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Dynamic optimization of fixed bed chemical-looping combustion systems integrated in thermal power plants

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Abstract: Optimization methods can be used to improve the design and operation of batch processes. In this work, we illustrate the application of dynamic simulation and optimization for power generation in chemical-looping combustion systems, carried out in batch reactors. We present a framework for the optimization strategy, in which the objective is to maximize a measure of the energy efficiency, bounded by inequality constraints reflecting the industry standards. Several case studies are used to illustrate the applicability of the present problem formulation to enhance the process performance.

Keywords: dynamic modeling, batch models, optimization problems, power generation

1. INTRODUCTION

Batch processes are widely used to manufacture specialty chemicals, pharmaceutical products and polymers. Typically, batch processes are used when production volumes are low, isolation is required for sterility or safety, and the raw materials involved are difficult to handle. Scheduling of batch operations can be optimized to improve performance and quality. These problems can be computationally expensive due to the complexity of the production sequence (Méndez et al., 2006). The problem should take into account the sequence of products, equipment assignment and connectivity, variable batch sizes, and mixed intermediate storage policies.

This paper presents an application of dynamic optimization batch operations for power generation. We explore high-pressure fixed bed reactors for Chemical-Looping Combustion (CLC) as the application of the presented framework. CLC is a novel technology that combines power production with CO_2 capture. In CLC, an oxygen carrier is used to transfer the oxygen from the air to oxidize a hydrocarbon fuel. While most of the CLC studies focus on continuous processes using interconnected fluidized bed reactors, the use of fixed bed reactors enables the process to operate at high pressures. High-pressure operation is necessary for the seamless integration of CLC with a highly-efficient combined cycle power plant. Other advantages associated with the fixed bed reactor are better bed utilization, elimination of particle attrition, and simplicity in gas/solid separation (Hamers et al., 2013).

The operation of a fixed bed CLC reactor is an inherent batch process. The oxygen carrier is static while the gaseous streams alternate between reduction and oxidation stages. During the reduction step, a gaseous hydrocarbon fuel is oxidized to produce CO_2 and steam. The bed is regenerated in the subsequent oxidation step with air. This step is exothermic and produces a high-temperature, high-pressure gas that can be converted into electricity in a downstream gas turbine of a combined cycle power plant. A short purging step is required between the oxidation and reduction cycles to avoid mixing of the combustible gases. The performance of the CLC reactor inside the power plant is highly constrained, because it needs to adhere to acceptable levels of CO_2 capture efficiency, while producing a suitable gas for the gas turbine, which is usually operated at steady-state conditions. Thus, an adequate cycle strategy is necessary for the process to achieve optimal operating conditions for the combined cycle power plant.

Simulation of power plants with CLC is complex due to the large number of units involved, interaction between CLC and power plant components, and presence of streams of diverse compositions and properties. Thus, investigators typically rely on thermodynamic models for calculating the mass and energy balances throughout the system (Iloeje et al., 2015). Kinetics is often ignored, in favor of assuming the conversion of the fuel and solid, on the basis of experimental measurements from bench-scale units (Erlach et al., 2011). Kolbitsch et al. (2009) showed that incorporation of the relevant kinetics and hydrodynamics is essential to accurately predict CLC performance. In their calculations they found incomplete conversion of solids in the air reactor, which was contrary to previous assumptions by Lyngfelt et al. (2001). Also, ideal assumptions of the mixing patterns inside the reactors can overpredict the conversion inside the fuel reactor, leading to an under-prediction of the solid inventory (Porrazzo et al., 2014). The accuracy of system models can be improved by including the process kinetics and hydrodynamics. Therefore, dynamic optimization should guide reactor design and operation of the CLC system and explore the theoretical limits of the efficiency of a combined cycle power plant with CLC.

The objective of this work is to present a mathematical formulation to optimally design and tune the cycle strategy of batch fixed bed reactors for power generation with in-situ CO_2 capture. We explore the optimality of batch CLC systems and investigate their batch operation with different

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oxygen carriers and fuel types, to showcase the applicability of this generic approach. A reverse-flow reactor is proposed to increase the efficiency of batch CLC reactor systems. A model-based approach is used to compare the novel reverse-flow reactor with the conventional fixed bed and a fluidized bed processes. The novelty of this work is the formulation of dynamic optimization problems to enable a comparison between batch and continuous systems proposed for CLC. The most efficient process should be eventually nominated for scale-up.

2. OPTIMIZATION STRATEGY

2.1 Process Options

In this work, we explore three CLC reactor configurations: (1) fixed bed; (2) reverse-flow fixed bed; and (3) fluidized bed reactors. Due to the different modes of operation, the utilization of the energy produced from the combustion process is different. In the fixed bed processes, the oxygen carrier material is static and the inlet gas varies between fuel, air, and inert. The direction of the gas is switched intermittently during the reduction and/or oxidation cycles in the reverse-flow fixed bed reactor. A high-temperature gas stream is produced after the oxidation step, which can be used for power generation in a downstream gas turbine of the combined cycle. The gas stream produced from the reduction cycle leaves the reactor at lower temperatures and can be used for heat recovery within the power plant. In contrast, the fluidized bed process operates in continuous mode, by transferring the oxygen carrier material between the so-called fuel and air reactors. The depleted air stream downstream the air reactor can be sent to the combined cycle to generate power.

To operate at high efficiencies, the inlet to the gas turbine should be at the maximum allowable temperature and pressure permitted by the CLC reactor. The energy recovery components downstream the gas turbine should also be optimized, to balance the efficiency of the steam turbine with the preheating demands of the CLC reactor. The performance of the CLC reactor is also heavily constrained by the limits of CO_2 capture, melting points of the materials, and pressure drops. All of these considerations must be taken into account in the process design phase of an integrated power plant with CLC. Thus, there is potential to improve the efficiency of these systems and to explore the envelope of performance through process optimization.

2.2 Problem Formulation

A generic optimization problem for the CLC reactor system can be formulated as follows:

$$\begin{aligned} \max_{\boldsymbol{\varphi}\in\boldsymbol{\Phi}} \quad & J \\ s.t. \quad & \mathbf{f}(\dot{\mathbf{x}}(t),\mathbf{x}(t),\mathbf{u}(t),\boldsymbol{\theta})=0, \\ & \mathbf{g}(\dot{\mathbf{x}}(t),\mathbf{x}(t),\mathbf{u}(t),\boldsymbol{\theta})\le 0, \end{aligned} \tag{1}$$

where J is the scalar performance index to be maximized, $\mathbf{x}$ is the vector of state variables, $\mathbf{u}(t)$ is the vector of control variables, $\mathbf{f}$ is the set of differential equations describing the dynamics of the system, $\mathbf{g}$ is the vector of path constraints for the state variables, and $\boldsymbol{\varphi}$ is the vector of design variables to be optimized bounded by the design space $\boldsymbol{\Phi}$.

2.3 Batch Process

The input to a batch CLC reactor is the feed gas, which dynamically alternates between fuel, air, and inert. This time-varying control vector is modeled with piecewise functions, $\mathbf{u}(t)=\mathbf{u}_i$, over the duration of the CLC step τ_i, *i.e.*, reduction, oxidation, heat removal, and purge. For a given reactor design and fuel flow, the optimizer manipulates the air flow rate and inlet temperature, time intervals for the CLC steps, and active metal content in the oxygen carrier, ω. These variables are contained within the design vector, $\boldsymbol{\varphi}'$:

$$\boldsymbol{\varphi}'=[\mathbf{u}_i,\tau_i,\omega] \ . \tag{2}$$

In the reverse-flow reactor, the direction of the gas flow is periodically reversed during the reduction and/or oxidation steps. The number of flow switches n_{sw} and the time period for each flow switch $\tau_{n_{sw}}$ can be optimized. The design vector for the reverse-flow reactor is thus:

$$\boldsymbol{\varphi}''=[\mathbf{u}_i,\tau_i,\tau_{n_{sw}},n_{sw},\omega] \ . \tag{3}$$

The power generation efficiency of the batch CLC process can be quantified in terms of the duration of the heat removal phase (J') and the fraction of heat leaving the reactor during heat removal (J''):

$$J'=\int_{t_0}^{\tau_{HR}} \alpha T_{out}(t)dt \ , \tag{4}$$

$$J''=\frac{\int_{t_0}^{\tau_{HR}}\left(\dot{m}_{out}(t)h_{out}(t)\right)dt}{\int_{t_0}^{\tau_{cycle}}\left(\dot{m}_{out}(t)h_{out}(t)\right)dt} \ , \tag{5}$$

where $T_{out}(t)$ is the temperature of the exhaust gas, τ_{HR} is the time interval of the heat removal step, τ_{cycle} is the time interval for one complete redox cycle, $\dot{m}_{out}$ is the exit mass flow rate, and h_{out} is the enthalpy of the exhaust gas. In (4), the process efficiency is measured by the time interval of the heat removal step to produce electricity in the gas turbine. The process efficiency of (5) is a function of the heat leaving the reactor during heat removal relative to the total heat produced during one complete redox cycle.

We incorporate several path constraints, $\mathbf{g}$, in the dynamic problem of (1) to ensure that the CLC process behaves within performance specifications. The exhaust gas temperature during heat removal should be within 50°C of the set-point value of the turbine inlet temperature, *TIT*. The CLC reactor during the entirety of the reduction step needs to achieve a high fuel conversion ($X_{fuel}(t)\geq 98\%$) and high CO_2 capture efficiency ($S_{CO2}(t)\geq 90\%$), calculated from the cycle-averaged gas flow rates. To prevent sintering of the oxygen carrier, the temperature at any time and point inside the reactor needs to be less than 50°C below the melting point of the material, $T(t,z)\leq T^{max}$-50°C. The pressure drop across the reactor needs to be below 8% of the inlet pressure ($\Delta P/P(t,z=0)\leq 8\%$), to