Control of regenerative catalytic oxidizers used in coal mine ventilation air methane exploitation

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Abstract

Ventilation air methane in coal mining has an important environmental impact, since methane is a strong greenhouse gas (1 kg of methane is equivalent to 28 kg of carbon dioxide). The oxidation of methane in regenerative oxidizers can be an attractive technique to exploit this resource. Thus, part of the heat released by the reaction can potentially be recovered, in addition to decreasing methane environmental impact. However, the concentration of methane in the mine ventilation air may change considerably with respect to the oxidizer design value, which have negative consequences. An increase in concentration can produce overheating (with possible damage to the unit), while a decrease in concentration may cause the extinction of the reaction. In this work, three control systems have been considered in order to deal with these issues: proportional-integral-derivative (PID) and proportional-integral (PI) feedback controllers, and model predictive controller (MPC).

The control action is based on regulating the heat extracted from the oxidizer by adjusting a hot gas purge from the centre of the reactor. First, the control systems have been designed (i.e. the tuning parameters of the controller have been calculated). To carry out the design of the controllers, a simplified dynamic model was obtained from a complex model of the oxidizer. Then, the performance of the controlled oxidizer has been simulated for different types of disturbances. In these simulations, the simple PID controller performed well, and the MPC exhibited the fastest response.

Keywords: dynamic reactor; periodic operation; reverse flow reactor; methane emissions; reactor modelling.

1 Introduction

In the last years, the environmental impact caused by coal mining and utilization has raised a great concern. During mining operations, methane gas from the coal bed is released to the atmosphere in great amount (Díaz et al., 2012; Zheng et al., 2019). Methane is a powerful greenhouse gas with a global warming potential (GWP) of 28 (i.e. 1 kg of methane produces a radiative forcing over a period of 100 years equivalent to 28 kg of carbon dioxide) (Myhre et al., 2013). The present work is focused on the reduction of methane gas emissions in underground coal mining.

To reduce the risk of explosion, methane released in coal mining is diluted and vented to the outside of the mine using a shaft (Wang et al., 2019). Therefore, the diffusive methane emissions of the inside of the mine are collected together and typically vented to the outside through a single point. This is a great advantage for the implementation of a treatment technique. Thus, different techniques have been proposed and those based on combustion being the most promising (Warmuzinski, 2008). Given their difference in GWP, the oxidation of methane to carbon dioxide leads to a strong reduction of the net radiative forcing caused by these emissions. However, typical methane concentration in these emissions is very low, so the use of efficient combustors, such as, regenerative oxidizers is necessary.

Regenerative oxidizers (also called reverse flow reactors) are a type of reactors that work under forced unsteady state conditions. In short, they consist of a fixed bed in which the feed flow direction is periodically reversed. The fixed bed can be formed of inert material, catalyst or combinations of both. For exothermic reactions, reversing the flow direction allows the storage of a great amount of heat inside the reactor in consecutive cycles. This makes it possible the auto-thermal operation, even for slightly exothermic reactions (e.g., combustion of organic compounds in air at very low concentration). When the concentration is higher and more heat is released, part of this heat can be recovered, e.g. for generation of steam. There are two main types of regenerative oxidizers: regenerative catalytic oxidizers (RCO) and regenerative thermal oxidizers (RTO). The first group uses a catalyst to decrease the temperature of the combustion reaction, while the second depends only on the high temperature thermal combustion (Matros and Bunimovich, 1996; Fissore et al., 2005; Zagoruiko, 2012).

In regenerative oxidizers, the system is started by pre-heating the feed or the bed, so that the feed reaches the ignition point in the catalytic bed. Once the feed flow reversing is started, the pre-heating is switched off. When the reactor design and operating switching time are adequate, a pseudo-steady state is reached after some cycles. This state is characterized by the repetition of the evolution of temperature and concentration profiles between cycles, resulting in the autothermal combustion of the feed and very high reactant conversion. For constant inlet conditions, and assuming no change in the catalyst activity, this pseudo-steady state can be maintained indefinitely (Barresi et al., 2007; Marín et al., 2019).

In practice, feed conditions, most commonly volumetric flow rate and concentration of methane, can change with time. This can produce changes in the reactor temperature profile and reaction conversion, affecting the adequate reactor operation. The two main problems of regenerative oxidizers are reaction extinction (null or very low exit conversion) and reactor overheating.

Extinction occurs when the reactor temperature is too low (this can be produced, for instance, by a decrease in the feed concentration), while overheating (produced, for instance, by an increase in the feed concentration) can lead to catalyst damage, un-desired side reactions, or other operational problems.

In underground coal mining, methane concentration in the ventilation air can change unexpectedly (Fernández et al., 2016). This affects the process safety considerably, leading to equipment overheating or even explosions for very high methane concentrations (the flammability range of methane in air is 5-15% vol.) (Baldissone et al., 2016). In order to avoid this, and allow satisfactory operation (i.e. stable with high conversion and no overheating), a control system is needed. Additional objectives of the control system can be to minimize the consumption of external energy or maximize the energy recovery (when this is feasible). Effective control system design is a difficult task, due to the hybrid continuous-discrete nature of regenerative oxidizers. In addition, the complex transport-reaction phenomena occurring in these devices results in a complex non-linear behaviour. The difficulty of controlling these reactors is considered one of the main barriers for their more widespread industrial use.

There are many published works related to the control of regenerative oxidizers and reactors. Most of them use mathematical simulations for their studies, being the experimental studies scarce. Table 1 summarizes the most important.

Table 1. Summary of the controllers proposed for regenerative oxidizers.

Type of controller	References	ferences Manipulated variables	
Logic controller	(Barresi and Vanni, 2002)	Switching time	
	(Marín et al., 2010)	Switching time	
	(Li et al., 2013a; Li et al.,	Hot gas withdrawal	
	2013b; Li et al., 2017)	Fuel addition	
,		Air dilution	
Fuzzy logic controller	(Li et al., 2014)	Fuel addition	
		Air dilution	
Feedback	(Budman et al., 1996)	Switching time	
		Heat transfer	
	(Hua et al., 1998)	Electrical heating	
	(Mancusi et al., 2007)	Switching time	
	(Fissore and Barresi, 2008)	Electrical heating	
		Air dilution	
Model predictive controller	(Dufour et al., 2003; Dufour	Electrical heating	
	and Touré, 2004)	Air dilution	
	(Edouard et al., 2005a)	Electrical heating	
		Air dilution	
	(Fuxman et al., 2007)	Hot gas withdrawal	
	(Balaji et al., 2007)	Fuel addition	
		Air dilution	
Linear quadratic regulator	(Edouard et al., 2005b)	Electrical heating	
		Air dilution	
	(Fuxman et al., 2008)	Hot gas withdrawal	

The variable most commonly used to detect changes in the reactor performance (i.e. control variable) is temperature. A single temperature measurement, generally situated at the reactor centre has been used by some authors. In other cases, there are two temperature-measuring points, situated one at each reactor end. In order to prevent both extinction and overheating, it has been reported that better results are achieved when measuring temperature at the reactor inlets and centre. Other authors include in their designs more temperature probes along the reactor (Marín et al., 2010).

Feed flow rate can also be measured, as well as, feed composition. The on-line measurement of feed composition can be difficult and expensive. Thus, some authors have developed "observers", based on detailed mathematical models, which allow the inference of feed composition and other characteristics from measurements of temperature along the reactor (Edouard et al., 2004; Fissore et al., 2006; Hua et al., 1998).

One of the possible manipulated variables is switching time (Barresi and Vanni, 2002; Budman et al., 1996; Mancusi et al., 2007; Marín et al., 2010). For given feed conditions (e.g. flow rate and concentration), a reactor unit exhibits stable operation only for a range of switching time values. For too large switching times, the reactor extinguishes, while, for too short ones, the "wash out" phenomenon becomes relevant. This phenomenon consists in the emission of the unreacted feed contained in the reactor and piping (between the switching valves and the reactor), which leaves the reactor every time the flow direction is switched. For too short switching times, the average reaction conversion is considerably reduced due to the "wash out". Considering these constrains, switching time can be used as manipulated variable in the control system, though some authors report poor performance in these systems.

Another alternative (that can be combined or not with the manipulation of the switching time) is the addition of external energy, when the reactor temperature is too low, and the extraction of energy, when the reactor temperature is too high. Energy can be added to the reactor with an internal or external heater, using electricity or hot fluids, or by adding a fuel to the reactor feed (Fissore and Barresi, 2008; Hua et al., 1998). Reactor temperature can be decreased by extracting energy from the reactor with an internal or external refrigeration system, by diluting the feed with air (at the reactor inlet or in the middle point), or by by-passing part of the cold feed to the reactor centre (thus, this fraction of the feed is not pre-heated in the hot first part of the bed) (Balaji et al., 2007; Edouard et al., 2005b). Other possibility consists of withdrawing part of the hot gas, generally in the central part of the reactor (Fuxman et al., 2007; Li et al., 2013a; Li et al., 2013b; Li et al., 2017).

Control algorithms of different complexity have been studied. The simplest controller consists of a logic controller actuating on the switching time. In these systems, the controller only has two states, direct and reverse flow (Barresi and Vanni, 2002; Li et al., 2013a; Li et al., 2013b; Li et al., 2017; Marín et al., 2010). More advanced fuzzy logic controllers of types 1 and 2 have also been used, type-1 controllers being sufficient for getting good results for RCO (Li et al., 2014). Conventional proportional-integral-derivative (PID) feedback controllers have also been proposed (Budman et al., 1996; Hua et al., 1998; Marín et al., 2014).

Several authors use Model Predictive Controllers (MPC); in some cases, in combination with Repetitive Model Predictive Control, which updates the values of the model variables online

(Dufour et al., 2003; Dufour and Touré, 2004; Edouard et al., 2005a; Fuxman et al., 2007) (Balaji et al., 2007). Model Predictive Controllers are based on the use of a process model to calculate the optimum values of the manipulated variables. This type of control is complex and computational intensive. For this reason, the success of these controllers is usually based on the use of simplified models and observers. Linear Quadratic Regulators (LQR) have been also used (Edouard et al., 2005a; Fuxman et al., 2008).

The present work addresses the safe operation of regenerative catalytic oxidizers, used in the treatment of coal mine ventilation air methane. Safety is achieved by the use of controllers aimed at preventing reactor overheating and extinction. The methodology is based on the use of computer simulations based on a complex phenomenological model of the regenerative oxidizer. In the first place, a reactor is designed for a typical methane emission and its operating window in the absence of controller is analysed. Three controllers, PID and PI feedback and model predictive controllers, are proposed and designed. Then, the performance of the controlled regenerative oxidizer when dealing with disturbances in methane feed concentration is analysed.

2 Materials and methods

2.1 Mathematical modelling

The modelling of regenerative oxidizers has been addressed by many authors, as summarized elsewhere (Marín et al., 2019). In the present work, a phenomenological dynamic heterogeneous 1D model has been used. The model is formed by the mass and energy balances applied separately to the gas and solid phases:

$$\begin{split} \epsilon_b \frac{\partial c_{Gi}}{\partial t} &= -\frac{\partial (uc_{Gi})}{\partial z} + \epsilon_b D_{ei} \frac{\partial^2 c_{Gi}}{\partial z^2} - aK_C (c_{Gi} - c_{Si}) \\ -aK_C (c_{Si} - c_{Gi}) + (1 - \epsilon_b) \rho_S r_{cat} \eta &= 0 \\ \epsilon_b \rho_G C_{PG} \frac{\partial T_G}{\partial t} &= -\rho_G C_{PG} u \frac{\partial T_G}{\partial z} + \epsilon_b \kappa_{Ge} \frac{\partial^2 T_G}{\partial z^2} - ah(T_G - T_S) - Q_{lost} \\ (1 - \epsilon_b) \rho_S C_{PS} \frac{\partial T_S}{\partial t} &= (1 - \epsilon_b) k_S \frac{\partial^2 T_S}{\partial z^2} - ah(T_S - T_G) + (1 - \epsilon_b) \rho_S r_{cat} \eta \Delta H_R - Q_{lost} \end{split}$$

Initial conditions:

$$c_{Gi}|_{t=0} = 0$$
 and $T_G|_{t=0} = T_S|_{t=0} = T_{pre-heating}$

Boundary conditions:

Direct flow,
$$u > 0$$

 $c_{Gi}|_{z=0} = c_{Gi,0}$
 $T_{G}|_{z=0} = T_{G0}$
 $T_{G}|_{z=L} = T_{G0}$
 $T_{G}|_{z=L} = T_{G0}$
 $T_{G}|_{z=L} = T_{G0}$
 $\frac{\partial c_{Gi}}{\partial z}|_{z=L} = \frac{\partial T_{G}}{\partial z}|_{z=0} = \frac{\partial T_{S}}{\partial z}|_{z=0} = \frac{\partial T_{S}}{\partial z}|_{z=0} = \frac{\partial T_{S}}{\partial z}|_{z=0} = 0$
 $\frac{\partial c_{Gi}}{\partial z}|_{z=0} = \frac{\partial T_{G}}{\partial z}|_{z=0} = \frac{\partial T_{S}}{\partial z}|_{z=0} = 0$

This type of model is able of predicting the evolution with time (t) of gas (G) and solid (S) concentration (c) and temperature (T) profiles along the reactor length (z) is the axial coordinate): $c_{Gi}(t,z)$, $c_{Si}(t,z)$, $T_G(t,z)$ and $T_S(t,z)$. The switch of the flow direction is simulated by a change in the sign of the gas superficial velocity (u) and by reversing the boundary conditions (as indicated above).

The modelled regenerative catalytic oxidizer is formed by two beds made of honeycomb monolith blocks and an open chamber in the middle. In this work, the properties of the blocks have been selected to match those of typical commercial monoliths used in oxidizers: cell density 100 cpsi, channel width (D_h) 1.8 (mm), bed porosity (ϵ_b) 0.64, surface area (a) 1310 m²/m³, density (ρ_S) 2400 kg/m³, heat capacity (C_{PS}) 865 J/kg K and thermal conductivity (k_S) 1.6 W/m K. The open chamber in the middle can be used to implement systems of heat extraction from the oxidizer, such as, heat exchange or hot gas purge. In the modelling, it is assumed to contain only gas and to have the same cross-section as the beds and a length of 0.40 m.

The monolith blocks can be either inert or catalytic. The catalytic monoliths incorporate a washcoating layer made of a porous material that supports the catalytic active phase. The properties of the catalyst correspond to a commercial Pd-based monolithic catalyst, as published elsewhere (Fernández et al., 2016), and summarized in the following. The reaction kinetics is of first-order with respect to methane: reaction rate $-r_{cat} = \rho_{cat} k_w R T_s C_{CH_4}$, with reaction rate constant $k_w = 1.56 \, e^{-9622/T_S} \, \text{mol/kg}_{cat} \, \text{s} \, \text{Pa}$. The influence of mass transfer inside the washcoating layer (thickness $L_w = 76 \, \mu \text{m}$) is accounted for using the effectiveness factor, $\eta = (\tanh \phi)/\phi$ (Levenspiel, 1999), being the Thiele modulus $\phi = L_w \sqrt{\rho_{cat} k_w R T_s/f_w D_{ep}}$. The Thiele modulus is calculated using experimentally measured textural properties of the catalyst (average pore size 0.12 nm and internal porosity 0.16) and literature correlations for methane diffusion coefficients (Fernández et al., 2016; Levenspiel, 1999). The enthalpy of the reaction is $\Delta H_R = -802.5 \, \text{kJ/mol}$ methane.

The transport properties on the honeycomb monoliths are calculated using correlations from the literature for the gas-to-solid mass and heat transfer (K_C and h, respectively) (Hayes and Kolaczkowski, 1997) and mass and heat axial dispersion (D_{ei} and κ_{Ge} , respectively) (Levenspiel, 1999).

An insulation layer is used to minimize heat losses through the wall of the oxidizer. However, they cannot be completely avoided and, for this reason, the energy lost (Q_{lost}) is also accounted for in the model. These losses are calculated considering heat transfer in the insulation (layer of 0.20 m with thermal conductivity of 0.32 W/m K) and surrounding air (at 5°C with heat transfer coefficient 20 W/m² K).

2.2 Model solving

The proposed model is formed by a set of partial differential equations and algebraic expressions (equations used to calculate the properties and reaction and transfer rates). On solving the model, temperature and concentration profiles upon time are obtained. This can be computationally demanding.

The model has been solved using COMSOL Multiphysics software. This software is based on the Finite Element Method. In particular, this problem was solved using the default PARDISO solver. The switch of the flow direction has been implemented in COMSOL by changing the sign of the velocity vector: positive during direct flow and negative during reverse flow.

3 Results and discussions

3.1 Design case study

The nominal operating conditions considered for the regenerative catalytic oxidizer are: gas flow rate 2.25 Nm³/s and methane feed concentration 0.40% vol. These conditions correspond to coal mines with moderate to rich methane emissions (Díaz et al., 2012). However, the oxidizer has been designed for a minimum methane feed concentration of 0.20% vol., in order to have some room for implementing the actions of the control system. This means that lower concentrations than 0.20% vol. will produce the extinction of the reactor, while higher concentrations may cause overheating. In the latter case, a control system is used to extract the excess heat from the reactor and prevent heat accumulation.

The design has been carried out using the complex reactor model introduced in the Materials and Methods section. For a given set of design specifications, the model is used to simulate the regenerative combustor with different bed sizes. For a too small bed, the combustor extinguishes, while for a very large one, too much heat is stored in the bed and, hence, the combustor suffers from overheating (progressive increase in bed temperature with time). The target bed size is the one providing a balance between these two extreme behaviours, with bed temperature profiles being reproducible upon time (pseudo steady-state).

The regenerative catalytic oxidizer is formed by two beds and an open chamber in-between, where in order to control the temperature of the reactor, the hot gas can be cooled or extracted. Typical design specifications consist of nominal superficial velocity of 1 m/s and switching time of 90 s. For these conditions, each of the beds (with square section) require a cross-sectional area of $1.5 \times 1.5 \, \text{m}$ and a length of $0.60 \, \text{m}$. The beds are formed by honeycomb monolithic blocks of $60x60 \, \text{channels}$ (cell density $100 \, \text{cpsi}$) with one catalytic block (length $0.30 \, \text{m}$) stacked on top of another inert block (length $0.30 \, \text{m}$).

Figure 1 shows the evolution of the bed temperature profile at the pseudo-steady state. The evolution with time between two consecutive switches of the flow direction (gas flow from left to right) is illustrated by different lines (each one every 15 s). The conditions of the simulations are: feed concentration 0.20% vol. methane, superficial velocity 1 m/s and switching time 90 s.

The displayed profiles are identical cycle after cycle, unless there is any disturbance (e.g. change in feed concentration or gas flow rate). The two sets of lines correspond to the temperature profile in each of the two regenerative beds. The solid temperature of a given point in the left regenerative bed decreases upon time, because the heat stored is being transferred to the gas entering the reactor. On the contrary, solid temperature increases gradually in the right regenerative bed, as it stores heat from the hot gas produced in the combustion. This is the fundamental of the regenerative principle.

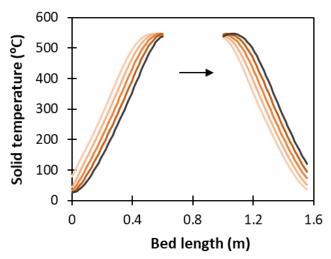


Figure 1. Design base case of the regenerative catalytic oxidizer: solid temperature profiles every 15 s (the arrow indicates the flow direction).

Figure 2 shows the stability map of the designed regenerative catalytic oxidizer. This map consists of a plot of the minimum methane concentration required to achieve stable operation as a function of the gas superficial velocity. The rest of the parameters (e.g. flow rate, switching time or bed size) are maintained the same. This map is very useful to set the operational conditions of an oxidizer. Below the line, the reactor extinguishes, because the reactor is unable of storing enough heat. Above the line, the operation is stable, but heat extraction may be required to prevent overheating. The corresponding maximum bed temperature is also depicted on the right axis. For 1 m/s and 0.20% vol. methane, maximum bed temperature is 535°C, as observed in the profiles in Figure 1.

It should be noted that on increasing the superficial velocity, a higher methane concentration is required to prevent the extinction. In addition, pressure drop and maximum temperature also increase, which are strongly related to the operation cost. For this reason, the superficial velocity is typically maintained at a reasonable low value.

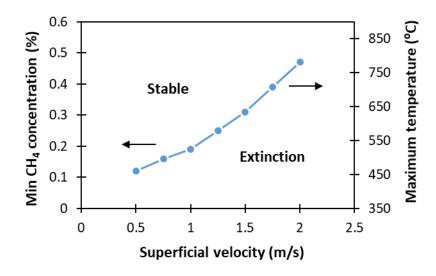


Figure 2. Stability map of the designed regenerative catalytic oxidizer.

3.2 Implementation of the control action

The aim of the reactor control is to maintain the operation of the reactor stable, overcoming disturbances in the feed methane concentration that could lead to reactor overheating or extinction. The first step for the implementation of a reactor control system is the selection of the controlled and manipulated variables.

In this case, the controlled variable is the reactor central temperature (i.e. the temperature of the gas in the open chamber of the centre of the reactor), a good indicator of the state of the reactor, which anticipates reactor overheating (temperature build-up) or extinction (temperature decrease, cycle after cycle).

The manipulated variable is chosen to deal with the excess of heat in the reactor. In the literature, several means of heat extraction from regenerative oxidizers have been considered. One of them consists of the use of a heat exchanger, placed in the open chamber of the reactor centre. This chamber is the part of the reactor at the highest temperature and, hence, where heat extraction is more efficient. This heat extraction produces a drop in the gas temperature, which affects the amount of sensible heat stored in the regenerative bed placed downstream. This sharp drop in the gas temperature has been found to cause important disturbances on the reactor performance (Marín et al., 2009).

Other heat extraction system consists of withdrawing hot gas from the centre of the oxidizer. In the centre of the reactor, temperature is the highest, so the purge of a small fraction of gas can have a marked influence on the amount of heat stored in the beds between cycles. As demonstrated elsewhere (Marín et al., 2014; Marín et al., 2009), this way of heat extraction is easy to implement using a valve, providing a good range of reactor control and causing little disturbance to the operation of the oxidizer. For this reason, the fraction of hot gas purge from the reactor centre is chosen as the manipulated variable.

3.3 Design of the control system

Two types of controllers have been selected for study: feedback and model predictive controllers. The tuning of the control action (e.g. the determination of the best parameters of the control algorithm) requires the use of a simple model describing the reactor dynamics. The following section describes the development of this simple model.

3.3.1 System identification

The system identification consists of the determination of a simple dynamic model of the oxidizer, relating the changes in the manipulated (fraction of hot gas purge) and disturbed (methane feed concentration) variables to the controlled variable (reactor centre temperature). It is usually desired an easy-to-solve model, i.e. a model mathematically simple. The stimulus-response technique is usually helpful in the system identification task. According to this technique, a change (typically a step change) is set in the manipulated and disturbance variables, and the dynamic response of the controlled variable is recorded.

In this case, the mathematical model of the regenerative catalytic oxidizer described in the Materials and Methods section has been used to simulate the dynamic response to the changes imposed using the stimulus-response technique. The procedure is summarized as follows. First, the designed regenerative oxidizer is simulated up to the pseudo-steady state for the nominal operating conditions using the complex model. Note that the nominal methane feed concentration was set to 0.40% vol., while the design one was 0.20% vol. This means that at nominal conditions the fraction of hot gas purge must be set at 0.135 to prevent temperature build-up and maintain a pseudo-steady state regime with a central temperature of 535°C.

Once the pseudo-steady state was reached, a step change in the fraction of the hot gas purge or methane concentration is introduced. As an example, Figure 3 shows the dynamic evolution of the central temperature for a change of -0.04 in the fraction of the hot gas purge (e.g. valve closing from 0.135 to 0.095). As shown, the central temperature oscillates, due to the change in the flow direction. However, in addition to these oscillations, there is a clear progressive rise in temperature, as more hot gas is fed to the second section of the reactor and hence more heat accumulates. This medium-term dynamic response is the one required by the control system. In order to have a better representation of this dynamic component of the response, the temperature signal has been cycle-average and then filtered. The results of the filtered average temperature at the reactor centre are shown in Figure 4a (as symbols).

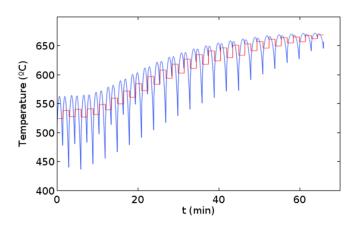


Figure 3. Dynamic response of the reactor centre temperature to a step change in the fraction of the hot gas purge of -0.04 (from 0.135 to 0.095): instantaneous temperature (—) and discrete cycle-average temperature (—).

Results of a single stimulus-response test, e.g. that of Figure 4a, can be used to fit a simple model, but the resulting model would be useful only near the conditions tested. This can be an important drawback, particularly for systems with non-linear dynamics. To extend the application range of the model, additional stimulus-response tests have been done, as summarized in Table 2. In a similar way, tests have been carried out for changes in methane feed concentration (Δy_0), which evaluates the influence of this disturbance on the dynamics of the system. In Table 2, the nominal values of the variables are included, together with the changes considered to characterize the dynamics of the system toward that variable. As shown, these tests have covered a broad range of positive (valve opening) and negative (valve closing) changes in the fraction of the hot gas purge (Δf).

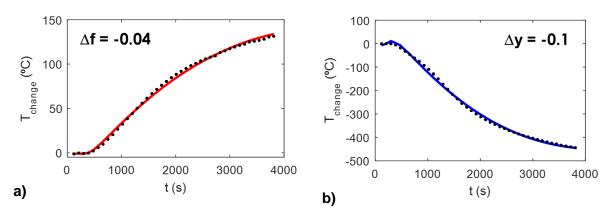


Figure 4. Dynamic response of the reactor central temperature to a step change in a) the fraction of hot gas purge of -0.04 (from 0.135 to 0.095) and b) methane concentration of -0.1% vol. (from 0.4 to 0.3% vol.). Filtered temperature (···) and ARX model predictions (—) and (—).

Table 2. Summary of the step changes performed to characterize the dynamics of the reactor by the stimulus-response test.

Variables	Fraction of hot gas purge (f)	Methane feed concentration (y ₀) (vol. %)	
Nominal	0.135	0.4	
Step changes	0.02	0.1	
	-0.02	-0.1	
	-0.04	0.05	
	-0.06	-0.05	
	-0.09		

Considering the results of all the stimulus-response tests, the following autoregressive with exogenous term model (ARX) has been fitted using MATLAB software:

$$\Delta T_{k} = -66.18 \Delta f_{k-2} + 29.56 \Delta f_{k-3} + 35.98 \Delta f_{k-4}$$

$$+ 80.45 \Delta y_{k-1} - 56.73 \Delta y_{k-3} - 23.87 \Delta y_{k-4}$$

$$+ 1.744 \Delta T_{k-1} - 0.1119 \Delta T_{k-2} - 1.032 \Delta T_{k-3} + 0.3996 \Delta T_{k-4}$$

This proposed simplified model is a discrete model; this means that the model is evaluated only at predefined time intervals (k), rather than continuously. Given the characteristic cycle nature of regenerative oxidizers, the chosen step time is equal to the switching time (in this work, maintained constant at 90 s). The model is able of predicting changes in the centre temperature, ΔT_k , at a given time k, as a function of the observed changes in the fraction of hot gas purge in previous times, Δf_{k-2} , Δf_{k-3} ..., and methane concentration, Δy_{k-1} , Δy_{k-3} ... The model is autoregressive, which means that changes of temperature in previous step times, ΔT_{k-1} , ΔT_{k-2} ...,

are also required. The model is of order 4, since information of the previous four time steps is required.

The performance of the model can be assessed in Figure 4a and b, where a case of overheating and a case of extinction are chosen as representative examples. As shown, the model is able of predicting the central temperature in both situations very well. Thereby, the model is ready to be used in the design of the control systems.

3.3.2 Feedback controller

The feedback control is based on the measurement of the controlled variable, the reactor central temperature in this case. The control variable is used to determine the control error, i.e., the difference between the actual temperature and the desired one (or set-point). The control action is calculated as a function of this error according to the control algorithm and executed on the manipulated variable (the fraction of hot gas purge, adjusted by opening or closing a valve). Controlled and manipulated variables form a loop, characteristic of feedback controllers.

The control algorithm most commonly used by feedback controllers is called PID algorithm, because it can execute up to three different types of control actions: proportional (P), integral (I) and derivative (D). As explained in the previous section, temperature is measured continuously, averaged on every cycle period and filtered to obtain the intrinsic mid-term dynamics that the controller must deal with. Hence, a new temperature value is available for the controller only every cycle period (Δt equal to the switching time, 90 s). For this reason, the discrete version of the PID control algorithm should be used (Seborg et al., 2011):

$$f_k = f_{k-1} + K_C \left[(e_k - e_{k-1}) + \frac{\Delta t}{\tau_I} e_k + \frac{\tau_D}{\Delta t} (e_k - 2e_{k-1} + e_{k-2}) \right]$$

Where f is the manipulated variable (fraction of hot gas purge), e is the control error (difference between the actual temperature and the set-point) and K_C , τ_I and τ_D are the controller parameters, that must be calculated prior to the use of the controller in a procedure called *controller tuning*. In the brackets, the first term corresponds to the proportional action P, the second to the integral I, and the third to the derivative D. If the derivative action is not included, the controller is called PI.

A typical criterion to carry out the tuning is the minimization of the integral timed-absolute error (ITAE) when a disturbance is introduced into the system (in this case, an increase in methane concentration of $\Delta y = +0.1$ is used). The mathematical formulation corresponds to the following optimization problem:

$$\min_{K_c, \tau_I, \tau_D} ITAE = \sum_j t_j |e_j| \Delta t$$

This optimization problem has been solved using the simple dynamic model obtained in the previous 'system identification' section. All the calculations have been done using a MATLAB code. The controller parameters (K_C , τ_I and τ_D) calculated with this method are displayed in Table 3 for PI and PID feedback controllers.

Table 3. Feedback controller parameters tuned by minimization of the ITAE for a methane feed concentration change of +0.1% vol.

Controller	Kc	τ _ι (s)	τ _D (s)
PI	- 0.0037	820	
PID	- 0.0051	325	123

The evolution of the average central temperature and the fraction of hot gas purge, corresponding to the optimum (tuned) controller parameters, are depicted in Figure 5. Hence, temperature initially increases due to the excess of heat accumulated in the reactor. The control action overcome this build-up by increasing the hot gas purge. The PID controller acts faster and temperature is maintained below 550°C. The new steady state is also achieved earlier with the PID controller and with very little oscillation. This is due to the derivative control action, which is able of incorporating to the control algorithm the measurement of the change rate of the error. However, the feedback controller acts only once the disturbance has produced a marked influence in the controlled variable. Given that the controlled variable is cycle-averaged, it takes two cycles (i.e. 180 s) for the control to have the updated information necessary to calculate and implement a change in the control action. To overcome this issue and provide theoretically faster responses, the use of model predictive controllers is suggested.

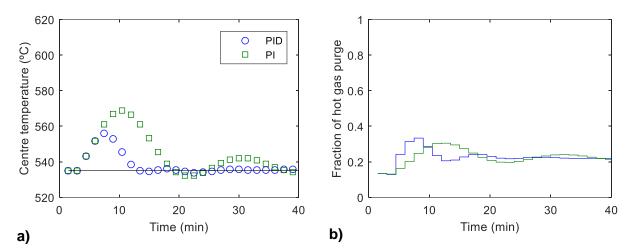


Figure 5. Response of the reactor provided with PID or PI controllers tuned using the ITAE method to a disturbance in methane feed concentration of $\Delta y_0 = +0.1\%$ vol. Controllers: (O —) PID and (\Box —) PI. a) Controlled variable (centre temperature). b) Manipulated variable (fraction of hot gas purge).

3.3.3 Model predictive controller

Instead of using fixed control algorithms with tuneable parameters, like the PID algorithm, model predictive controllers (MPC) incorporate to the control algorithm a model of the process able of predicting future changes in the controlled variable. As shown in the box diagram of Figure 6, MPCs predict what will happen to the process for a given measured disturbance, so they are capable of anticipating the response to overcome this disturbance. For this reason, they are theoretically faster than traditional feedback controllers.

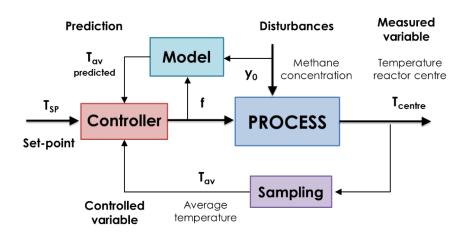


Figure 6 – Box diagram of a model predictive controller (MPC).

The objective of the MPC control algorithm is to determine a sequence of control moves, so that the predicted response is the closest to the set-point. The algorithm is based on the following optimization problem. An entire set of values of the manipulated variable $(f_{k+1}, f_{k+2}, \dots f_{k+M})$ are determined for the following M cycles, based on the minimization of the integral timedabsolute error (ITAE) (Seborg et al., 2011).

$$\min_{f_{k+1}, f_{k+2} \dots} ITAE = \sum_{j=k+1}^{k+P} t_j |e_j| \Delta t$$

Where $e_j = T_{SP} - T_j$ is the difference between the set point and model predictions and $T_j = \text{Model}\left(f_j, y_j\right)$ is the temperature predicted in the future by the simplified model over the prediction horizon P (in this case P has been fixed to 5). Thus, these predictions account for both the influence of the control moves and the measured disturbance. Since disturbances change upon time, the previous optimization problem must be solved every cycle to take into account the updated measurements and calculate the new control moves. This is an important difference with respect to feedback controllers, where the optimization problem is solved only once (in the controller tuning). These calculations have been implemented in a MATLAB code.

A good dynamic model of the process is critical for MPC, as the success of the controller to determine good control moves depends on the capability of the model to give good predictions of the output variable. In addition, the model must be simple (and fast to solve), because the optimization problem must be solved at every sampling interval, each one requiring many

evaluations of the model. The optimization problem should be solved by the computer in charge of the implementing the control action in seconds; otherwise, the information will become outdated and the control algorithm will introduce important time lags in the control action.

3.4 Simulation of the controlled regenerative catalytic oxidizer

In this section, the performance of the feedback and MPC controllers explained in the previous section is tested by simulating step disturbances of methane feed concentration.

3.4.1 Feedback controller

Figure 7 shows how the feedback PID and PI controllers perform for a disturbance in methane feed concentration of +0.2% vol. (from 0.4 to 0.6% vol.). It should be noted that the tuning of the feedback controllers was carried out assuming a disturbance of +0.1% vol. Hence, the simulation of the controlled process for a disturbance of higher magnitude will test the capability of the controller. The response of the system in the absence of controller is also depicted.

The results of Figure 7 shown that the PID controller is faster than the PI controller. For this reason, the peak temperature is lower for the PID controller (577°C against 602°C) and the time required to achieve a new steady state is shorter (29 min against more than 40 min). The PI feedback controller is based on the simplest algorithm (only with proportional and integral action), leading to a marked underdamped dynamic response (e.g. temperature decreasing to the set point with an oscillatory response). The hot gas purge (i.e. valve opening) calculated by the controller evidences the superiority of the PID controller to respond to the disturbance: the response on the valve is faster and the new steady state value of the fraction of gas purge is calculated accurately.

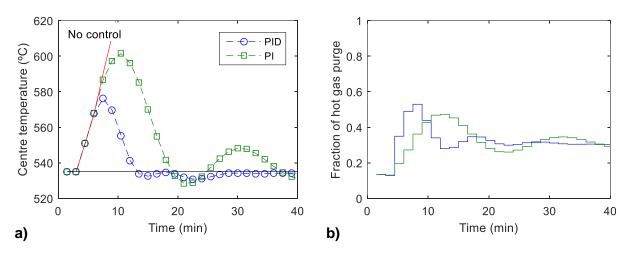


Figure 7. Simulation of the response of the feedback controllers to disturbances of +0.2% vol. in methane feed concentration (from 0.4 to 0.6% vol.). Controllers: (- -) no control, (○ --) PID and (□ --) PI. a) Controlled variable (centre temperature). b)

Manipulated variable (fraction of hot gas purge).

The influence of the controller gain (Kc) on the response of the PID controller has been evaluated by means of a sensitivity analysis. Figure 8 shows the simulated dynamic responses for a decrease and an increase of 25% in the controller gain with respect the optimum value (optimum $K_C = -0.00507$, as determined in section 3.3.2). The same disturbance of +0.2% vol. in methane feed concentration is introduced, like in Figure 7. On decreasing 25% the controller gain (to $K_C = -0.0038$), the response becomes slower (i.e. less aggressive). Thus, when methane feed concentration is increased, the increase in the fraction of hot gas purge is lower. As a result, the peak temperature and the time required to reach the new steady state are a bit higher (temperature increases from 577 to 580°C and time increases from 29 to 31.5 min).

When the controller gain is increased 25% (to $K_C = -0.00634$), the controller becomes more aggressive. This means that the fraction of hot gas purge will be manipulated to a higher extent in order to compensate changes in disturbances. As a consequence, the response is fast and the peak temperature is reduced. However, this outcome is obtained at the expense of an increase in the oscillations of the response, which means a very high time is needed to reach the new steady state (more than 40 min). This behaviour is characteristic of underdamped dynamic systems.

A lower controller gain has the advantage of reducing the oscillations of the dynamic response, which is interesting for regenerative oxidizers. Thus, when the valve that sets the fraction of hot gas purge is continually adjusted to high or low values, due to the oscillations of the underdamped dynamic response (see Figure 8b), the heat storage in the regenerative oxidizer is highly disturbed. This can have negative consequences to the stability of the oxidizer and, hence, it is preferable having a slower and safer response.

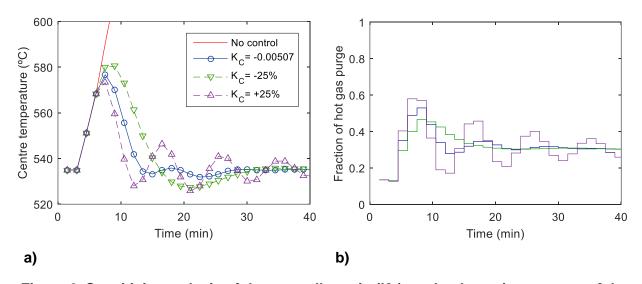


Figure 8. Sensitivity analysis of the controller gain (K_c) on the dynamic response of the PID feedback controller. Disturbance: +0.2% vol. in methane feed concentration (from 0.4 to 0.6% vol.). Controller gain: (\bigcirc —) K_c = -0.00507, (\bigvee —) K_c = -0.00380 (-25%) and (\triangle —) K_c = -0.00634 (+25%). a) Controlled variable (centre temperature). b) Manipulated variable (fraction of hot gas purge).

3.4.2 Model predictive controller

In a similar way to the PID feedback controller, the performance of the MPC has been analysed by means of simulations using two step disturbances in methane feed concentration (+0.1 and +0.2% vol.). Figure 9 shows that the dynamics of the reactor is very similar for both disturbances, temperature returning to the set-point value in less than 10 min. As shown in the figure, in the absence of control, temperature would increase above 600°C very fast (less than 12 min). The main differences between both disturbances are observed in the peak temperature, which is higher for the case of the higher concentration step (+0.2% vol.). In this case, the controller is able of overcoming this disturbance by means of a higher increase in the fraction of hot gas purge (up to 0.80). Once temperature decreases, the controller is able of reducing the fraction of hot gas purge to a lower value to prevent an excessive drain of energy from the reactor. The new steady state value required for the fraction of hot gas purge is calculated by the controller accurately.

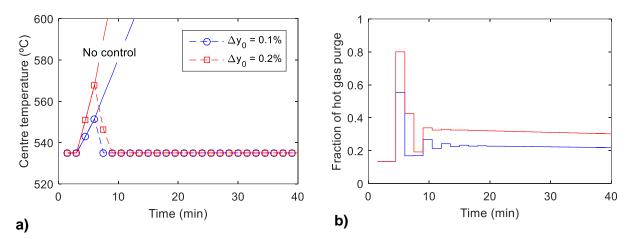


Figure 9 – Simulation of the MPC for the following disturbances in methane feed concentration: (○ —) +0.1% and (□ —) +0.2% vol. a) Controlled variable (centre temperature). b) Manipulated variable (fraction of hot gas purge)

3.4.3 Comparison of PID feedback controller and MPC

The main differences between PID feedback controllers and MPC are discussed in this section. Figure 10 compares the controllers for an increase in methane concentration of +0.2% vol. (from 0.4 to 0.6% vol., which represents a 50% -relative- increase in methane concentration, above what is expected in practical operation). It can be observed that the MPC is faster: temperature returns to the desired value in less than 10 min. In the same way, the maximum temperature achieved in the reactor is lower (568 and 577°C, respectively, for the MPC and PID controllers). Table 4 summarizes the main performance parameters of the controllers when methane feed concentration is increased or decreased, such as, the error, the maximum and minimum temperature differences or the time required to reach a new steady state.

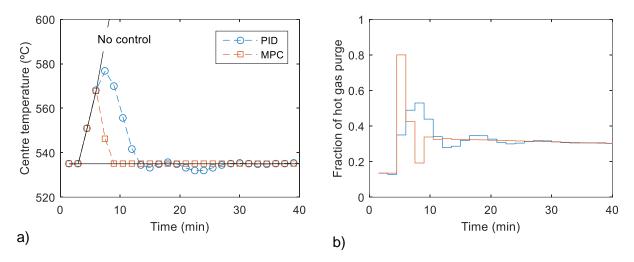


Figure 10. Comparison of MPC (\square —) and PID (\bigcirc —) feedback controller for a disturbance of +0.2% vol. in methane concentration (from 0.4 to 0.6% vol.) a) Controlled variable (centre temperature). b) Manipulated variable (fraction of hot gas purge)

Table 4. Comparison of the performance of the PID feedback controller and MPC.

	Change in methane feed concentration				
	+0.2% vol.		-0.05	% vol.	
	PID	MPC	PID	MPC	
ITAE x 10 ⁻⁶	8.3	1.9	5.0	0.57	
ΔT_{max} (°C)	42	33	1	0	
ΔT_{min} (°C)	3	0	10	8	
t _{error ±1°C} (min)	29	9	29	9	

The reason for the fast response of the MPC is the anticipation capability. Once methane concentration increases, the process model incorporated in the control algorithm can predict the future increase in temperature. For this reason, the control action starts to overcome the disturbance in the following cycle after detecting the concentration change. This can be observed in the valve opening calculated by the controller, which raises to 0.80 to drain to excess of heat. This aggressive action is the reason for the fast response of the MPC. On the contrary, the PID feedback controller needs a temperature error to trigger the control action.

The PID feedback controller exhibits an underdamped response, but within a very small temperature range and, hence, this has no negative effect in the performance of the reactor. The best controller in this regard is the MPC, because the fraction of hot gas purge is adjusted at every step time to the optimum value. As a result, it does not show the underdamped response observed for the PID controller.

The performance of the controllers has also been compared for a decrease in methane concentration of 0.05% vol. (from 0.4 to 0.35% vol.). Figure 11 shows the dynamic response of the reactor temperature and the fraction of hot gas purge. As observed, after the introduction of

the concentration change, reactor temperature decreases. The heat released by the reaction is lower and the control system must change the fraction of hot gas purge to prevent the extinction of the reactor (conversion drop to zero). The controllers deal with this disturbance by reducing the fraction of the hot gas purge from an initial value of 0.135 to zero (or very close to zero), so that more heat accumulates in the reactor, compensating the decrease of heat released by the reaction. This control action produces an increase in the reactor temperature, approaching the set-point.

The time and value of the minimum temperature reached by the reactor can be used as an indicator of the performance of the controllers, as summarized in Table 4. Thus, the MPC is the controller that performs the best: temperature never decreases below 527°C, and returns to the set point in 9 min. This response is so fast because the controller closes completely the valve of the hot gas purge during a few cycles to prevent a critical heat drainage from the reactor. The PID controller performs also well, being only slightly slower than the MPC: the temperature set point is reached after 29 min, but being very close after 12 min. The response is slightly underdamped, but with temperature oscillating within a narrow range.

Both controllers calculate the fraction of hot gas purge required to reach the new steady state correctly. The MPC does this calculation faster, but at the expense of aggressive changes in the valve regulating the fraction of hot gas purge.

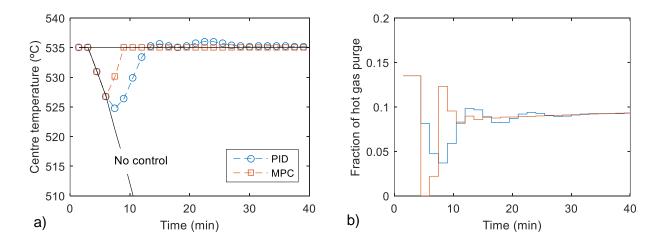


Figure 11. Comparison of MPC (□ —) and PID (○ —) feedback controller for a disturbance of -0.05% vol. in methane concentration (from 0.4 to 0.35% vol.). a) Controlled variable (centre temperature). b) Manipulated variable (fraction of hot gas purge)

According to the simulations, the MPC is the controller that performs the best in both situations found in regenerative oxidizers, an increase and a decrease in methane feed concentration. However, it is more complicated to implement, as the control algorithm is not a simple function, but a complete minimization problem that calculates the value of the manipulated variable at each step time. For this reason, a computer is required to make all the calculations, together with skilled operating and maintenance personnel. On the other hand, the PID controller is much easier to implement and supervise, its performance being also satisfactory, and only a bit worse (the response time is a bit slower and the dynamics slightly underdamped).

4 Conclusions

Regenerative oxidizers require the use of controllers to adjust the operating variables, as a function of the operating conditions (mainly, feed concentration), which can change with time. The selected controlled variable is the temperature in the middle point of the oxidizer, a good indicator of reactor overheating or extinction (to be prevented by the control system). The change of the fraction of hot gas purge from the centre of the oxidizer is a suitable manipulated variable for the controller, affecting the energy storage in the reactor and, hence, being able of overcoming the increase/decrease of feed concentration.

Three controller types have been designed and simulated for the case of a regenerative oxidizer used in the treatment of coal mine ventilation air methane. The proportional-integral (PI) and proportional-integral-derivative (PID) feedback controllers have a control algorithm easy to implement with two (PI) or three (PID) tuneable parameters. These parameters have been calculated, so that the integral timed-absolute error (ITAE) is minimized when a change in methane feed concentration is introduced. Since central temperature varies during a cycle, due to the inherent dynamics of the oxidizer, discrete cycle-averaged central temperatures are calculated by the controller every cycle time. For this reason, all the responses of the controller have a lag time equal to the cycle time. Considering this, the simulations of the PID controller showed good and fast responses, being able of dealing with high and low concentration disturbances.

The model predictive controller (MPC) is more complicated, incorporating into the control algorithm a full optimization problem based on a simple process model. This controller requires a powerful control hardware (computer) to make all the calculations on time (every cycle time). The performance of the controller is a bit better than that of the PID controller, mainly in terms of velocity response, but it is more complex to implement and operate.

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