v}_2 be their respective eigenvectors. The temporal evolution of some small perturbations from the steady state, δ_x and δ_y is given by,

$$\begin{pmatrix} \delta_x \\ \delta_y \end{pmatrix} = c_1 \vec{v}_1 \exp(\lambda_1 t) + c_2 \vec{v}_2 \exp(\lambda_2 t) \quad (31)$$

where c_1 and c_2 are constants to be determined from initial conditions. Then, it follows from equation (31) that the steady state is stable if the eigenvalues have negative real parts and the functions δ_x and δ_y converge to zero monotonically. Let ψ and ϕ denote the trace and the determinant of the Jacobian matrix respectively, i. e.,

$$\psi = f_x + g_y$$

$$\phi = f_x g_y + f_y g_x$$

Therefore, the eigenvalues then can be calculated as,

$$\lambda_1 = \frac{1}{2}(\psi + \sqrt{\psi^2 - 4\phi}) \quad (32)$$

$$\lambda_2 = \frac{1}{2}(\psi - \sqrt{\psi^2 - 4\phi}) \quad (33)$$

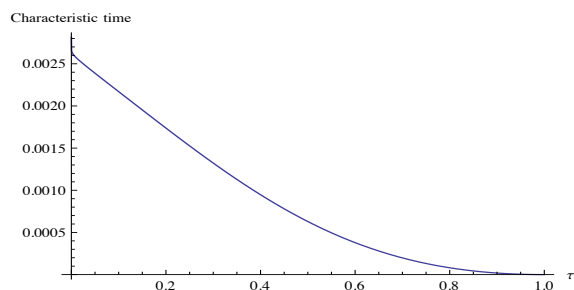


Figure 3. Characteristic time t_1 vs the parameter $\tau = n_x/n_y$.

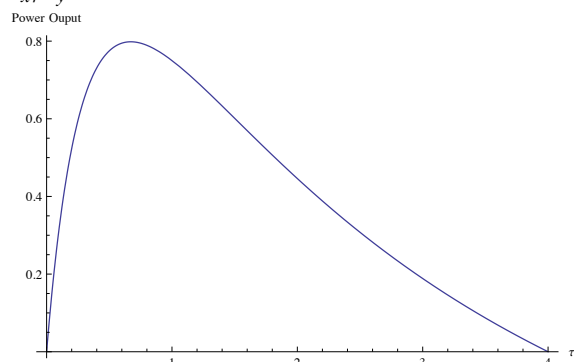


Figure 4. Power Output vs the parameter $\tau = n_x/n_y$.

4 Numerical results for the endoreversible chemical reactor

By taking into account (32) and (33), it is possible to find the eigenvalues λ_1 and λ_2 of the characteristic equation. For this purpose, the computation using the Mathematica software code is implemented showing that one of the eigenvalues λ_1 is real and negative, and the second eigenvalue has no physical sense since it is not real.

It is possible to determine the time required for the concentrations n_x and n_y to vary significantly in the neighborhood of \bar{n}_x and \bar{n}_y respectively. They are called characteristic time scales and are given as:

$$t_{1,2} = \frac{1}{|\lambda_{1,2}|} \quad (34)$$

In fact, using the characteristic time t_1 given by equation (34), verifies that the smaller λ_1 is the faster the system returns to equilibrium after a small perturbation. Figure 3 depicts the time characteristic t_1 against the parameter $\tau = n_x/n_y$, where the behavior described above is observed.

It can be noted that both characteristic times are proportional to C_1 and $h_1 = h_2 = h$ (taking the latter for simplicity), this means, improving the system stability

may decrease or increase C_1 or h . Clearly, the smaller t_1 or t_2 are the faster the system returns to its steady state after a small perturbation. Therefore, Figure 3 shows that the characteristic time t_1 depends on τ . In this case, the system has a better stability as τ increases to one. The power output in the steady state and the efficiency also depend on τ . Figure 4 shows this dependency for the case of power output.

Concluding Remarks

The analysis of local stability of an endoreversible chemical reactor (ECR) has been presented. Considering the ECR as an endoreversible engine and taking into account that it works at a maximum power regime, transport equations at steady state can be obtained. Moreover, it was found that the steady state efficiency and the engine power output can be expressed in terms of $\tau = n_x/n_y$ and h .

To gain more information about the system stability, its behavior under small perturbations was analyzed. It was found that the system declines exponentially to the steady state with either of two characteristic times for any value of $\tau = n_x/n_y$, C_1 and h . Therefore, considering an increase of C_1 or h , the system stability can be improved.

The time scales show that both characteristic times depend on $\tau = n_x/n_y$. The characteristic system time scale decreases the stability depending on whether increases or decreases its value.

Finally, it is important to note that both the power output and efficiency depend on τ , and both are functions related to this parameter. Comparing the energetic properties (the output power or the efficiency) with system stability, it was observed that both depend on τ ; this could suggest that there is a trade-off between energetic properties, and the system stability operated by τ . Both system stability and power output by increasing h can be improved. It should be stressed that the stability properties, together with the energetic characteristics, enhance the dynamic robustness of the chemical engines, in such a way, that both are important for the design of such engines.

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