CO$_2$ METHANATION: OPTIMAL DYNAMIC OPERATION OF A CATALYTIC WALL REACTOR FOR POWER-TO-GAS APPLICATIONS

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Abstract

In contrast to the traditional steady state operation for chemical production systems, we consider dynamic operations for the purpose of utilizing volatile renewable energy sources (solar, wind). Taking the example of a methanation reactor in the context of power-to-gas applications, a dynamic optimization approach is used to identify optimal control trajectories when the hydrogen inlet stream of the reactor is changing over a reasonable time horizon (up to days). For the optimization, we develop a dynamic, two-dimensional model of a catalytic wall tube reactor for CO$_2$ methanation which is based on the reaction network of the underlying Sabatier reaction mechanism. With respect to past production scenarios of wind power, we prove the feasibility of our methodology and investigate the feasibility of dynamic CO$_2$ methanation.

Keywords

Methanation, Optimal Control, Renewable Energy, CO$_2$

Introduction

An example of current interest for dynamic reactor operation is the utilization of renewable resources for chemicals production. The challenge in dealing with these resources lies in their volatile nature (Güttel, 2013), such that a deeper understanding of the dynamic reactor behavior becomes crucial. In this context, the production of appropriate synthetic molecules, such as SNG [1], needs to be reconsidered for optimal process design and optimal operation.

In this contribution, we consider a typical process route from renewable energy through to methane, as shown in Fig. 1. Within this process unsteady operations appear due to the volatile input of renewable electricity (e.g. coming from wind energy). Especially the methanation unit, a catalytic tubular reactor, is very sensitive against volatile inputs, such that the produced methane may not fulfill the product requirements. For this reason, this type of reactor is mostly analyzed under steady state operation conditions (Parlikkad et al., 2013). Recently, also the dynamics around a steady state operation have been investigated in terms of step responses for control and system analysis (Li et al., 2015). More relevant within the context of renewables, Güttel (2013) investigated the system dynamics with respect to cycling inputs in time

ranges of seconds. Nevertheless, for real applications the consideration of a longer time horizon (up to days) is more reasonable, but this is still not available.

In the present work we investigate the optimal operation of a CO$_2$ methanation reactor under fluctuating hydrogen feed, based on typical, renewable production scenarios. To achieve better controllability of the reaction, we use a catalytic wall tubular reactor, which is cooled at the outer wall. More details on the model are given in the next section.
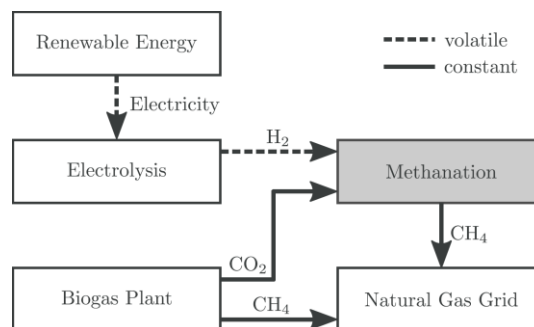

Figure 1. Power-to-gas process route for the conversion of renewable, electrical energy.

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Reactor Model

The model for the methanation reactor is derived by mass and energy balances over an arbitrary control volume. The flow field in the reactor was assumed to be known, either laminar or turbulent. Thus, the governing PDE system is as follows:

$$\frac{\partial \rho_\alpha}{\partial t} = -v_z \frac{\partial \rho_\alpha}{\partial z} + D_{r,\alpha}\left[\frac{\partial^2 \rho_\alpha}{\partial r^2} + \frac{1}{r}\frac{\partial \rho_\alpha}{\partial r}\right], \tag{1}$$

$$\frac{\partial T}{\partial t} = -v_z \frac{\partial T}{\partial z} + \frac{\lambda_r}{\rho c_p}\left[\frac{\partial^2 T}{\partial r^2} + \frac{1}{r}\frac{\partial T}{\partial r}\right], \tag{2}$$

where ρ_α relates to the mass concentration of each component α, $D_{r,\alpha}$ is the radial gas diffusion coefficient of each component, T is the gas temperature, λ_r is the radial thermal gas conductivity, ρ is the mass density of the gas mixture, c_p stands for the heat capacity of the gas mixture, and v_z relates to a given laminar velocity profile. Due to symmetry at the central axis and the catalytic reaction at the inner wall, Neumann boundary conditions (B.C.) are used, in which we implemented the reaction mechanism and the rate expressions proposed by Xu and Froment (1989). The PDE system is discretized using the finite volume method (FVM), such that we obtain an ODE system of the following form:

$$\dot{x}(t) = f(x(t), u(t)),$$
$$x(t_0) = x_0, \quad u(t) \in \Omega, \tag{3}$$

where $x(t)$ represents the entire mass concentration and temperature field of the reactor interior. The control $u(t)$ is related to the cooling temperature at the outer reactor wall and to the inlet flows of all components, except hydrogen. The initial conditions $x(t_0)$ relate to assumed steady state operation established prior to reactor perturbation.

Dynamic Operation

As mentioned before, we consider typical harvesting scenarios for volatile renewable energy. Therefore, daily wind power generation profiles, as shown in Figure 2, are used. Assuming water electrolysis for hydrogen production, where the hydrogen output quickly follows the electricity input, the hydrogen feed flow rate profile is obtained and used as dynamic input for the methanation reactor. The absolute values of the hydrogen input profile are limited with respect to the capacity of the methanation unit, according to the reactor model. With this time-dependent system input, a reactor simulation of an entire production day can be simulated.

Dynamic Optimization

To solve the main objective of an optimal operation for given dynamic hydrogen inputs, we formulate the following optimal control problem:

$$\min_{u(t)} \quad -\int_{t_0}^{t_f} \dot{m}_{CH_4}(x(t), u(t))\, dt, \tag{4}$$

$$s.t. \quad \dot{x}(t) = f(x(t), u(t)), \quad \forall t \in [t_0, t_f]$$
$$g(x(t)) \leq 0,$$
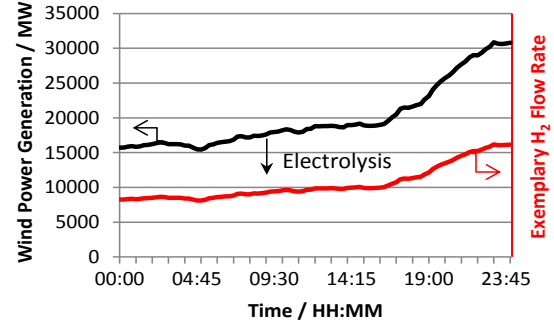$$x(t_0) = x_0, \quad u(t) \in \Omega,$$

Figure 2.   Wind power generation in Germany (date: 2015/11/17, source: eex-transparency.com) and derived hydrogen flow rate profile.

which is seeking for the optimal control trajectories (e.g. for the cooling temperature) to maximize the methane production over the given time horizon. Therefore, we consider boundaries for temperatures to ensure process safety and material demands, as well as product purity to meet the distribution requirements (e.g. within gas grids).

Conclusions

We will show in a comprehensive way that the unsteady state operation of a methanation reactor is possible for various scenarios of renewable energy production. Moreover, we demonstrate that our approach allows a more flexible usage of renewable electricity for chemical energy production systems.

Acknowledgments

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