UNIVERSIDADE DE SÃO PAULO
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FABRÍCIO FEIJÓ DE LIMA

Temperature control strategies comparison: a case study of an industrial semi-batch hydrogenation reactor

Comparação de estratégias de controle de temperatura: um estudo de caso de um reator de hidrogenação industrial semi-batelada

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Temperature control strategies comparison: a case study of an industrial semi-batch hydrogenation reactor

Dissertation presented to Engineering School of Lorena – University of São Paulo to obtain the title of Master of Science of the Postgraduate Program in Chemical Engineering in the concentration area of Product and Process Development.
Advisor: Profª. Drª. Daniela Helena Pelegrine Guimarães

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ABSTRACT

FEIJO DE LIMA, FABRICIO. **Temperature control strategies comparison: a case study of an industrial semi-batch hydrogenation reactor.** 2020. 124p. Dissertation (Master of Science) – Escola de Engenharia de Lorena, Universidade de São Paulo, Lorena, 2020.

Nowadays, semi-batch processes are widely used due to their high added-value products and their production flexibility. Therefore, the optimization of this kind of process plays a major role to guarantee economic performance of the business. In this work, an industrial hydrogenation reactor from a multinational chemical company located at São Paulo state in Brazil was mathematically modeled and then simulated at MATLAB. The model, based on the reaction's kinetics, and mass and energy balances, was validated through real plant data and then, the open-loop dynamic of the process was evaluated through a model sensitivity analysis. In the light of model sensitivity analysis, three new temperature control strategies were proposed: cascade, override and cascade+override. These techniques were then compared with each other and with the current feedback controller of the industrial hydrogenation reactor. As a result, the cascade controller showed good results regarding not only temperature tracking but also for product quality and then was chosen as the most suitable for this case study. In addition, several future work proposals were done, aiming the use of tools to refine the model, use of the model to perform operational training and process safety analysis, development and comparison with other control strategies, and investigation of anti-reset windup and controller's tuning methodologies.

Keywords: Hydrogenation. Semi-batch reactor. Temperature control strategy.

RESUMO

FEIJO DE LIMA, FABRICIO. **Comparação de estratégias de controle de temperatura: um estudo de caso de um reator de hidrogenação industrial semi-batelada.** 2020. 124p. Dissertação (Mestrado em Ciências) – Escola de Engenharia de Lorena, Universidade de São Paulo, Lorena, 2020.

Nos dias de hoje, os processos semi-batelada são amplamente utilizados devido ao grande valor agregado de seus produtos e a flexibilidade de produção. Portanto, a otimização deste tipo de processo é muito importante para garantir a performance econômica do negócio. No presente trabalho, um reator de hidrogenação industrial de uma empresa química multinacional localizada no estado de São Paulo no Brasil foi modelado matematicamente e depois simulado no MATLAB. O modelo, baseado na cinética da reação e nos balanços de massa e energia, foi validado através de dados reais da planta e então, a dinâmica do processo em malha aberta foi avaliada através de uma análise de sensibilidade do processo. Sob a luz da análise de sensibilidade, foram propostas três novas estratégias de controle de temperatura: cascata, override e cascata+override. Estas técnicas foram então comparadas entre si e com o atual controle feedback do reator industrial de hidrogenação. Como resultado, o controlador cascata apresentou bons resultados não apenas para o controle de temperatura, mas também, para a qualidade do produto e então, foi escolhido como o mais adequado para este estudo de caso. Adicionalmente, diversas propostas de trabalhos futuros foram feitas, focando no uso de ferramentas para refinar o modelo, uso do modelo para executar treinamentos operacionais e análises de segurança de processos, desenvolvimento e comparação com outras estratégias de controle e investigação de metodologias de anti-reset windup e sintonia de controladores.

Palavras-chave: Hidrogenação. Reator semi-batelada. Estratégia de controle de temperatura.

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ABBREVIATIONS LIST

APCS	Adaptative Predictive Control Systems
CV	Controlled Variable
DMC	Dynamic Matrix Control
EOT	Estimation-Optimization Task
GMV	Generalized Minimum Variance
GMC	Generic Model Control
IMC	Internal Model Control
ISA	International Society of Automation
MV	Manipulated Variable
MPC	Model Predictive Control
MPHC	Model Predictive Heuristic Control
MIMO	Multiple Input Multiple Output
NMPC	Non-Linear Model Predictive Control
NLP	Non-Linear Programming
PVC	Polyvinyl Chloride
PLC	Programmable Logic Controllers
PI	Proportional and Integral
PID	Proportional, Integral and Derivative
STR	Self-Tuning Regulators
SAC	Stable Adaptative Controller
STR	Stirred Tank Reactor
SLP	Successive Line Program
SQP	Successive Quadratic Program
FOPDT	First Order Plus Dead Time
VPC	Valve Position Controller

SYMBOLS LIST

C_A	Concentration of A	mol/L
Cp_A	Reactant A heat capacity	J/g.K
Cp_B	Product B heat capacity	J/g.K
Cp_C	Product C heat capacity	J/g.K
$Cp_{H_{2,e}}$	Inlet hydrogen heat capacity	J/g.K
Cp_i	Substance i heat capacity	J/g.K
Cp_J	Cooling fluid heat capacity	J/g.K
Cp_R	Reactor content heat capacity	J/g.K
Cp_{sol}	Solvent heat capacity	J/g.K
C_{v,H_2}	Hydrogen control valve flow coefficient	kg/h.bar$^{0.5}$
$C_{v,J}$	Cooling fluid control valve flow coefficient	L/h.bar$^{0.5}$
F_J	Cooling fluid volumetric flowrate	L/h
F_{k,H_2}	Hydrogen specific heat ratio factor	dimensionless
F_{L,H_2}	Hydrogen control valve pressure drop correction factor	dimensionless
F_{p,H_2}	Hydrogen piping geometry correction factor	dimensionless
$F_{p,J}$	Cooling fluid piping geometry correction factor	dimensionless
$f(X)_{H_2}$	Hydrogen flow control valve characteristics	%
$f(X)_J$	Cooling fluid flow control valve characteristics	dimensionless
Kc	Controller gain	-
k_{H_2}	Hydrogen specific heat ratio	dimensionless
m_0	mass of the mixture reactant A + solvent coming from the previous vessel	kg
m_A	Reactant A mass	kg

m_{A0}	Initial mass of reactant A	kg
m_B	Product B mass	kg
m_C	Product C mass	kg
m_{H_2}	Hydrogen mass	kg
$\dot{m}_{H_{2,e}}$	Inlet hydrogen mass flowrate	kg/h
$\dot{m}_{H_{2,e,SP}}$	Hydrogen mass flowrate set point	kg/h
m_i	Substance i mass	kg
m_{kat}	Catalyst mass	kg
MM_A	Reactant A molecular mass	g/mol
MM_B	Product B molecular mass	g/mol
MM_C	Product C molecular mass	g/mol
MM_{H_2}	Hydrogen molecular mass	g/mol
MM_i	Substance i molecular mass	g/mol
MM_{SOL}	Solvent molecular mass	g/mol
m_R	Reactor total mass	kg
m_{sol}	Solvent mass	kg
N_A	Number of mols of reactant A	mol
N_{A0}	Initial number of mols of reactant A	mol
N_B	Number of mols of product B	mol
N_{B0}	Initial number of mols of product B	mol
N_C	Number of mols of product C	mol
N_{C0}	Initial number of mols of product C	mol
N_{H_2}	Number of mols of hydrogen	mol
N_{H_20}	Initial number of mols of hydrogen	mol
$\dot{N}_{H_{2,cons}}$	Consumed hydrogen molar flowrate	mol/h
$\dot{N}_{H_{2,e}}$	Inlet hydrogen molar flowrate	mol/h
N_i	Number of mols of substance i	mol
N_{SOL}	Number of mols of solvent	mol
$P_{H_{2,e}}$	Inlet hydrogen pressure	Bar
P_R	Reactor Pressure	bar
P_{R_0}	Reactor initial pressure	bar
Q_J	Heat exchanged trough the jacket	J/h

Q_R	Reaction heat	J/h
R	Universal gas constant	J/mol.K
$-r_A$	Reaction rate in terms of reactant A	-
T_{eJ}	Cooling fluid inlet temperature	ºC
$T_{H_{2,e}}$	Inlet hydrogen temperature	K
T_J	Jacket temperature	ºC
T_{J_0}	Jacket initial temperature	ºC
T_R	Reactor Temperature	ºC
$T_{R,SP}$	Reactor temperature set point	ºC
T_{R_0}	Reactor initial temperature	ºC
U	Overall heat transfer coefficient	J/h.K.m^2
$\overline{UA}$	Average global heat transfer coefficient times area	J/h.K
V_{hs}	Reactor headspace free volume	L
V_J	Jacket volume	L
V_L	Reactor liquid volume	L
V_{SOL}	Solvent volume	L
V_{TOTAL}	Reactor total volume	L
x	Dimensionless generic variable value	dimensionless
x^*	Real generic variable value	dimensionless
x_{A0}	mass fraction of reactant A in the mixture A + solvent coming from the previous vessel	kg/kg
x_{eff,H_2}	Hydrogen control valve chocked flow pressure drop ratio	dimensionless
X_{H_2}	Hydrogen flow control valve opening	%
X_J	Cooling fluid control valve opening	%
x_{kat}	Catalyst mass fraction	kg/kg
x_{ref}	Reference generic variable value	-
x_{T,H_2}	Hydrogen control valve pressure drop ratio	dimensionless

Y_{H_2}	Hydrogen expansion factor	dimensionless
Z	Compressibility factor	dimensionless
ΔH_R	Reaction enthalpy	J/mol
ΔP_J	Pressure drop at cooling fluid control valve	bar
ΔT	Temperature difference	K
ρ_A	Reactant A density	g/L
ρ_B	Product B density	g/L
ρ_C	Product C density	g/L
$\rho_{H_2@P_{H_{2,e}}}$	Hydrogen density at inlet hydrogen pressure	kg/m3
ρ_J	Cooling fluid density	g/L
τ_I	Integral time	s

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1 INTRODUCTION

Batch and semi-batch processes are widely used due to their high added-value products and their production flexibility. Therefore, the optimization of these kinds of processes plays a major role to guarantee a high business economic performance. A more competitive environment and social pressure for less environmental and social impacts are heading industries towards more reliable, cost-efficient and less waste generator plants. Thus, the further development of automatic process control has an important role in this mindset and technological change to guarantee the quality, safety, and economic performance.

In this actual context pointed before, the control of batch and semi-batch reactors temperature is very important and plays a fundamental role to achieve desired product specification and guarantee reaction thermal safety. However, batch and semi-batch reactors can be very hard to control especially due to their inherent dynamic nature what leads the process variables and parameters to be not constant in the time, such as, temperature, pressure, conversion, heat of reaction, viscosity, density, and specific heat among several others. Furthermore, these reactors usually have a nonlinear behavior because the heat generation term in energy balance has a strong dependency on temperature and follows Arrhenius law.

Although many control approaches had been proposed as alternatives to the drawbacks of PID traditional strategy, a question remains: how deeply the PID strategy was explored and optimized to be comparable to the more advanced techniques? (ROTSTEIN; LEWIN, 1992).

1.1 Objectives

The goal of this work was to perform a case study of temperature control strategies applied to an industrial hydrogenation reactor from a multinational chemical company and has the following specific objectives:

a) Modeling an industrial jacketed STR (Stirred Tank Reactors) in a semi-batch hydrogenation reaction;
b) Validate the model with real plant data;
c) Understand process behavior through model sensitivity analysis;

d) Implement four different temperature control strategies in the simulation environment;
e) Compare temperature control strategies’ responses in the simulation environment focusing on standard and non-standard operation conditions, disturbance handling, reduction of batch time and quality parameters.

2 LITERATURE REVIEW

2.1 Hydrogenation reactions

The earliest register of hydrogenation reactions applied to the production of a marketable product comes from 1823, when the Döberener's Lamp, developed by the German chemist Johann Wolfgang Döberener, was commercialized as a result of a platinum-catalyzed reaction of hydrogen and oxygen. Later in 1897, the French scientist Paul Sabatier performed an inedited hydrogen addition to hydrocarbons by a nickel-catalyzed gas-phase reaction. In 1902, the German chemist Wilhelm Normann received a patent for liquid-phase hydrogenation process reaction and enabled a huge application of it at oil and fat industries. The new process was commercialized in the United Kingdom at Joseph Crosfield & Sons and in Germany at Ölwerke Germania. After that, over the first two decades of the 20th century, catalyst hydrogenation had an important role in the development of major processes as the production of methanol, ammonia (Haber-Bosch process) and liquid hydrocarbons (Fischer-Tropsch process) (JACKSON, 2018).

The hydrogenation reactions consist of reduction reactions, that uses hydrogen as a reactant to add it to a chemical compound (BRUICE, 2015).

The catalytic hydrogenation is a type of reaction that adds hydrogen to the double or triple bonds of an organic compound, in the presence of a catalyst, generally of metallic nature, for example, palladium (Pd), nickel (Ni), platinum (Pt). The presence of the metallic catalyst weakens the bond $H-H$ of the hydrogen (H_2), which is very stable when the catalyst is not present. The catalytic hydrogenation mechanism is complex, but it is known that the hydrogen is adsorbed on the metal surface, creating two radicals ($H\cdot$) (homolytically break), and that the organic compound also interacts with the metallic surface weakening its double or triple bond. The formation of the unstable compounds and the approximation of reactants enable the occurrence of the chemical reaction that takes place, consequently, also on the metal surface. This type of reaction is intrinsically exothermic, and the released heat is referred to as the heat of hydrogenation. The more stable the double or triple bond of the organic compound is, smaller is the heat of hydrogenation (BRUICE, 2015).

According to (BRUICE, 2015), the catalytic hydrogenation can reduce double or triple bonds to single bonds of different organic compounds:

a) Carbon-Carbon double or triple bonds resulting, for instance, on the reduction of alkenes to alkanes; alkynes to alkenes and then to alkanes; benzene to cyclohexane, etc.
   Example:

$$CH_3CH = CHCH_3 + H_2 \xrightarrow{Pd/C} CH_3CH_2 - CH_2CH_3$$

b) Carbon-Nitrogen double and triple bonds resulting, for example, on the reduction of imines and nitriles to primary amines.
   Example:

$$RC \equiv N + 2H_2 \xrightarrow{Raney\ Nickel} RCH_2 - NH_2$$

c) Carbon-Oxygen double bond resulting, for instance, on the reduction of aldehydes to primary alcohols, ketones to secondary alcohols, etc.
   Example:

$$R - C = O - H + H_2 \xrightarrow{Raney\ Nickel} R - CH_2 - OH$$

As the resonance in aromatic compounds stabilizes the double bonds, the reduction of aromatic compounds double bond occurs only at high temperature. In that manner, the use of catalytic hydrogenation to reduce, and then modify, only the aromatic compounds substituents, is another application of this kind of reaction. It can change (BRUICE, 2015):

a) Nitro substituent to an amino substituent:

$$C_6H_6 - NO_2 + H_2 \xrightarrow{Pd/C} C_6H_6 - NH_2$$

b) Nitrile substituent to amino substituent:

$$C_6H_6 - C \equiv N + 2H_2 \xrightarrow{Raney\ Nickel} C_6H_6 - CH_2NH_2$$

c) Alkylidene groups to alkyl group:

$$C_6H_6 - CH = CH_2 + H_2 \xrightarrow{Pd/C} C_6H_6 - CH_2CH_3$$

Nowadays, the hydrogenation reaction is widely used in several industries:

a) Food industry: highly used to transform polyunsaturated fatty acids into fats, as margarine (GUNSTONE, 2009);

b) Energy industry: biofuel stabilization (SOUZA *et al.*, 2012), light hydrocarbons production via biomass (HUBER *et al.*, 2005; SOTELO-BOYÁS; LIU; MINOWA, 2011), conversion of alkanes and aromatics into paraffins and naphthenes (KONG *et al.*, 2015), hydrocracking of petrol heavy residues (SAHU *et al.*, 2015).
c) Fine chemistry industry: oxo process (DU *et al.*, 2016), amines process (LANGE *et al.*, 2016);
d) Pharmaceutical Industry (KLINGLER, 2007).

2.2 Process classification

The chemical industry is plural regarding types of processes and each process has many individual characteristics being that one of the most important facets of a process is its mode of operation.

Many researchers and industry professionals have a consensus regarding the classification of the two opposite modes of operation: batch vs continuous.

A batch process is the one where there is no input or output of material during the operation time (FELDER; ROUSSEAR; BULLARD, 2016; HARRIOTT, 2003; LUYBEN, 2007; MISSEN; MIMS; SAVILLE, 1999; ÖZGÜL EVRANUZ; KILIÇ-AKYILMAZ, 2012). For instance, substance A and B are added to a reactor and remain there until the end of the reaction. Other common examples of batch processes are a washing machine, kitchen oven, coatings, artisanal spirits production, beverages, pharmaceutical products, fine chemistry industries, etc (FELDER; ROUSSEAR; BULLARD, 2016; HARRIOTT, 2003; LUYBEN, 2007; MISSEN; MIMS; SAVILLE, 1999; ÖZGÜL EVRANUZ; KILIÇ-AKYILMAZ, 2012).

On the other hand, a continuous process is the one where the input and output flows are permanent throughout the operation time (FELDER; ROUSSEAR; BULLARD, 2016; HARRIOTT, 2003; LUYBEN, 2007; MISSEN; MIMS; SAVILLE, 1999; ÖZGÜL EVRANUZ; KILIÇ-AKYILMAZ, 2012). For instance, a good example is a petrochemical distillation column which is continuously fed with nafta and has several continuous output streams.

However, when it comes to a process which is not fully batch or fully continuous, it seems that there is no agreement regarding its classification as can be seen in the following paragraphs. At least three other classifications can be found in literature: semi-batch, semi-continuous and fed-batch.

Missen, Mims and Saville (1999) formally described four kinds of processes: batch, semi-batch, semi-continuous and continuous. The authors define a semi-batch reactor operation as an intermittent or continuous addition of one reactant to another one which is already inside the reactor or a product that is removed intermittently or continuously as the reaction occurs. On the other hand, a semi-continuous reactor is defined as a multiphase reactor in which one phase is already inside the reactor while the other phase flows continuously through the reactor, for example in a fixed-bed catalytic reactor or a fixed-bed gas-solid reactor.

Harriot (2003) mentions only three kinds of processes: batch, semi-batch and continuous. He interprets a semi-batch reactor as a variation of a batch reactor in which one reactant is already charged into the reactor and the second is fed continuously or as frequent pulses during the batch cycle.

Korovessi and Linninger (2006) presented a second interpretation for the semi-continuous process. The authors defined as semi-continuous process as the one where several machines are working in staged batch times, as in solid/liquid separation processes.

Luyben (2007) also classified processes as batch, semi-batch and continuous. However, he differentiated two types of semi-batch reactors. The first one was nominated as fed-batch where an initial charge of reagent is done into the reactor, but another substance (reactant or catalyst) is continuously fed. The second type of semi-batch reactor works with the continuous removal of some material during the batch. As examples, he pointed the ethanol fermenters in which carbon dioxide is continuously removed from the reaction throughout batch time.

Özgül and Kiliç-Akyilmaz (2012) classified processes as batch, semi-continuous or continuous. The authors defined a semi-continuous process as any process that is neither batch nor continuous and also mention the term semi-batch as a synonymous for semi-continuous.

Felder, Roussear and Bullard (2016) classified processes as batch, semi-batch and continuous. The authors use the same definition for the semi-batch process as Özgül and Kiliç-Akyilmaz (2012).

Some others researches (BRESAOLA *et al.*, 2019; LI *et al.*, 2019) defined a semi-continuous reactor when it starts as a batch but at some time, part of the content inside the reactor is removed and restoration is done by adding fresh product. Usually, this process is done in cycles.

In general, when a process is not a batch neither continuous, its classification may vary. In recent literature, each classification seems to be used depending of the kind of process as can be seen in Table 1.

Table 1 - Different mode of operation classification

Mode of operation classification	Process	References
Semi-batch	Fine chemistry, pharmaceutical industry	(ÁLVAREZ *et al.*, 2019; JANG; LEE; BIEGLER, 2016; MALIK *et al.*, 2019)
Fed-batch or pulse-fed-batch	Biotechnology industry	(FAN *et al.*, 2015; KEIL *et al.*, 2019; ÖZTÜRK; ÇALIK; ÖZDAMAR, 2016)
Semi-continuous	Petrochemical, biotechnology, food industry and waste water treatment	(ABAIDE *et al.*, 2019; ASHRAF *et al.*, 2019; BOONPRASOP; CHALERMSINSUWAN; PIUMSOMBOON, 2019; BRESAOLA *et al.*, 2019; LI *et al.*, 2019)

Source: Author

For this work, the term semi-batch will be used as a continuous addition of one reactant during the reaction time to another one which is already inside the reactor.

2.3 Batch and semi-batch processes

In the very beginning, this kind of process was seen as old-fashioned when compared to the big continuous ones, mostly driven by the petrochemical industry (RIPPIN, 1983). Indeed, as mentioned by Rippin (1983), the first chemical engineering students were too attached to the idea of the burden held by chemical engineers to transform batch processes into continuous ones.

However, the students, researches, and engineers have continued to struggle for several years since the technological advances applied to continuous processes were usually not applicable for batch processes (SOROUSH; KRAVARIS, 1993a).

Batch processes are applied to many branches of the chemical industry. They are usually used to produce high added-value products, like pharmaceuticals, industrialized food, pesticides, herbicides and, but not limited to, specialty chemicals (KOROVESSI; LINNINGER, 2006; SMITH, 2014).

The main features that characterize batch processes against continuous processes are (SMITH, 2014; SOROUSH; KRAVARIS, 1993a):

- Transient operation, while continuous processes operate in a steady-state;
- Large operation range due to transient behavior, while continuous processes fluctuates around the stationary point (shorter operation ranges);
- Initial conditions (loading) defines the batch trajectory, while for continuous process this is only relevant when a multiplicity of steady states exists;
- Higher production flexibility since small production runs can be planned accordingly to different strategies regarding market demands and adaptability to new products development;
- Difficulty to correct upset conditions due to batches common irreversible behavior, while continuous process can be adjusted to find the desired steady-state;
- The operation period is limited to the batch cycle, while at continuous processes, the operation period is related to maintenance issues or equipment life cycle.

Beyond the examples above, the development of batch processes came from its economic performance. For sure, this performance is determined by several factors, but Friedrich and Perne (1995), two Bayer AG engineers, highlighted three as the main objectives to achieve the economic benefit of batch processes. They are basic functionality, process safety, and optimum yield (as product quality). Moreover, they wisely highlighted a fourth very important factor which is the education of operating personnel even though many processes were already very automated.

More recently, besides the basic functionality, process safety, optimum yield, and operational expertise, Smith (2014) includes the intellectual property, strict tolerances of raw material usage and temperature control as a fundamental factor to achieve economic performance in batch and semi-batch processes.

2.4 Process control strategies for batch and semi-batch reactors

Batch and semi-batch reactors can be hard to control due to their inherently dynamic nature what leads to a not constant in time variables and parameters, such as, temperature, pressure, conversion, heat of reaction, viscosity, density, and specific heat among several others. Furthermore, these reactors usually have a nonlinear behavior because the heat generation term in energy balance has a strong dependency on temperature and follows Arrhenius law. Moreover, usually in semi-batch processes, the heat transfer area is changing while the reactor is being feed. This time-dependent heat transfer area brings additional non-linearities to the processes (SEBORG *et al.*, 2017).

The inherent nonlinearities affect the open and closed loops' characteristics responses. Therefore, it is one of the most relevant factors to stablish process control for chemical reactors. What makes the control problem analysis more difficult for batch and semi-batch processes is the fact that these processes are not operated at steady state due to a temporally varying trajectory of the controlled variable (HELBIG; MARQUARDT; ALLGÖWER, 1998).

Temperature control is essential for batch and semi-batch reactors since the chemistry behind has usually a strong dependence on temperature. So, the requirements for the controller design and the heat exchange equipment need to be very well evaluated to guarantee a robust and reliable operation (SMITH, 2014).

2.4.1 Early development

The initial development of process control strategies applied to batch and semi-batch processes started in the '60s when researchers began to test the already developed control strategies for continuous plant at batch processes.

Shinskey and Weinstein (1965) proposed a dual-mode temperature control consisting in an on-off control for the heat-up phase where full heating is applied to reach the desired temperature set point and then maximum cooling to stop further heating. After that, a conventional PID controller is used to maintain the temperature.

Millman and Katz (1967) started to evaluate a linear temperature control in batch reactors where parallel and consecutive first-order reactions were taking place. Mathematical models were proposed to maximize the yield of a given batch by choosing the best PID control parameters and strategy. The authors showed how each term of the PID framework impacted in the temperature profile and consequently in the reaction yield.

However, Luyben (1968) figure out that the nonlinearities, especially present in batch processes, could impair the effectiveness of the control and thus should be considered in the design of feedforward controllers. The author made an enriching comparison between the effectiveness of linear and nonlinear feedforward controllers applied to batch and continuous reactors. It was clearly shown that nonlinear methods improve a lot the quality of the control of chemical reactors.

Marroquin and Luyben (1972) identified that little experimental verification was done in the previous works of process control for batch processes. So, the authors proposed an experimental investigation of four types of nonlinear controllers applied to a cascade temperature control of a lab-scale batch reactor. They have demonstrated a better performance of the cascade control when the error in the primary loops varies the gain of the secondary controller when compared with standard linear control.

Rippin (1983) summarized extensive literature regarding optimal batch operation including different processes such as polymerization, enzymes, batch distillation, and batch crystallization. He exhibited and point out the pros and cons of the currently available quantitative methods for optimal batch operation. As examples of areas in which methods were available at that time were, but not limited to optimal operation of existing equipment items, design of multi-product plant at minimum equipment cost and multi-purpose plant capacity planning.

During this early period, researchers were struggling with the use of basic linear controllers to highly non-linear, and non-stationary characteristics of exothermic batch and semi-batch reactors. They introduced several techniques as dual-mode control, PID tuning assessment, non-linear strategies, and others, to overcame the firsts challenges of this kind of control. Later on, based on their achievements, many other researchers developed much more robust techniques as is shown above.

2.4.2 Middle development – Adaptative control

During the development of control strategies of batch and semi-batch process, researchers were very interested in adaptative and self-tuning techniques. This makes sense considering the dynamic and nonlinear behavior of this kind of processes since the process characteristics often change in each step and sometimes even from batch to batch. Therefore, from the 1960s to the middle 1980s, much effort was employed to study strategies of adaptive control. Approaches including Self-tuning Regulators (STR) to Adaptive Predictive Control Systems (APCS) had been developed. In this context, the understanding of control stability is of great interest for the advances in this research field and several papers were published as follows.

Kiparissides and Shah (1983) compared the use of two adaptative controllers, the self-tuning regulator (STR) and a globally stable adaptative controller (SAC) and the classical PID controller applied to a dynamic model of PVC polymerization. It was shown that even with strong nonlinear process characteristics, both strategies, STR and SAC, gave excellent results, avoiding the temperature overshoot and achieving a very stable reactor temperature. Moreover, both adaptative controllers showed to be very robust against noise in parameters.

Martin-Sánchez (1984) demonstrated the global stability of an APCS, using a general formulation of error estimation minimization, applied to a discrete linear process. The main challenges of the research are related to the presence of time delays in the process and the use of unmeasured disturbances and noise, to make the stability analysis more robust.

Cluett, Shah and Fisher (1985) were interested in this new kind of adaptative predictive control and evaluated a globally stable adaptative predictive control system applied to the PVC batch reactor investigated earlier by Kiparissides and Shah (1983). The researches were aiming to develop a control strategy to a batch reactor with a wide range of operating conditions and repeated startups and shutdowns. They highlighted the use of adaptative control as a good approach to initial tuning and also continuingly since there are often changes in batch processes.

Despite the previous advances in batch process control, for decades the focus was at continuous processes due to its large application and profitability. However, batch processes started to be seen as economically feasible and hence has offered attractive challenges for the process control research community (RIPPIN, 1989).

For that reason, over the years, the research regarding process control for batch and semi-batch processes has increased a lot and several papers have been published since then.

In 1986, a new study is published contrasting the main challenges and features of batch process control when compared to continuous process control. Due to the transient nature of batch processes and, consequently, their large operation range with no central steady-state and their different process dynamics along the batch trajectory, the controller strategy and tuning can be very difficult. However, if a well-designed control is set to batch processes, it can directly influence the batch productivity and can bring, thus, beneficial results (JUBA; HAMER, 1986).

Ponnuswamy, Shah and Kiparissides (1987) continued to evaluate theoretically and experimentally the open- and closed-loop of batch polymerization reactors, but now, it was done for the methyl methacrylate polymerization reaction. Besides the open-loop discussions where the time-optimal temperature policy was determined for a given initial initiator concentration, the researchers investigated a linear-quadratic feedback control to keep the process parameters within the optimal state trajectories even in case of processes perturbations and they obtained success.

A strategy of Adaptive Pole-Assignment Control was experimentally and computationally compared to a classical PID controller (with fixed gain) and to a Generalized Minimum Variance (GMV) Controller, applied to a discrete batch polymerization process. The Adaptive Pole-Assignment Control consists of the continuous resolution of a matrix equation (Sylvester equation) for obtaining the roots of the closed-loop system characteristic equation. As those roots are related to the control stability, the method calculates the controller transfer function so that the poles are located in the desired region. The analytical resolution of the Silvester equation represents one of the methods biggest challenges (TZOUANAS; SHAH, 1989).

Tzounas and Shah (1989) also highlights the effect of sampling time for a discrete process. The availability of fast measurements can change completely the selection of the best controller strategy. In his studies, the PID strategy is the best when large sampling times are available. The shorter the sampling time is the better the adaptative strategy performs, once it requires a continuous parameters estimation.

The control of exothermic batch reactor is of great interest of the research academy since proper temperature control is crucial to avoid thermal runaway and product off-spec. This regulatory issue can sometimes bring complex solution due to the process's strong non-linearities. Although many control approaches had been proposed as an alternative to the drawbacks of PID traditional strategy, a question still remained: how deep was the PID strategy explored and optimized so that it was comparable to the more advanced techniques? (ROTSTEIN; LEWIN, 1992).

The characteristics of self-tuning and adaptive control techniques were then further studied by Rotstein and Lewin (1992). They compared the performance of an adaptive control strategy applied to an unstable batch reactor against a PID controller based on Internal Model Control (IMC) for tuning purposes. When open-loop unstable processes are to be controlled, the PID strategy can work well if a deep knowledge of process behavior is available so that a proper tuning can be performed. For those kinds of processes, the adaptive strategy can, at first impression, seems easier. However, if the adaptive scheme's model is not properly selected, for example by modeling a lower order process then it is, the control strategy is prone to failure. The researchers propose then a set of rules for PI and PID controllers tuning so that they can be put at the same comparison base of the alternative strategy. Moreover, they study the challenges and risks of upgrading a traditional PID to a self-tuning approach and discuss the criteria to upgrade a simple control strategy.

The dynamic characteristic of semi-batch reactors temperature during the reactant feed was studied by Defaye *et al.* (1993). The authors demonstrated that an adaptative-predictive technique was able to handle these inherent temperatures changes during reactant addition.

2.4.3 Middle development – Model based control

In the same period, in the late 1970s and early 80s, researchers started to realize the opportunities and benefits of using process models to predict or at least understand the dynamic behavior to design better control strategies.

Richalet *et al.* (1978) introduced a so-called Model Predictive Heuristic Control (MPHC) where an internal model built from the controlled variables represented by its impulse-responses is used for on-line prediction.

Independently, Culter and Ramaker (1980) showed a successful technique that had been used for years at Shell Oil Company. The Dynamic Matrix Control (DMC) algorithm represents the process dynamics with a set of numerical coefficients based on a model.

Garcia and Morari (1982) defined a so-called Internal Model Control (IMC) where process disturbances can be anticipated, and corrective action taken before more severe problems occur. The control adjustments are directly related to the model parameters.

After these developments, several studies popped out regarding the use of process models embedded to process control strategies. The MPCs (model predictive control), based on the works of Richalet *et al.* (1978) and Cutler and Ramaker (1980), have been widely used since the middle 1980s as can be seen in a very good MPC historical review from Lee (2011).

However, in the middle 1980s, the researches were struggling when non-linear and dynamic processes were in place and then the previous MPC strategies developed mostly for big continuous processes (i.e. Petrochemical industry) didn´t give the desired results.

Lee and Sullivan (1988) presented a Generic Model Control (GMC) as the first application of model-based control taking into account the nonlinear behavior present in many chemical processes. The main advantage of this control structure is the fact that no linearization is needed to embed a nonlinear control into the controller.

Based on the control framework of Lee and Sullivan (1988), Cott and Macchietto (1989) developed a controller using the GMC to control the temperature of exothermic batch reactors. They used a deterministic on-line estimator to determine the amount and rate of the reaction heat released and they were also able to properly combine the nonlinear feedforward and feedback effects. In the end, the model-based control strategy showed a very good performance in the heat-up and temperature control steps and robustness regarding process parameters changes and model mismatch.

Peterson *et al.* (1989) proposed a nonlinear MPC to control a semi-batch methyl methacrylate polymerization reactor by an extended DMC algorithm and achieved a good performance applied to a MIMO (multiple input multiple output) control. The key feature of their method is the introduction of the effects of nonlinearities in the prediction horizon through a disturbance vector which updates the DMC model.

2.4.4 Further development

According to Soroush and Kravaris (1993a), when thinking about a batch reactor optimization, generally, the focus should be on maximizing its productivity and quality, rather than minimizing its operational or raw-material costs. This occurs because batch processes are usually not large scale (if compared to continuous processes) and often produce great added-value chemicals. The authors propose a systematic approach for optimal design and operation of batch and semi-batch reactors. The method not only introduces some key terms of interest in batch reactor operation but establishes some steps to support automatization and proper design. Due to the important role that batch processes play in the production of chemicals (generally related to great added value) and also due to the competitive industrial environment, their efficient design and operation are crucial.

In a second work, Soroush and Kravaris (1993b) applied their proposed method on a polymerization batch reactor. Through the optimum design (aspects as flexibility, controllability, and safety are taken into account) and control strategy (a nonlinear temperature control is selected), they can maximize conversion and specify the final product properties.

As mentioned before, the dynamic nature of batch and semi-batch processes can lead to non-constant physical properties with the time and hence good temperature control is hard to achieve. Chylla and Haase (1993) investigated this characteristic in a generalized industrial case study. They evaluated how should the nonlinearities, features, and constraints of the process be addressed to the control problem.

Chylla and Haase (1993) also highlighted the deficiencies of the standard PID controller applied to the investigated case. The control strategy was defined as a cascade control where the reactor temperature is the primary controller and the reactor jacket temperature is the secondary controller actuating in a split range control. Some of the deficiencies were summarized as the following:

- PID tuning parameters were set regardless of the weather conditions or products used. This entails control poor performance;
- The experience level of operators causes much variation in the regulation of temperature set point;

Friedrich and Perne (1995), from Bayer AG, shared several interesting examples of industrial modeling, simulation, and control techniques applied to batch reactors. It was evaluated not only the financial benefits of using dynamic simulation and advanced control methods but also the effort needed to develop and implement these kinds of techniques regarding complexity and process knowledge.

Ruppen, Bonvin and Rippin (1997) presented the problem of a very sensitive batch reactor to raw-material composition due to the presence of undesired side reactions. As each batch can have distinct behavior, the employed strategy consists of online (at given points in time) estimation of reaction rate parameters followed by real-time optimization of the load strategy to achieve minimum batch time respecting yield and concentration constraints. The authors called this strategy as Estimation-Optimization Task (EOT). The estimation and optimization are solved through a successive quadratic program (SQP) and successive linear programming (SLP), both non-linear programming (NLP) methods, selected according to the problem features.

The main challenge regarding the estimation step is the difficulty of measuring online concentrations. Although some applications of model-based measurements are used to tackle this issue, Ruppen, Bonvin and Rippin (1997) use off-line analytical data of reactor samples to provide the complementary information to the estimation step. Finally, the proposed strategy is tested in a laboratory reactor and the results are satisfactory showing that the on-line optimization could significantly improve the batch performance.

Louleh and Cabassud (1999) developed a new control strategy for a jacketed batch reactor with different heating and cooling fluids. Through a cascaded model control based on the thermal flux as the manipulated variable, the method was able to establish the better heating/cooling fluid pair in each step of the process. It was shown a better temperature control and energy saving.

The inherent time-varying trajectories of transient processes, very present in batch and semi-batch operations, bring several difficulties to the control system as described in several papers above. Helbig, Abel and Marquardt (2000) understand that many problems from this context are naturally an optimization problem. The flexibility and ability to handle the process and/or market constraints and disturbances contribute to operate the processes as close as possible to the optimum economical and safety point. The authors showed that this optimization problem approach is well established for big continuous processes and has been used often as a combination between model predictive controllers (MPC) and real-time optimization (RTO) schemes, however, for transient processes the optimization problem approach wasn´t clear. Therefore, they proposed the base concepts for optimization-based control of transient processes and highlighted the need of further development in this field of research.

2.4.5 Recent development

Over the years, math, statistics, computer science, and data analysis went through huge progress culminating in the advent of supercomputers and powerful tools. With a bigger computational power and more mathematical tools, the researchers could be much more creative regarding the application of these new techniques to other areas of knowledge, for instance, process control.

Therefore, from the 2000s on, it can be seen a “boom” in the number of papers published regarding batch and semi-batch reactors control. Thus, the following paragraphs section will compile the main papers regarding each main topic researched within the last two decades, 2000 to 2020.

After the development of the first applications of model-driven control frameworks to batch and semi-batch processes during the end of the 1980 decade, several more approaches were proposed to tackle the countless number of challenges coming from transient, very non-linear and constrained processes. As a result, the non-linear model predictive control (NMPC) and its variations, has shown very good results to keep the temperature at batch and semi-batch reactors under control.

Nagy and Braatz (2003) realized the lack of a comprehensive robustness analysis of the performance of the NMPCs. To overcome this issue, they proposed one of the first NMPC that explicitly took the process uncertainty into account in the controller design and in the proposed extended Kalman filter to reduce biases in the state estimates.

During the development of new model-based control frameworks, the researches have faced many issues such as accuracy of the model, stability of control loop, robustness of the MPC´s frameworks, real processes' uncertainties, constraint violations, and high computational effort. In this sense, several more studies have been published to tackle these issues (ANILKUMAR; PADHIYAR; MOUDGALYA, 2017; BRADFORD *et al.*, 2019; HOLTORF; MITSOS; BIEGLER, 2019; LUCIA; PAULEN; ENGELL, 2014; THANGAVEL *et al.*, 2018).

On the other hand, several researchers started to investigate how data-driven techniques can be combined with model-driven architectures to handle the model accuracy problem of model driven architecture. This fact is a real concern since the daily-life of industrial plants can be sometimes "tricky". For instance, the reactant's quality/purity not as expected, reactant's amount or heat/cooling supply can be out of expected trajectory due to equipment/instrument problems, strong weather conditions changes, too much operational intervention, operational error, catalyst activity, etc.

In this sense, Wakabayashi *et al.* (2009) investigated the application of a fuzzy control strategy to a semi-batch reactor of nylon 6 using a phenomenological model with operational conditions from a real process. The authors compared the results of a conventional PID control versus a PI-fuzzy control strategy. The advantage of the fuzzy control is that it doesn't require an internal mathematical model and therefore with personal expertise, it is possible to build a very robust control strategy. The PI-fuzzy strategy was shown to have great potential to deal with the nonlinearity and interactive aspect of the process.

Several more studies regarding the application of data-driven techniques were developed to tackle this problem. Artificial Neural Networks (ANN) and Neuro-Fuzzy Networks solely or in combination with MPC´s and GMC´s, were used in semi-batch reactors control and/or optimization (DOVŽAN; ŠKRJANC, 2010), (FONSECA *et al.*, 2016), (KAMESH; RANI, 2017), (DAOSUD *et al.*, 2019).

Regarding product quality it is very usual in batch and semi-batch processes that the end of the batch can only be established after samples results and hence, depending on the velocity of the quality control laboratory, the cycle time of the batch can increase. Moreover, for some processes, the results can only be seen several steps after the first reaction therefore it is very important to guarantee the quality. For sure, there are several advances in on-line analytics, but it isn't widely implemented in chemicals plants yet. Therefore, the use of data-driven control techniques within a single batch, as mentioned before, maybe not as reliable as expected in some applications.

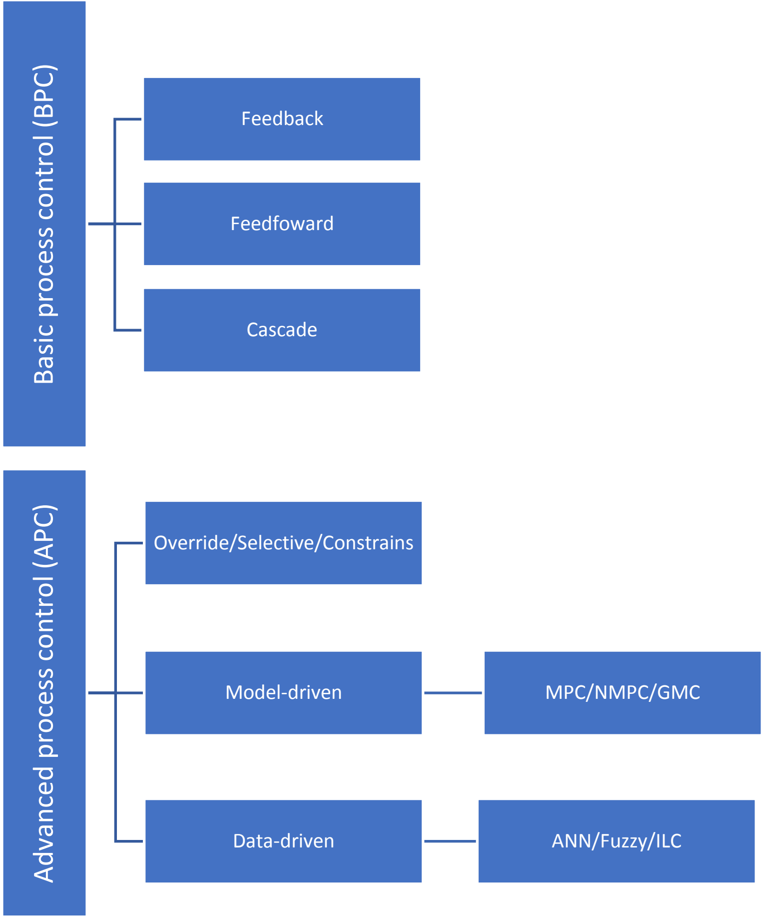
In that manner, researches figured out the information of the previous batches could be applied as a feedback control to the actual batch resulting in a called batch-to-batch (or run-to-run) control approach (XIONG; ZHANG, 2003). Therefore, the introduction of Interactive Learning Control (ILC) to batch and semi-batch processes were developed and several papers were published (MEZGHANI *et al.*, 2001), (LEE; LEE, 2003), (XIONG *et al.*, 2005), (BO *et al.*, 2017; LI; YU, 2020; WANG *et al.*, 2014)

Xiong, Dong and Zhang (2009), presented an iterative learning control approach in combination with a control affine feed-forward neural network CAFNN). A simulated isothermal semi-batch reactor was used to illustrate this application. The authors demonstrated how the CAFNN can help the ILC to improve the end-point product qualities by updating the control policy from previous batch control profiles. In the end, it was shown that the proposed method can gradually increase the endpoint product quality from batch to batch.

Besides the control techniques aforementioned, simpler control frameworks are still in use and have good results to keep batch and semi-batch reactors temperature under control. Jogwar (2015) proposed a novel control system for batch processes consisting of the use of a process modeling applied to a cascade control strategy. The work is situated in the context of energy integration of batch processes and thus, effective control enables not only better operation but also efficient energy savings. The success of using a cascade strategy was based on the fact that the process had different time scales. The model-based controller uses an input-output model, based on the interaction between the batches, and corresponds to a slow dynamic. The fast-dynamic scale is related to the batch itself. In that manner, the work proves to control the parameters of individual batches and integrate energy between batches. It is demonstrated that the proposed approach is effective against disturbances, different operational points and can integrate different batches energetically promoting an efficient overall operation.

As discussed in detail above, over the years there was a huge development of process control strategies of semi-batch reactors. The process control strategy strongly depends on process characteristics (e.g. batch or continuous, non-linearities, etc). An overview of these main strategies which can be used solely or in combination with each other is presented in Figure 1.

Figure 1 – Main process control strategies applied to semi-batch reactors



Source: Author

2.5 Dynamic simulation and modeling applied to semi-batch processes

Process simulation can bring several benefits for chemical plants whether used at plant conceptual and economic feasibility design or optimization of existing plants.

To perform a process simulation, a mathematical model is needed. The mathematical models can be classified as following (GARCIA, 2005):

- Static x Dynamic;
- Linear x Non-Linear;
- Lumped x Distributed;
- Deterministic x Stochastic.

And the way to obtain a mathematical model can be divided into (GARCIA, 2005):

- Theoretical or Phenomenological;
- Empirical.

Moreover, another usual classification of models is related to how many physical laws are included in the models (ARENDT *et al.*, 2018). This classification is shown below:

- White-box: entirely physics-based model;
- Black-box: entirely data-driven model;
- Gray-box: a mixture of white-box and black-box approaches.

In semi-batch processes, it is usual that the kinetics information is missing or is not updated due to process/technology changes. Nevertheless, it is possible to derive the missing parameters applying a tendency model as proposed by Filippi *et al* (1986). They developed a gray-box model combining the mass and energy balances (white-box approach) and experimental kinetics data (black-box approach) by matching the model prediction to experimental previous batches data. Through this method Filippi *et al* (1986) demonstrated that the simplified kinetic model proposed was enough to represent the overall kinetic behavior.

Another way to overcome missing parameter or strong non-linearities hard to model is the use of neural networks. Xiong and Jutan (2002) used a neural network to compensate model mismatches and achieved a good grey-box model proposal to control an exothermic batch reactor.

Therefore, the first step of this work was to create a model based on mass and energy balances which can properly represent the actual process.

3 METHODOLOGY

A mathematical model was proposed to describe an industrial hydrogenation reactor from a multinational chemical company in São Paulo, Brazil. Using the proposed model, computational simulations were performed and the results compared to real plant data in order to validate the model. After the model validation, an open-loop model sensitivity analysis was proposed to study the dynamics of the process and four different temperature control strategies were proposed and compared.

Due to information protection reasons, specific data regarding the process was presented in generic forms. Besides, the model variables were defined dimensionless as shown in equation (1):

$$x = \frac{x^*}{x_{ref}} \ [dimensionless] \tag{1}$$

Where

a) x is the dimensionless variable value;
b) x^* is the real variable value;
c) x_{ref} is the reference variable value;

3.1 Case explanation

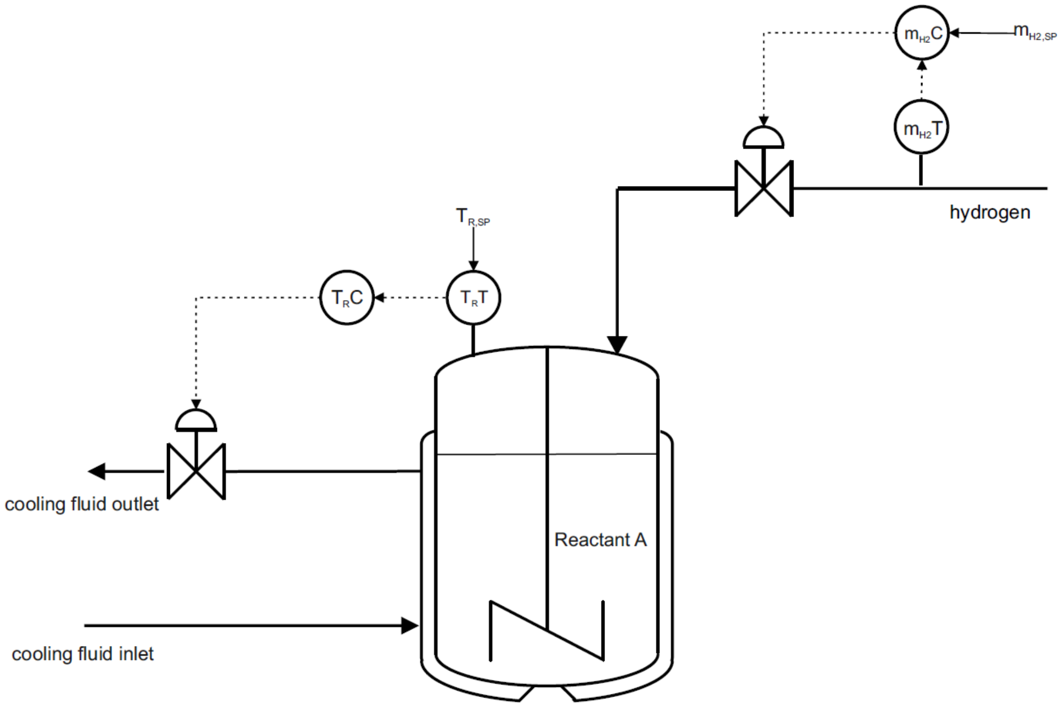
The reaction which takes place in the jacketed SBR (semi-batch reactor) is shown below:

$$A + H_2 \rightarrow B + C \tag{2}$$

A scheme of the process under study is presented in Figure 2. The first step is to dose the whole amount of A into the reactor. With reactant A inside the reactor, hydrogen is fed, and the temperature rises until a specific set-point, $T_{R,SP}$ at which the reaction is conduced. The compounds B and C are produced by the reaction of hydrogen (H_2) with reactant A which is already in the reactor. The reaction is considered completed when the reactor pressure equalizes with the hydrogen feed pressure, indicating there is no more consumption of hydrogen.

The reactor has an external jacket to remove the heat generated by the exothermic reaction.

Figure 2 – Current process scheme



Source: Author

3.2 Control problem

As mentioned in section 1.1, the goal of this work is to stabilize and optimize a real industrial hydrogenation reactor regarding temperature control and batch time which are narrowly dependent to the process cooling capacity and the constraints at the hydrogen flow.

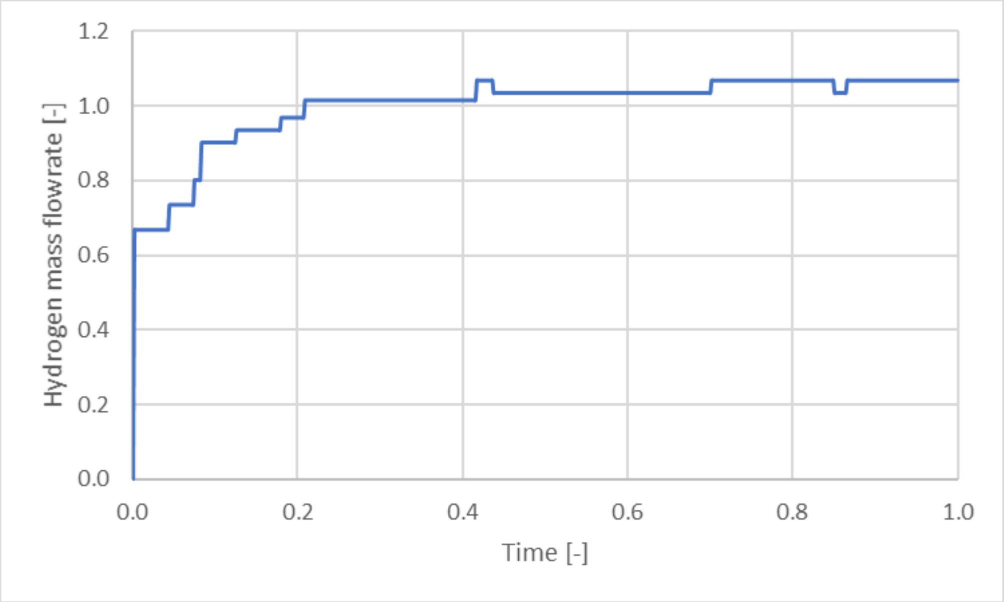
As in many batch or semi-batch reactions, regarding control, the process can be divided into three parts:

a) Heat-up step from lower temperature until reaction temperature;
b) Maintain temperature during the reaction path;
c) Post-reaction (heating/cooling/distillation/etc) before transfer;

This inherent dynamic behavior shows a moving trajectory of the temperature that can often cause variations of physical properties, representing non-linearities to be handled by the controller. Moreover, in the case of semi-batch processes, the dynamic trajectory of the feed of one of the reactants can also bring a quite challenge if the heat exchange area changes during the batch.

As can be seen in Figure 2, the case study reactor has two independent controllers. The hydrogen flowrate feed is controlled by a PI feedback controller which actuates directly on the hydrogen feed control valve keeping the hydrogen flowrate at its setpoint, $\dot{\mathrm{m}}_{\mathrm{H_{2},e,SP}}$. The reactor temperature is controlled at its setpoint, $T_{R,SP}$, by the direct actuation of a PI feedback controller at the cooling fluid flowrate control valve. Although the controllers are independent, the reactor temperature controller is influenced by the hydrogen flowrate control since the reaction rate, and thus, the energy released, depends on the hydrogen flowrate. As a consequence, in the industrial plant, operators often need to manually intervene and change the hydrogen flowrate set point as can be seen in Figure 3. Each step in the hydrogen mass flowrate represents a manual intervention.

Figure 3 – Real plant data: hydrogen mass flowrate set point manual changes during a real batch

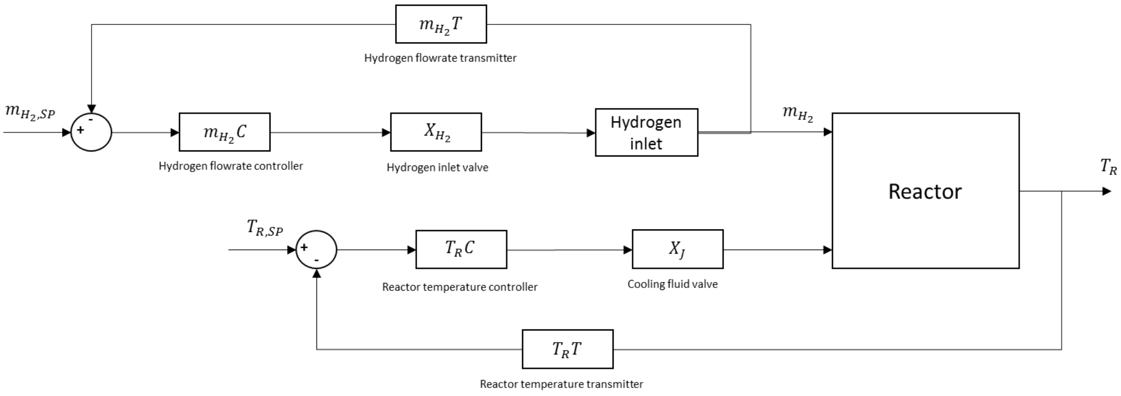


Source: Author

In the real plant, both control strategies and their parameters are the same during the heat-up and reaction phases of the batch. As the post-reaction phase can be satisfactorily conduced with the current control strategy, its control strategy will be not part of this work.

Figure 4 show the current controllers' block diagram.

Figure 4 - Current control block diagram



Source: Author

Moreover, Table 2 describes the current controllers' strategies and their constitutive equations:

Table 2 - Actual controllers´ constitutive equations

Controller	MV	CV	Controllers outputs	Equation
Hydrogen flowrate	X_{H_2}	$\dot{m}_{H_{2,e}}$	$Out_{H_2} = bias_{H_2} + Kc_{H_2}\left(\dot{m}_{H_{2,e}} - \dot{m}_{H_{2,e},SP}\right) + \frac{Kc_{H_2}}{\tau_{I,H_2}}\int\left(\dot{m}_{H_{2,e}} - \dot{m}_{H_{2,e},SP}\right)dt$	(3)
Reactor temperature	X_J	T_R	$Out_{T_R} = bias_{T_R} + Kc_{T_R}\left(\beta_{TR}.T_{R,SP} - T_R\right) + \frac{Kc_{T_R}}{\tau_{I,T_R}}\int\left(T_{R,SP} - T_R\right)dt$	(4)

Source: Author

Where:

a) MV is the manipulated variable;
b) CV is the controlled variable;
c) Out is the controller output (%);
d) $bias$ is the reference value for controller output (%);
e) K_C is the controller gain;
f) β is the reactor temperature setpoint weight;
g) $\dot{\mathrm{m}}_{\mathrm{H}_{2,\mathrm{e}}}$ is the hydrogen mass flowrate;
h) $\dot{\mathrm{m}}_{\mathrm{H}_{2,\mathrm{e}},\mathrm{SP}}$ is the hydrogen mass flowrate set point;
i) T_R is the reactor temperature;
j) $\mathrm{T}_{\mathrm{R,SP}}$ is the reactor temperature set point;

k) τ_I is the controller integral time (s);
l) X_{H_2} is the hydrogen flow control valve opening (%);
m) X_J is the cooling fluid flow control valve opening (%).

The values of the PI parameters for the current feedback controllers can be found in Table 3.

Table 3 - Feedback controllers parameters

Control parameters	Values
Kc_{H_2}	0.1
τ_{I,H_2}	6
Kc_{T_R}	-90
τ_{I,T_R}	1500
β_{TR}	0.8

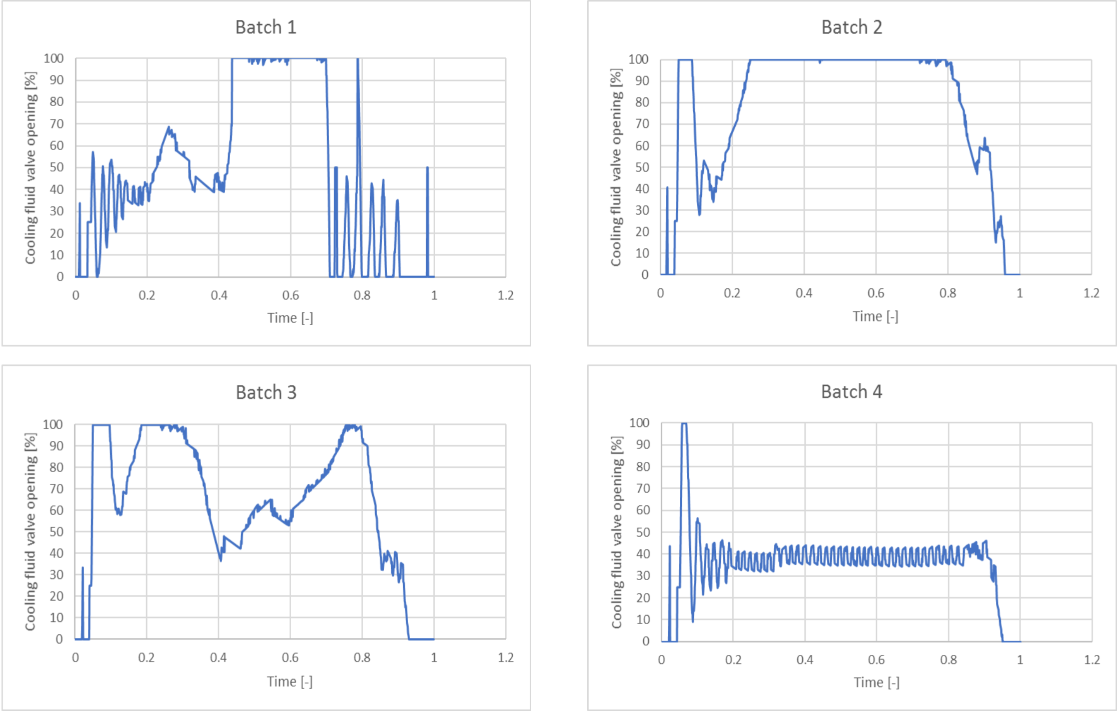
Source: Author

The main challenges of the actual process control strategies are:

a) During the first kilograms of hydrogen dosage the reactor temperature needs to be heat-up until $T_{R,SP}$ with minimum overshoot;
b) The process is sensitive to hydrogen flow rate. A high flowrate can increase very fast the reaction rate and hence, the reactor temperature can get out of control. On the other hand, a too low hydrogen flowrate can decrease temperature and increase reaction time;
c) The quality of the product is sensitive to the reactor temperature which must be controlled at $0.995.T_{R,SP} < T_{R,SP} < 1.005.T_{R,SP}$ during the reaction path. This narrow range introduces a challenge to the controller.
d) The cooling fluid network flowrate to the reactor jacket is not stable, as can be seen in Figure 5, by comparing the behavior of the cooling fluid control valve within 4 different batches. This variation affects directly the energy balance of the system and contributes

to the difficulty of keeping the hydrogen flowrate constant and demands the operator to manually intervene and change the hydrogen flowrate set point as previously shown in Figure 3.

Figure 5 – Real plant data: Cooling fluid control valve behavior in different batches


Batch 1

Batch 2

Batch 3

Batch 4

Source: Author

3.3 Model Development

The following assumptions were made:

i. The SBR is ideal: perfect and ideal mixing;
ii. The reaction takes place only in the liquid phase;
iii. Liquid phase is considered a perfect mixing (lumped model);
iv. Liquid phase behaves as a Newtonian fluid;
v. Viscosity, density and heat capacity of liquid phase are constant during the whole batch;
vi. Molecular interactions for computation of mixture properties can be neglected:

  - The total volume of the mixture is the sum of the volume of pure species;
  - The heat capacity of the liquid phase is the mass-weighted average of heat capacities of individual components;

vii. Reactor headspace (vapor phase) contains only hydrogen;

viii. Reactor headspace temperature is the same as the liquid phase temperature;

ix. The reactor jacket is always full;

x. Cooling fluid behaves as a Newtonian fluid;

xi. The heat transfer area change due to hydrogen feed is insignificant since the total amount of hydrogen is too small when compared to the total mass of reactor;

xii. The heat transfer area change due to density variations is neglected;

xiii. Heat transfer coefficient is constant during the batch;

xiv. The cooling fluid in jacket is considered a perfect mixture (lumped model);

xv. The reactor is perfectly insulated (there is no heat loss to the environment);

xvi. Hydrogen mass is negligible at mass and energy balances since its energy contribution is much smaller than reaction enthalpy;

xvii. Reactant A is 100% pure;

xviii. There is no side reaction;

xix. Mass transfer phenomena of hydrogen in liquid phase and catalyst is neglected;

3.3.1 Mass balance

With the assumptions made, the mass balance can be written as follows:

- Reaction rate, $(-r_A)$:

$$(-r_A) = f(\, C_A, P_R, x_{kat}) \tag{5}$$

Where:

  - C_A is the concentration of reactant A;
  - P_R is the reactor pressure;
  - x_{kat} is the mass fraction of the catalyst;

- Product B:

$$\frac{1}{V_L}\frac{dN_B}{dt} = (-r_A);\ [mol/L.h] \qquad N_{B0} = N_B(t=0) \tag{6}$$

Where:

  - N_B is the number of mols of product B
  - V_L is the liquid phase volume

- Product C:

$$\frac{1}{V_L}\frac{dN_C}{dt} = (-r_A);\ [mol/L.h] \qquad N_{C0} = N_C(t=0) \tag{7}$$

Where:

  - N_C is the number of mols of product C

- Reactant A:

$$-\frac{1}{V_L}\frac{dN_A}{dt} = (-r_A); [mol/L.h] \qquad N_{A0} = N_A(t=0) \tag{8}$$

Where:

  - N_A is the number of mols of reactant A

- Hydrogen:

$$\frac{dN_{H_2}}{dt} = \dot{N}_{H_{2,e}} - \dot{N}_{H_{2,cons}};\ N_{H_20} = N_{H_2}(t=0)\ [mol/h] \tag{9}$$

$$\dot{N}_{H_{2,e}} = \frac{\dot{m}_{H_{2,e}}}{MM_{H_2}}; [mol/h] \tag{10}$$

$$\dot{N}_{H_{2,cons}} = (-r_A).V_L; [mol/h] \tag{11}$$

Where:

- N_{H_2} is the number of mols of hydrogen;
- $\dot{N}_{H_{2,e}}$ is the inlet hydrogen molar flowrate;
- $\dot{N}_{H_{2,cons}}$ is the consumed hydrogen molar flowrate;
- $\dot{m}_{H_{2,e}}$ is the inlet hydrogen mass flowrate;
- MM_{H_2} is the hydrogen molecular mass;

Considering assumptions vii and viii, the number of mols of hydrogen contained in the reactor headspace can be expressed by the following:

$$N_{H_2} = \frac{P_R.V_{hs}}{R.T_R.Z}; \tag{12}$$

Where:

- P_R is the reactor pressure;
- V_{hs} is the reactor head space free volume;
- R is the universal gas constant;
- T_R is the reactor temperature;
- Z is the compressibility factor hydrogen

The variation of the number of mols of hydrogen regarding reactor pressure can be written as:

$$\frac{dN_{H_2}}{dP_R} = \frac{V_{hs}}{R.T_R.Z}; [mol/bar] \tag{13}$$

Applying equations (10), (11) and (13) on equation (9) follows:

$$\frac{V_{hs}}{R.T_R.Z}.\frac{dP_R}{dt} = \frac{\dot{m}_{H_{2,e}}}{MM_{H_2}} - (-r_A).V_L; [mol/h] \tag{14}$$

Finally, it is possible to obtain a time dependent equation for hydrogen pressure:

$$\frac{dP_R}{dt} = \frac{R.T_R.Z}{V_{hs}}.\left[\frac{\dot{m}_{H_{2,e}}}{MM_{H_2}} - (-r_A).V_L\right]; [bar/h] \tag{15}$$

3.4 Energy balance

By assumptions xiv, is was not considered in the model a spatial mean temperature and hence, the reactor temperature can be expressed by the following:

$$m_R Cp_R \frac{dT_R}{dt} = \dot{m}_{H_{2,e}} Cp_{H_{2,e}} T_{H_{2,e}} + (-r_A) \mathrm{V_L} \Delta H_R - \ (\overline{UA}) \Delta T \tag{16}$$

Where:

- m_R is the reactor total mass;
- Cp_R is the reactor content heat capacity;
- $Cp_{H_{2,e}}$ is the inlet hydrogen heat capacity
- $T_{H_{2,e}}$ is the inlet hydrogen temperature;
- ΔH_R is the reaction enthalpy;
- $(\overline{UA})$ is the average global heat transfer coefficient times area;
- ΔT is the reactor temperature difference;

The reactor mass can be expressed as the sum of the individual mass of each reactant, product and solvent.

$$m_R = m_A + m_{H_2} + m_B + m_C + m_{sol} \tag{17}$$

Where:

- m_i is the substance i mass $(i = A, B, C, H_2, Sol)$;

However, through assumptions xvi, the hydrogen mass can be neglected:

$$m_R = m_A + m_B + m_C + m_{sol} = \sum m_i \tag{18}$$

Or in terms of number of mols:

$$m_R = N_A MM_A + N_B MM_B + N_C MM_C + N_{SOL} MM_{SOL} = \sum N_i MM_i \tag{19}$$

Where:

- N_i is the substance i number of mols $(i = A, B, C, H_2, Sol)$;
- MM_i is the substance i molecular mass $(i = A, B, C, H_2, Sol)$;

By assumptions vi:

$$m_R Cp_R = m_A Cp_A + m_B Cp_B + m_C Cp_c + m_{sol} Cp_{sol} = \sum Cp_i N_i MM_i \tag{20}$$

Where:

- ○ Cp_i is the substance i heat capacity ($i = A, B, C, H_2, Sol$);

Finally, the reactor temperature is expressed as below:

$$\frac{dT_R}{dt} = \frac{Q_R - Q_J}{\sum Cp_i N_i MM_i}; T_{R_0} = T_R(t = 0)[K/h] \tag{21}$$

$$Q_R = (-r_A)\mathrm{V_L}\Delta H_R[J/h] \tag{22}$$

$$Q_J = (\overline{UA})\Delta T\ [J/h] \tag{23}$$

Where:

- ○ Q_R is the reaction heat;
- ○ Q_J is the heat exchanged trough the jacket;

As mentioned in assumptions xiv, the reactor jacket was modeled as a CSTR, so that the ∆T can be written as:

$$\Delta T = T_R - T_J\ [K] \tag{24}$$

Where:

- ○ T_J is the jacket temperature;

Jacket temperature:

$$\frac{dT_J}{dt} = \frac{F_J Cp_J \rho_J (T_{eJ} - T_J) + Q_J}{\rho_J Cp_J V_J}; T_{J_0} = T_J(t = 0)[K/h] \tag{25}$$

Where:

- ○ F_J is the cooling fluid volumetric flowrate;
- ○ Cp_J is the cooling fluid heat capacity;
- ○ ρ_J is the cooling fluid density;
- ○ T_{eJ} is the cooling fluid inlet temperature;
- ○ V_J is the jacket volume;

3.5 Constitutive equations

Volume of liquid inside the reactor as assumptions vi:

$$V_L = \frac{N_A MM_A}{\rho_A} + \frac{N_B MM_B}{\rho_B} + \frac{N_C MM_C}{\rho_C} + V_{SOL}[L] \tag{26}$$

Where:

- ρ_i is the substance i density $(i = A, B, C)$;
- V_{SOL} is the solvent volume;

Headspace volume:

$$V_{hs} = V_{TOTAL} - V_L \ [L] \tag{27}$$

Where:

- V_{TOTAL} is the reactor total volume;

Reactant A concentration:

$$C_A = \frac{N_A}{V_L}[mol/L] \tag{28}$$

Mass fraction of catalyst:

$$x_{kat} = \frac{m_{kat}}{m_{A0}}[kg/kg] \tag{29}$$

Where:

- m_{kat} is the mass of catalyst $[kg]$;
- m_{A0} is the initial mass of reactant A $[kg]$.

Initial mass of reactant A:

$$m_{A0} = m_0 . x_{A0}[kg] \tag{30}$$

Where:

- m_0 is the mass of the mixture reactant A + solvent coming from the previous vessel $[kg]$;
- x_{A0} is the mass fraction of reactant A in the mixture A + solvent coming from the previous vessel $[kg/kg]$.

Based on Emerson (2017) control valve hand book:

a) Hydrogen mass flow through control valve:

$$\dot{m}_{H_{2,e}} = 27{,}34. C_{v,H_2}. F_{p,H_2}. Y_{H_2}. f(X)_{H_2}. \sqrt{x_{eff,H_2}. P_{H_{2,e}}. \rho_{H_2 @ P_{H_{2,e}}}} \ ; [kg/h] \tag{31}$$

$$Y_{H_2} = 1 - \frac{x_{eff,H_2}}{3. F_{k,H_2} . x_{T,H_2}} \tag{32}$$

$$F_{k,H_2} = \frac{k_{H_2}}{1.4} \tag{33}$$

$$x_{T,H_2} = 0{,}84. F_{L,H_2}^2 \tag{34}$$

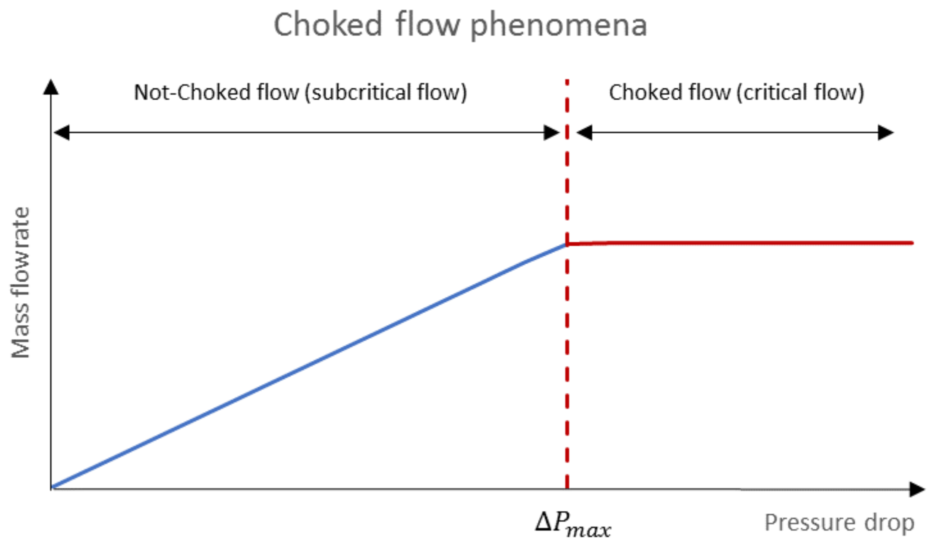
The $f(X)_{H_2}$ term represents the characteristic of the valve. For this case study, it was used a linear characteristic valve which means a direct and proportional relationship between the flow rate and the valve opening, X_{H_2}.

$$f(X)_{H_2} = X_{H_2} \tag{35}$$

For compressible fluids, it is very important to take into account the choked flow phenomenon. This condition determines a maximum pressure drop allowable where, from this point on, the mass flowrate is not more dependent on the downstream pressure but only on upstream pressure and temperature. (EMERSON, 2017)

The phenomenon is exemplified in Figure 6.

Figure 6 - Choked flow phenomenon



Source: Author

The proposed model is able to handle choked flow conditions by using the following ISA (International Society of Automation) equations below. The flow is considered sub-critical (not choked flow) if:

$$\frac{P_{H_2,e} - P_R}{P_{H_2,e}} < F_{k,H_2}.x_{T,H_2} \tag{36}$$

- For sub-critical flow (not choked flow):

$$x_{eff,H_2} = \frac{P_{H_2,e} - P_R}{P_{H_2,e}} \tag{37}$$

- For critical flow (choked flow):

$$x_{eff,H_2} = F_{k,H_2}.x_{T,H_2} \tag{38}$$

Based on Driskell equation (GARCIA, 2005):

b) Cooling fluid volumetric flow through control valve:

$$F_J = 27339.6C_{v,J}.F_{p,J}.f(X)_J.\sqrt{\frac{\Delta P_J}{\rho_J}} \ ;[L/h] \tag{39}$$

The $f(X)_J$ term represents the characteristic of the valve. For this case study, it was used a linear characteristic valve which means a direct and proportional relationship between the flow rate and the valve opening.

3.6 Model Classification

According to the model classification made by (GARCIA, 2005) and (ARENDT *et al.*, 2018) as mentioned in topic 3.1, the proposed model is classified as:

a) Theoretical: the model is based on physical laws (mass and energy balances) instead of empirical data;
b) Dynamic: variables trajectories vary with time. Transient operation and no steady-state;
c) Non-linear: as can be seen in the mass and energy balances, the non-linearities of the process comes from the reaction rate, time-dependent pressure trajectory, time-dependent reactor temperature trajectory, time-dependent cooling fluid temperature trajectory;
d) Lumped: properties and states are considered homogeneous within the volume of control. Spatial variations are neglected;

e) Deterministic: the dynamic trajectory result is always the same given a specific initial condition;
f) Gray-box approach: the model is based on mass and energy balance, however, the global heat transfer coefficient will be estimated by empirical data as described in section 3.9

3.7 Model Variables and Parameters

The variables used in the model can be divided into three main groups. The state variables, the input variables, and the output variables.

a) State variables define the state of the process in each instant of time. For the proposed model they are: $N_A, N_B, N_C, T_R, T_J, P_R, X_{H2}, X_J, V_L$;
b) Input variables define the operational conditions of the process and can be used as model disturbances. For the proposed model they are: $x_{A0}, m_{kat}, f_{kat}, T_{eJ}, P_{eJ}, P_{e,H_2}$;
c) Output variables are variables that result from the solution of model equations and which trajectories results are used to evaluate the process behavior. These variables can be either state variables or a transformation of state variables. For the proposed model they are: T_R, P_R.

The model parameters are divided into three groups as shown below:

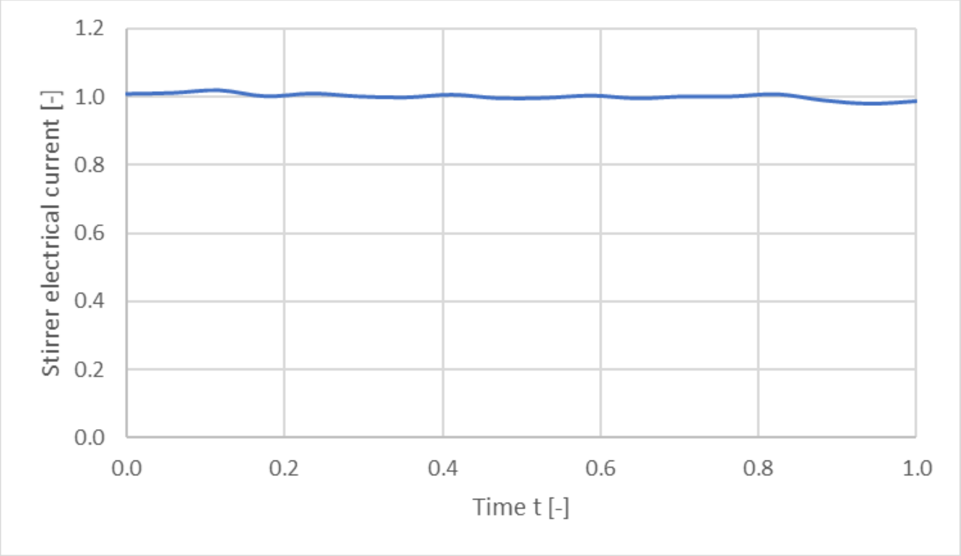
a) Initial conditions: values of the state variables at t (time) = 0;
b) Operational conditions: values of input variables;
c) Parameters: control parameters, valve parameters, physicochemical properties, and kinetic parameters

3.7.1.1 UA estimation

The global heat transfer coefficient (U) is highly dependent on viscosity and it is highly impacted by fouling (INCROPERA *et al.*, 2007).

As mentioned in assumption v, the viscosity is considered constant along the batch time. In the real plant, this fact can be indirectly observed through a constant electrical current consumption by the reactor stirrer, as can be seen below (Figure 7). If there was a significant change in viscosity, the stirrer would have to struggle more or less to keep the rotation speed at the same set point, and hence, an increase or decrease of the electrical current consumption would be seen.

Figure 7 – Real plant typical stirrer electrical current consumption along the batch time



Source: Author

Regarding fouling it was evaluated: (a) fouling at the reactor inside walls; (b) fouling at the service side (reactor jacket). Internal fouling is not expected due to components characteristics and plant historic internal inspection results. However, the service side (reactor jacket) is subjected to fouling problems that can gradually change the UA. Nevertheless, as it is a medium to long-term issue that cannot suddenly modify the reactor conditions, it will not be considered in this work.

Johnson *et al.* (2016) studied the application of a constant overall heat transfer coefficient in batch jacketed reactors, which can be inaccurate in a transient approach due to variations in reactor and jacket temperatures, physicochemical properties etc. Nevertheless, the authors remarked that it is usual to see this kind of approach in industry and academia. In this work, the overall heat transfer coefficient (U) dependency on time was neglected and the following approach was done to estimate a constant in time U.

Real jacket inlet and outlet temperatures and cooling fluid flowrate data were extracted from fifteen batches along one year of operation to support the calculation of UA coefficient. The data was fitted to the equation below.

$$\overline{Q_J^n} = \frac{\sum_0^i F_J^{i,n}\,\rho_J Cp_J\left(T_J^{i,n} - T_{eJ}^{i,n}\right)}{i} \tag{40}$$

Where:

- $\overline{Q_J^n}$ is the average heat removed by jacket in batch n;
- $F_J^{i,n}$ is the cooling fluid volumetric flowrate at time i of batch n;
- $T_J^{i,n}$ is the jacket outlet temperature at time i of batch n;
- $T_{eJ}^{i,n}$ is the jacket inlet temperature at time i of batch n;

$$\overline{\Delta T^n} = \frac{T_R^{i,n} - T_J^{i,n}}{i} \tag{41}$$

Where:

- $\overline{\Delta T^n}$ is the average ΔT in batch n;
- $T_R^{i,n}$ is the reactor temperature at time i of batch n;

From equation *(23),*

$$\overline{UA^n} = \frac{\overline{Q_J^n}}{\overline{\Delta T^n}} \tag{42}$$

Where:

- $\overline{UA^n}$ is the average UA in batch n.

Finally, the constant overall heat transfer coefficient times area, UA, used in the model was the average of the fifteen batches:

$$\overline{UA} = \frac{\sum_0^n \overline{UA^n}}{n} \tag{43}$$

The average of the fifteen batches, $\overline{UA}$, was 0.99 with a standard deviation of 0.03.

3.8 Simulation environment

The mathematical model was implemented at MATLAB as the numerical calculation tool in other to solve the differential equations.

It was used MATLAB ode15s solver which integrates the system of differential equations within a determined timespan given initial conditions. The solver is based on the numerical differentiation formulas (NDFs) of orders 1 to 5, which are related with the Gear's method (backward differentiation formulas – BDFs) and is recommended when two or more different variables have highly different dynamic variations, presenting a so-called stiff behavior (MATHWORKS®, 2019).

The tolerances for the numerical integration errors were established as the following: absolute and relative error tolerance of 1e-10.

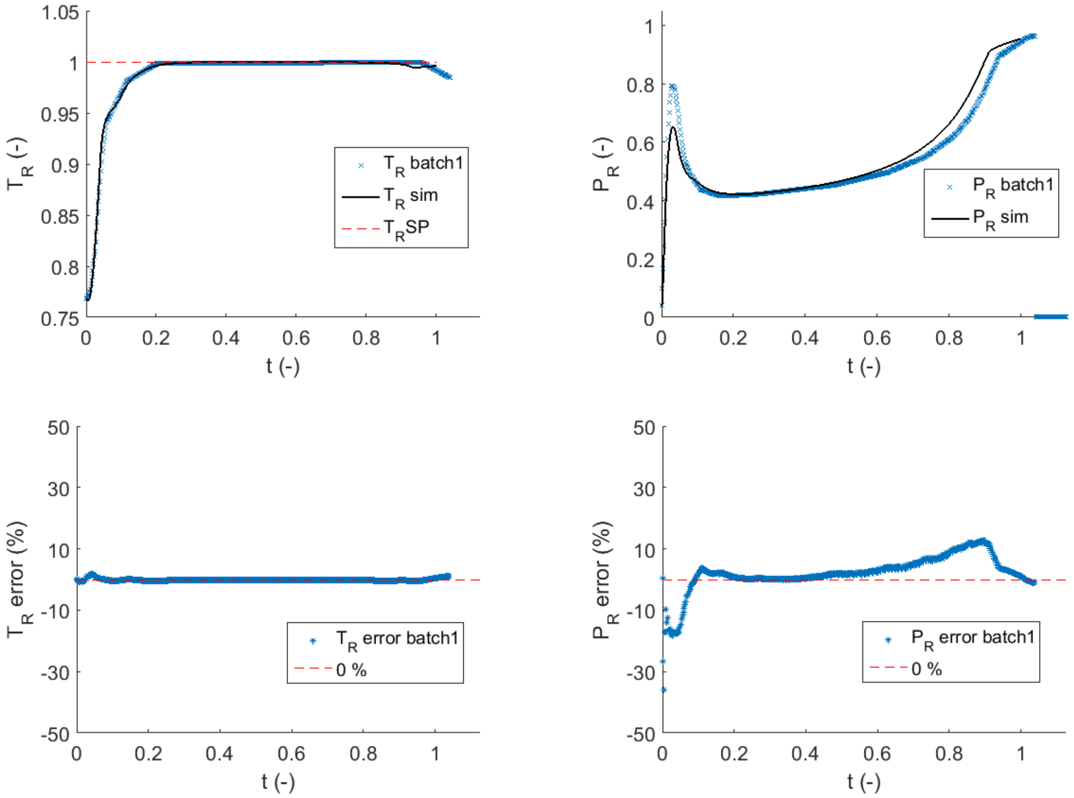
4 RESULTS AND DISCUSSIONS

4.1 Model validation

To validate the proposed model, the closed-loop simulation results were compared to real plant data from three batches. The batches were selected based on satisfactory results regarding pressure and temperature profiles, quality and batch time.

Figure 8 shows the simulations results against plant data for batch 1. As one can see, the proposed model fitted the real data. The temperature error between model and real data was below 2% during the whole batch, while the pressure error was below 10% in most of the batch time. The lower values of error for temperature are observed since it is the controlled variable of the process.

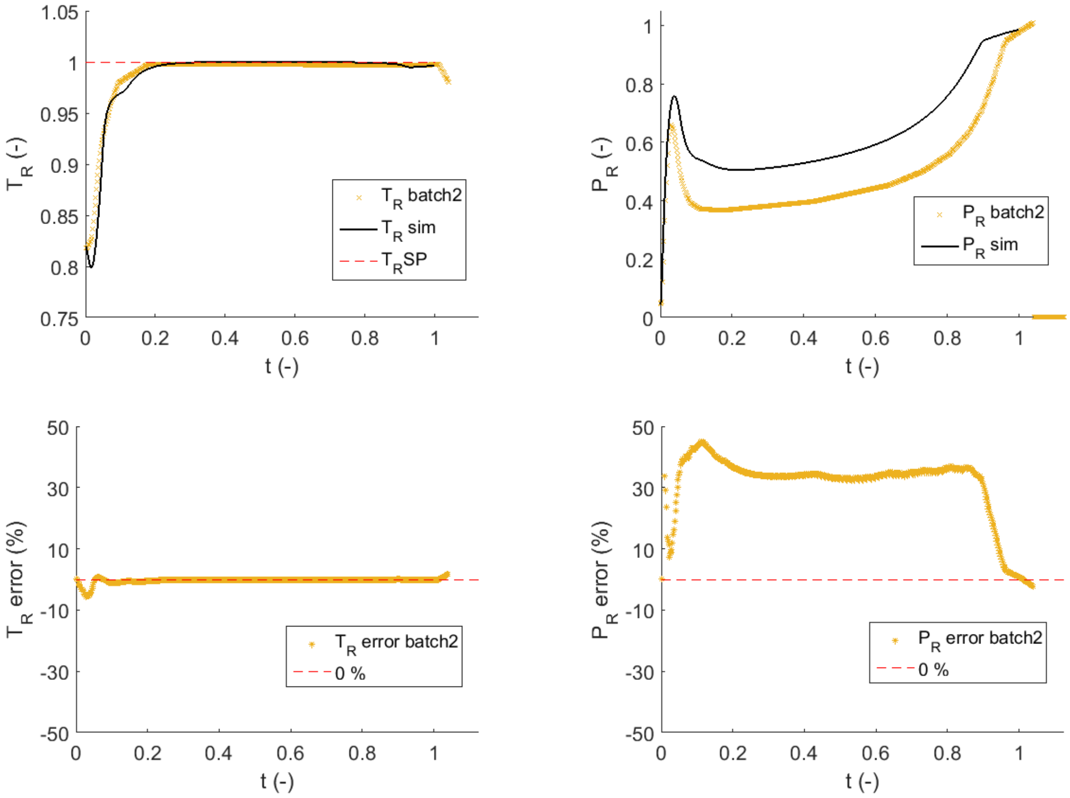
Figure 8 - Reactor temperature and pressure: a comparison between plant real data and simulation results – Batch1



Source: Author

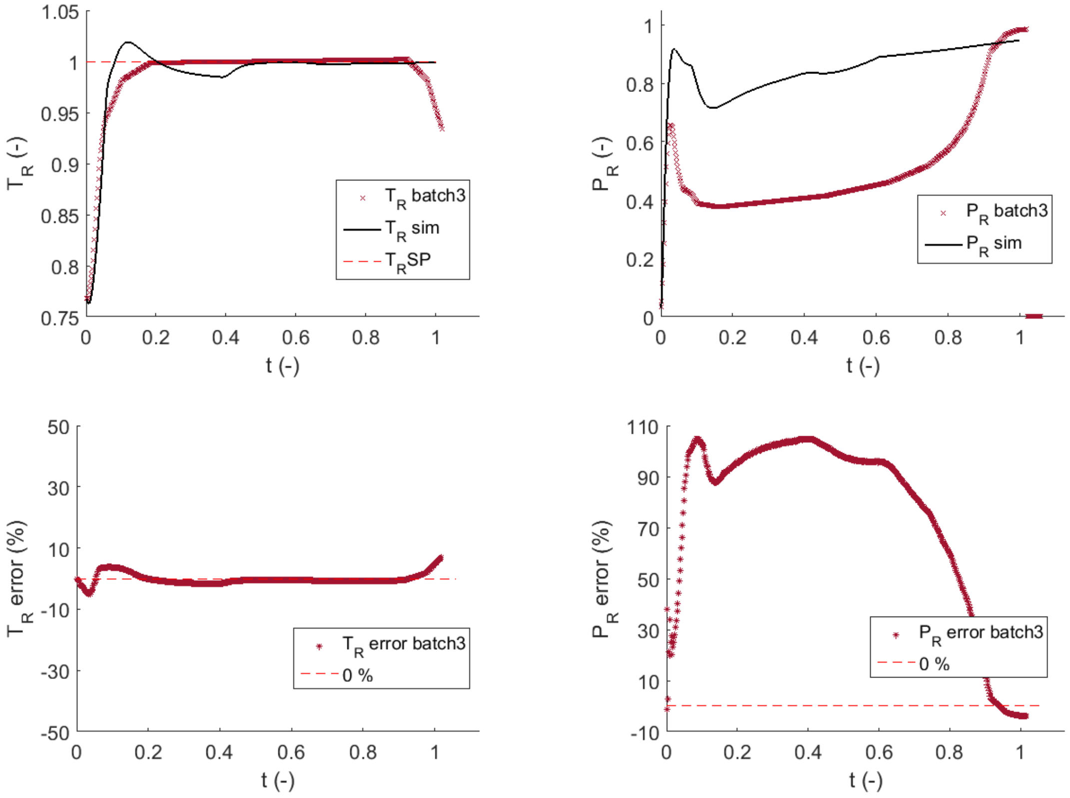
Comparing the same simulations results to different batch data (batch 2 and 3), as one can see in Figure 9 and Figure 10, a significant mismatch, especially in pressure, can be observed with errors above 30%.

Figure 9 - Reactor temperature and pressure: a comparison between plant real data and simulation results – Batch2



Source: Author

Figure 10 - Reactor temperature and pressure: a comparison between plant real data and simulation results – Batch3



Source: Author

Analyzing the differences between the real batches, it was found that the catalyst activity can significantly vary from one batch to other. These non-controlled variations affect the process behavior and are not related to the process itself, but with the catalyst supply. Therefore, to take into consideration this fact, a parameter was added in the reaction rate in the form of a kinetic factor as represented in equation (44):

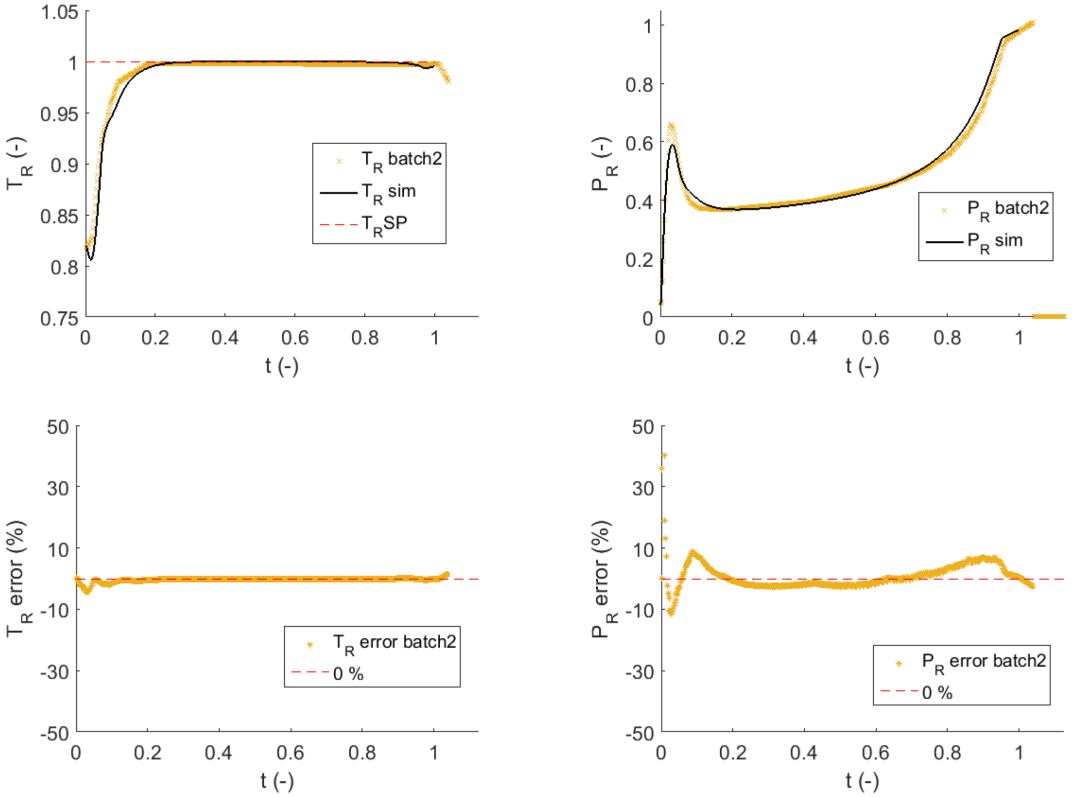
$$(-r_A) = f(f_{kat}, C_A, P_R, x_{kat}) \tag{44}$$

Where:

- f_{kat} is the kinetic factor which represents the activity of the catalyst;

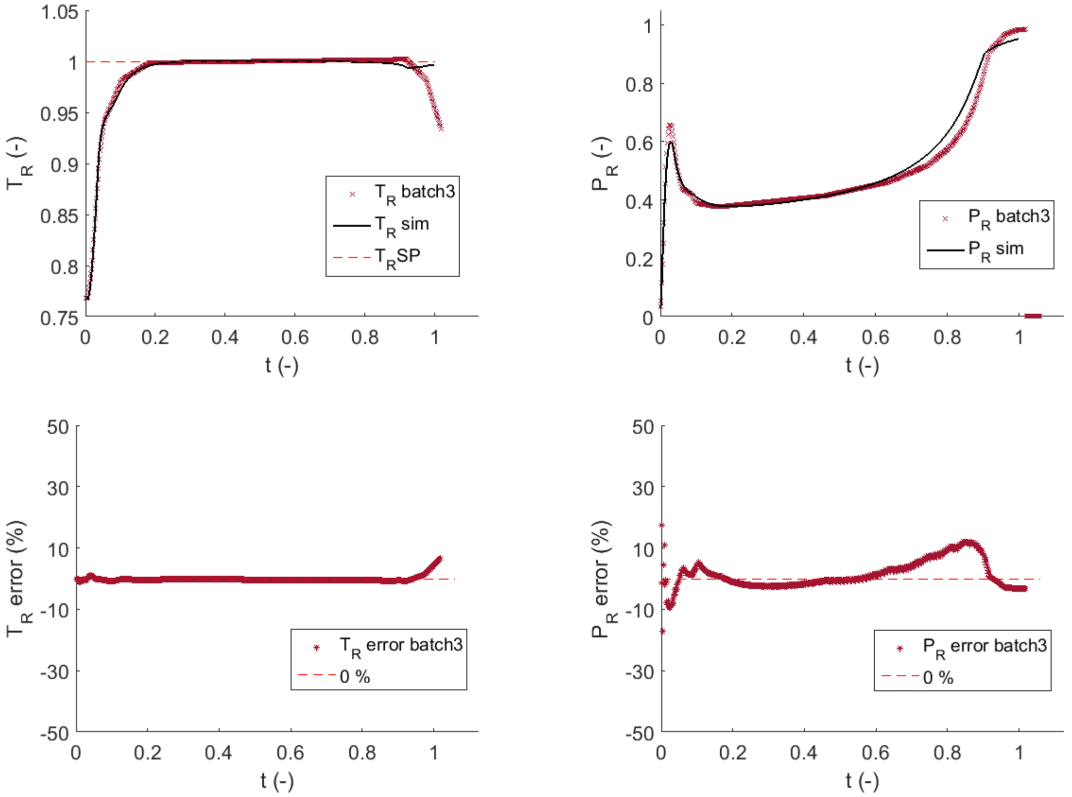
The kinetic factor was adjusted depending on the catalyst activity in each batch. Then, the simulations were performed and compared again to real data from batches 2 and 3. The simulation results showed a better fit to the real process data as can be seen in Figure 11 and Figure 12. The error was less than 5% for temperature during the whole batch, while the pressure error was below 10% in most of the batch time.

Figure 11 – Batch 2 results adjusted by kinetic factor



Source: Author

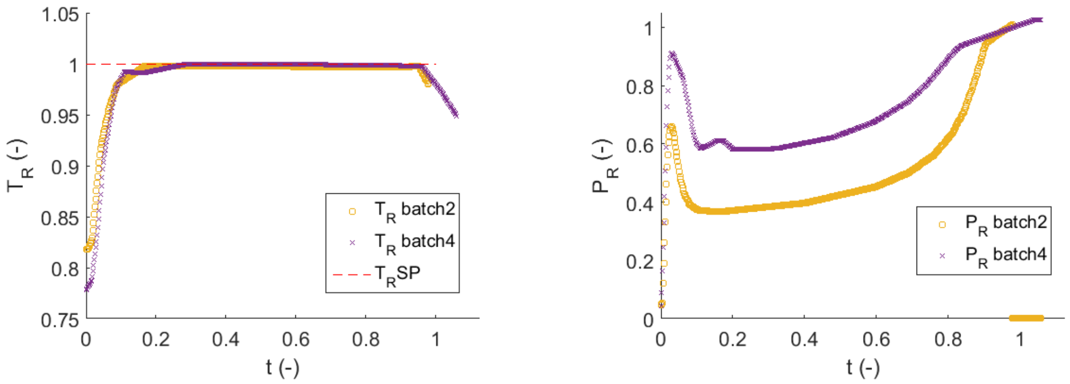
Figure 12 - Batch 3 results adjusted by kinetic factor



Source: Author

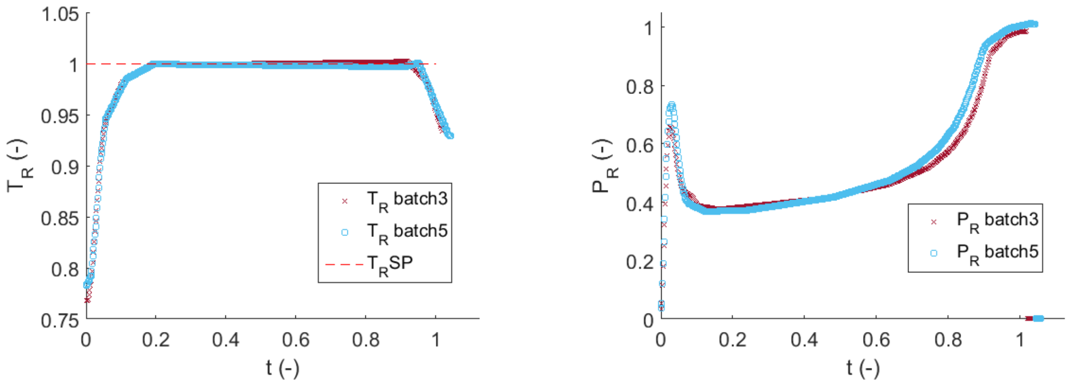
At a first sight, the consideration of the kinetic factor in the reaction rate equation allowed better description of the process real behavior and reduced mismatches between computational results and real data. Nevertheless, comparing real batches subjected to equivalent catalyst activities, the process behavior can vary significantly as can be seen in Figure 13. On the other hand, in some cases, the opposite effect occurs. Batches with different catalyst activity presented similar trajectories as can be seen in Figure 14.

Figure 13 - Batch 2 and 4: similar catalyst activity



Source: Author

Figure 14 - Batch 3 and 5: different catalyst activity



Source: Author

The phenomenon aforementioned indicates that the catalyst activity is not solely the reason for the model mismatch, but shows the existence of many uncertainties in the real process. The main uncertainties were identified and classified as the following:

- Process uncertainties:
    - Fluctuation of inlet jacket temperature and jacket cooling fluid inlet pressure related to weather conditions and other plant consumers using the same cooling fluid network;

  - Fluctuations of hydrogen inlet pressure related to weather conditions and pressure control at hydrogen tankfarm;
  - Fluctuations of hydrogen flow rate related to measurement noise;
  - Variations of the catalyst activity and performance due to different supplier, range of specification and contaminants;
  - Variation of purity of component A, which according to assumptions xvii, is considered 100% pure, but, in the real process, can be affected by downstream process oscillations;

- Parameters uncertainties:
  - Variations of UA discussed in topic 3.7.1.1. The UA was estimated as an average constant value, but in the real process, UA changes during the batch and can change significantly from each batch, as U depends on the cooling fluid flowrate, reactor and jacket temperatures;
  - Inaccuracy of kinetic parameters since they were based on laboratory research data and are subjected to errors that can occurs due to scale-up issues;

Despite of the discussed uncertainties and the observed errors between real data and simulation results, for the proposal of this dissertation, which is the comparison of temperature control techniques, the achieved model satisfactorily represents the real process.

Several techniques for uncertainty handling (LUCIA *et al.*, 2014; ROSTAMPOUR; ESFAHANI; KEVICZKY, 2015) and parameter estimation (GRAICHEN; HAGENMEYER; ZEITZ, 2006; KAMESH; RANI, 2016; KERN; SHASTRI, 2015) applied to batch and semi-batch processes have been studied to overcome these issues, however, it was not scope of this work the application of these techniques.

4.2 Model sensitivity analysis

After model validation, an open-loop model sensitivity analysis was performed. This evaluation is important to understand the process dynamics, which means, how the main variables trajectories behave after parameters disturbances and changes in the initial conditions. These results can bring clarity to determine the best control strategy and which variable to use to optimize the process. Moreover, this analysis can be used to support process troubleshooting and also to perform safety analysis.

Therefore, the batch trajectory profiles help to understand how the reactor pressure, reactor temperature, jacket temperature and reaction time (as a result of complete consume of the number of mols of A) are influenced by the following parameters and variables:

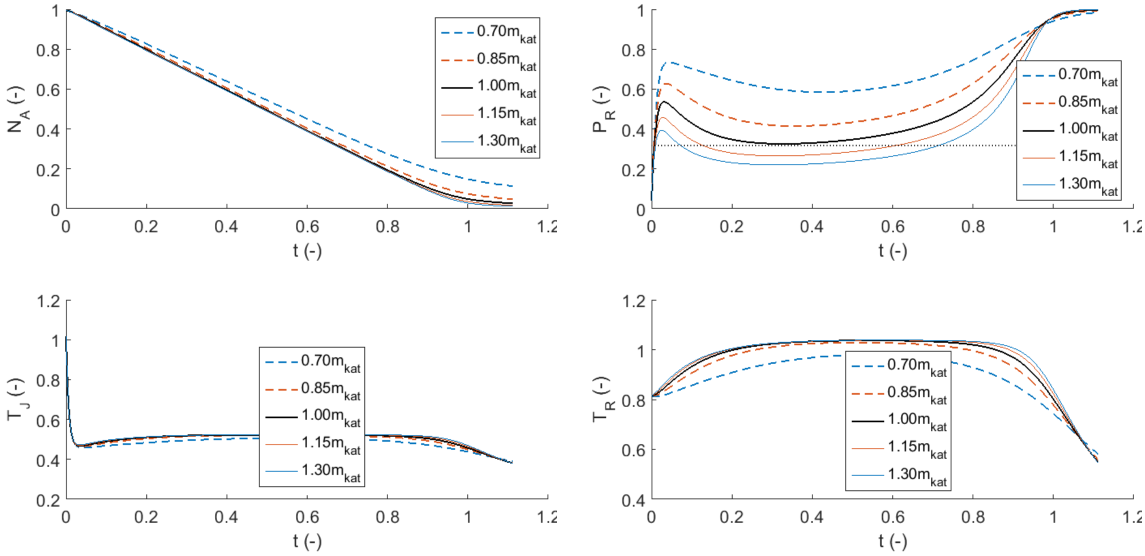
- Parameters:
  - Catalyst amount (m_{kat}) and initial mass fraction of A (x_{A0}) can be changed by plant personal. Therefore, it is important to evaluate what benefits or problems can appear due to a parameter change;
  - Kinetic factor (f_{kat}) represents the activity of the catalyst;
  - Global heat coefficient times area (UA) can change due to process conditions (i.e. reactor temperature, fouling, etc) and hence, it is important to know what impact in the process it might come up with;
- Initial conditions:
  - Initial jacket temperature, T_{J_0}, initial reactor temperature, T_{R_0}, and pressure, P_{R_0}, are different from each batch, therefore, it is fundamental to understand how these values change the dynamic of the process;
- Process conditions:
  - Cooling fluid inlet temperature, T_{eJ}, and hydrogen inlet pressure, $P_{H_{2,e}}$, have high fluctuation as explained before. Thus, it is important to evaluate how much impact these variables affect the process;

- Control final elements:
    - Cooling fluid control valve opening (X_J) and hydrogen control valve opening (X_{H_2}) are used in the actual control strategy as manipulated variables. Therefore, it is substantial to understand their behavior to establish the correct control of the process.

It is important to highlight that the model sensitivity analysis results forward in this work are true only for the variables and parameters range evaluated. For broader ranges, the effects can be different. However, the following ranges were chosen as they are closer to the real-life fluctuation of the process.

Figure 15 shows the model sensitivity against catalyst quantity.

Figure 15 – Model sensitivity: Catalyst quantity (m_{kat})



Source: Author

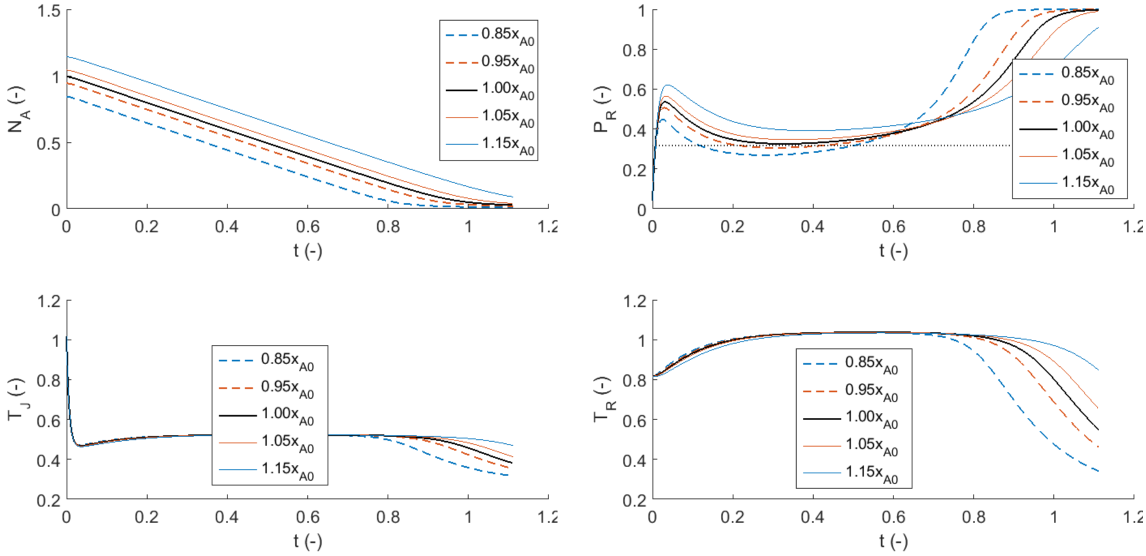
It is important to highlight the chocked flow phenomenon described by equation (36), where for a certain pressure value, the hydrogen flow rate through the valve doesn’t depend on the pressure difference but only on the valve opening. This limiting value is the black dotted line at P_R graph. For pressure values below this line, the flow is critical (choked) and for values above this line, the flow is subcritical and depends on both pressure difference and valve opening.

Thereupon, it can be seen at P_R graph, the m_{kat} directly affects the reactor pressure. A bigger amount of catalyst has the potential to increase the reaction rate, however, the reaction rate is limited by the amount of hydrogen that is entering in the reactor, which is fixed by the hydrogen valve opening due to the choked flow condition as mentioned before. This results in a pressure reduction to compensate the effect of higher catalytic activity. On the other hand, for smaller quantities of catalyst, the reaction rate is limited by the amount of catalyst and not by hydrogen flow. Therefore, there is enough hydrogen for the catalytic demand and hence, one can see a tendency in pressure increase and also in the reaction time (N_A graph). Nevertheless, it is interesting to notice at N_A graph, that for bigger amounts of catalyst the reaction time doesn´t change significantly. This result can be explained by a constant reaction rate as a result of a fixed hydrogen flow rate since the choked flow region occurs in most of the time in these cases.

The changes in the reactor temperature are explained by the same reason since the heat of reaction is a function of the reaction rate. For the catalyst amount values where the hydrogen flow is chocked, the temperature doesn’t change because in these cases the reaction rate is constant. However, for smaller amounts of catalyst, the reaction rate isn’t limited, and the temperature change is visible.

Figure 16 shows the model sensitivity against the initial mass fraction of A.

Figure 16 – Model sensitivity: Initial mass fraction of A (x_{A0})



Source: Author

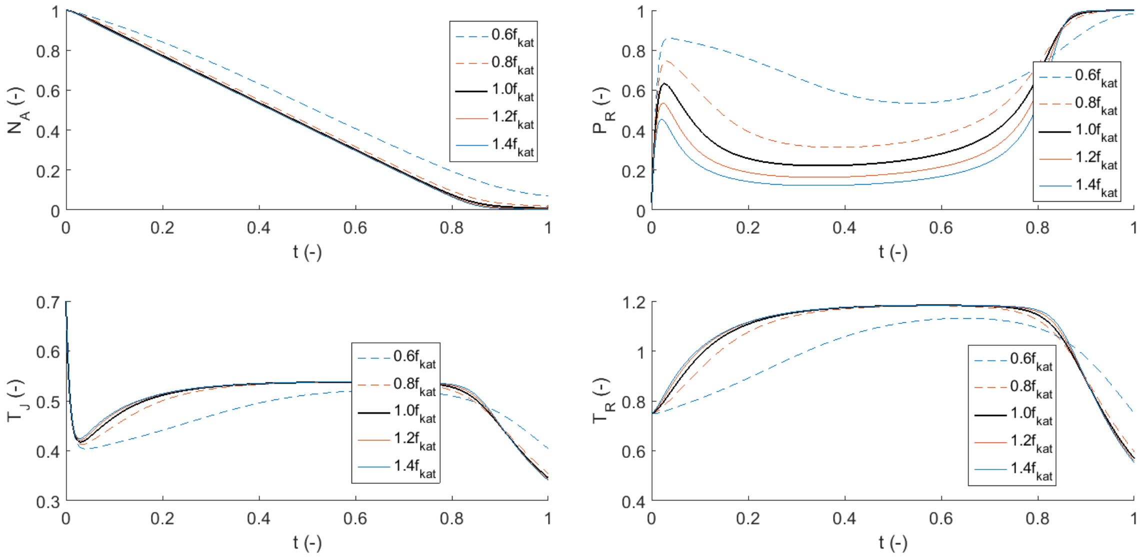
The initial mass fraction (x_{A0}) influences directly the concentration of A (C_A), which has a nearly a null effect in the reaction rate times liquid volume for the analysis range. The result of this effect can be seen through the almost parallel lines at N_A graph, showing a constant reaction rate for all cases.

As one can see at N_A graph, the x_{A0} directly affects the reaction time. A lower mass fraction means less reactant to be converted, therefore, the reaction time will be lower. The other way around is as well true. For the reactor pressure, it can be seen a strong influence of initial mass fraction of A. The initial mass fraction of A affects the proportion of catalyst and reactant A. This means the lowest initial mass fraction of A, the higher excess of catalyst will be present in the reaction and hence, as explained for Figure 15, the pressure will be lower. The pressure is shifted in time due to significant different reaction times.

As mentioned before, in the range evaluated, the reaction rate is nearly the same for all cases and hence, the reactor temperature does not change since the heat of reaction is a function of the reaction rate.

Figure 17 shows the model sensitivity against kinetic factor (catalyst activity).

Figure 17 – Model sensitivity: f_{kat}

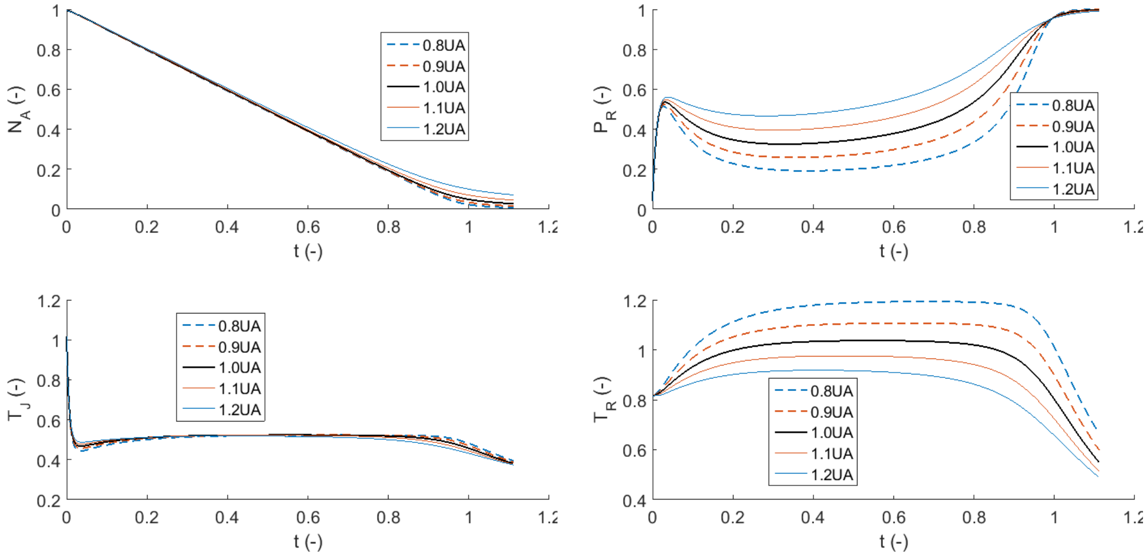


Source: Author

It is important to highlight that the catalyst activity impact is not fully comprehended, however, some insights can be derived from Figure 17. The kinetic factor was modeled as a multiplier of the reaction rate, therefore, bigger values mean a bigger reaction rate and as a consequence less accumulation of hydrogen and lower pressure level. The consumption of reactant A is limited by the amount of hydrogen that is entering the reactor, which is almost the same for values from 0.8 to 1.4 times the kinetic factor, since, the hydrogen valve opening is fixed and the hydrogen flow has a chocked condition as mentioned before. For kinetic factor below 0.8 times, the hydrogen flow is not critical and hence, depends on the pressure difference in the hydrogen valve. As a result, the hydrogen flow rate is lower and the reaction takes more time to finish. The changes in the reactor temperature are explained by the same reason since the heat of reaction is a function of the reaction rate. For the kinetic factor values where the hydrogen flow is chocked, the temperature doesn't change because in these cases the reaction rates are very similar since the hydrogen flow rate is equal for these cases. However, for lower values, the reaction rate isn't limited, and the temperature change is visible.

Figure 18 shows the model sensitivity against the global heat coefficient times area.

Figure 18 – Model sensitivity: UA

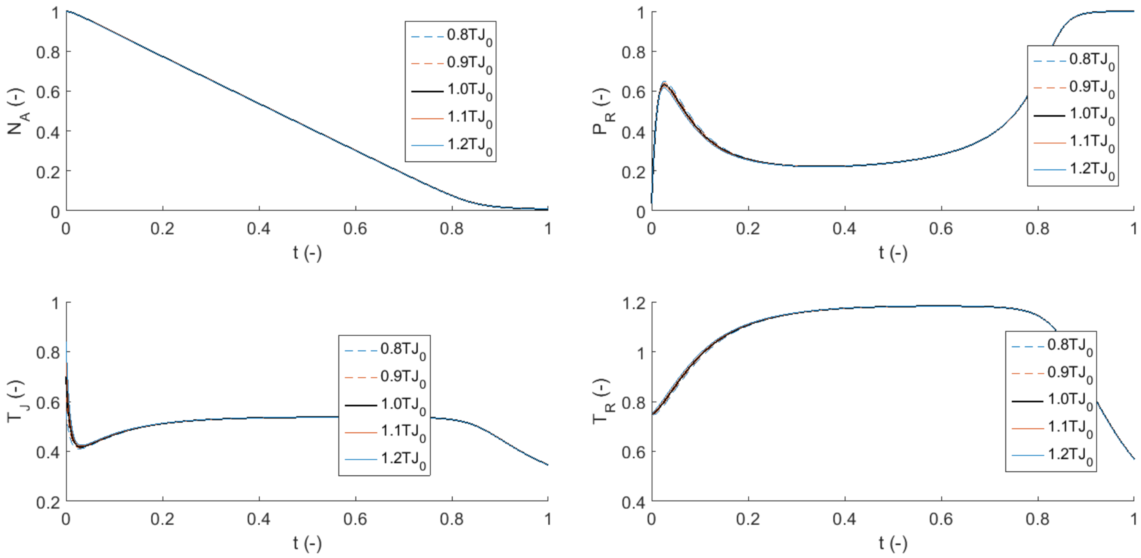


Source: Author

As one can see at T_R graph, the UA directly affects the reactor temperature. The UA represents the heat removal capacity of the system, therefore, for lower UA´s, the reactor temperature is higher. As a result of higher reactor temperature, the hydrogen consumption increases what can be seen as a deeper reactor pressure profile in P_R graph for lower UA´s. Nevertheless, the hydrogen mass rate is still limited by the constant hydrogen valve opening, which explains the fact that the UA does not significantly influence the reaction time, as one can see at N_A graph. Finally, at T_J graph, one could notice that the jacket temperature is only slightly affected by UA changes. This fact can be explained by a cooling fluid flowrate fast enough or a small jacket volume where the residence time heat is small.

Figure 19 shows the model sensitivity against the initial jacket temperature.

Figure 19 – Model sensitivity: T_{J_0}

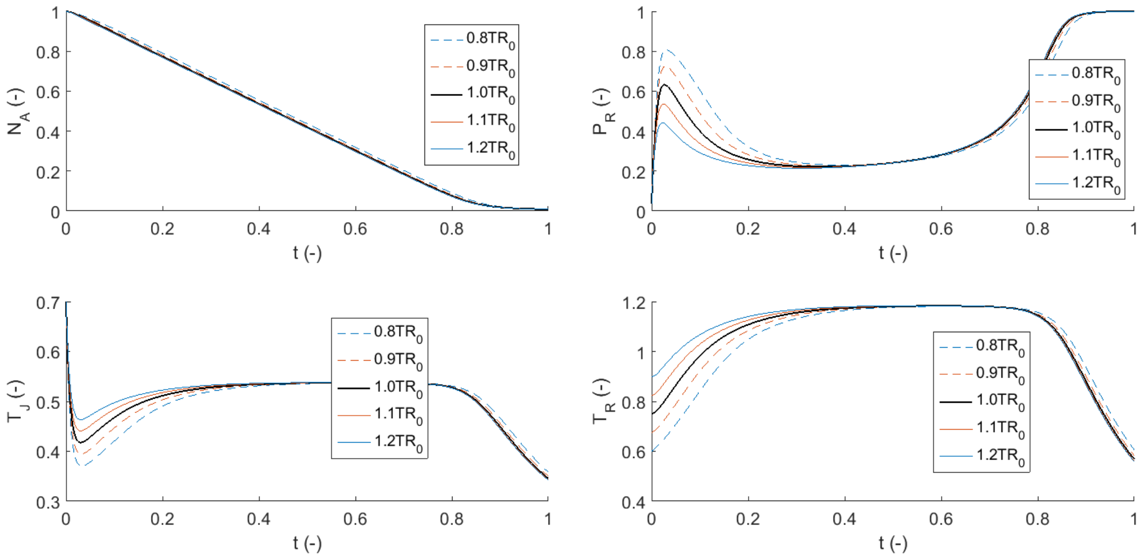


Source: Author

The initial jacket temperature has no significant effect on any parameter as one can see in the profiles shown in Figure 19. This fact can be explained by a cooling fluid flowrate fast enough and/or a small jacket volume where the residence time is too small and thus, as soon as, the cooling fluid enters the reactor jacket it expulses very fast the cooling fluid inside the jacket.

Figure 20 shows the model sensitivity against the initial reactor temperature.

Figure 20 – Model sensitivity: T_{R_0}

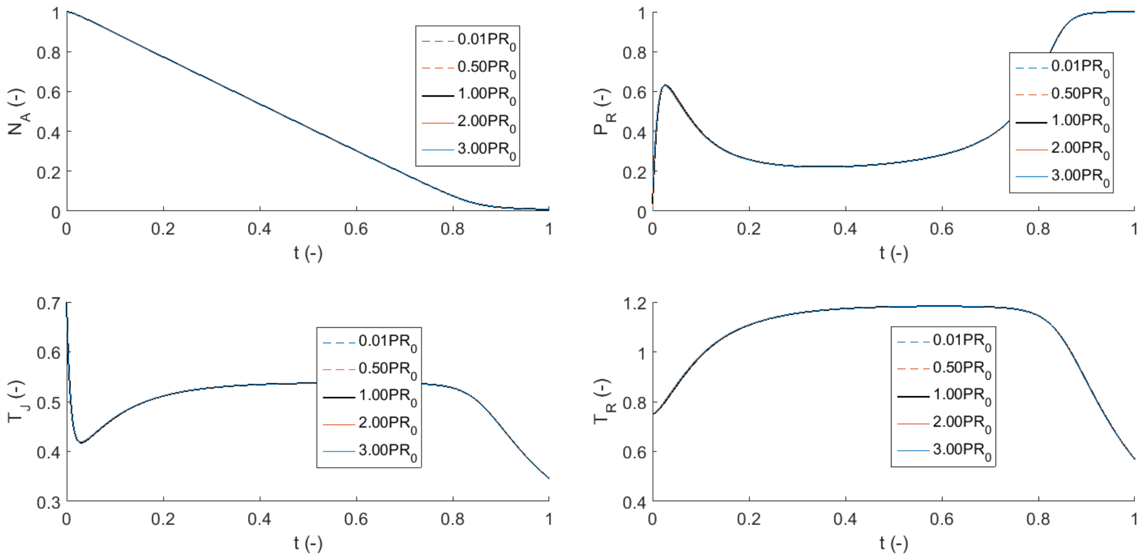


Source: Author

As one can see at P_R graph, the reactor initial temperature strongly affects the pressure profile. A smaller temperature directly reduces the reaction rate and, hence, less hydrogen is consumed by the reaction. As a consequence, the accumulated hydrogen pressures up the reactor. The same occurs for the reactor temperature. As the reaction rate is lower, it gets more time to increase the temperature inside the reactor. The opposite effect occurs for a bigger initial temperature, where the hydrogen consumption is faster and therefore, the pressure inside the reactor remains in a low level while the temperature rises fast. The jacket temperature follows the reactor temperature as expected. Nevertheless, it is interesting to see at N_A graph, that the reaction time doesn’t change. Although the reaction rate is faster for higher temperature, the hydrogen consumption is limited by the choked hydrogen flow rate which is the same for all cases.

Figure 21 shows the model sensitivity against the initial reactor pressure.

Figure 21 – Model sensitivity: P_{R_0}

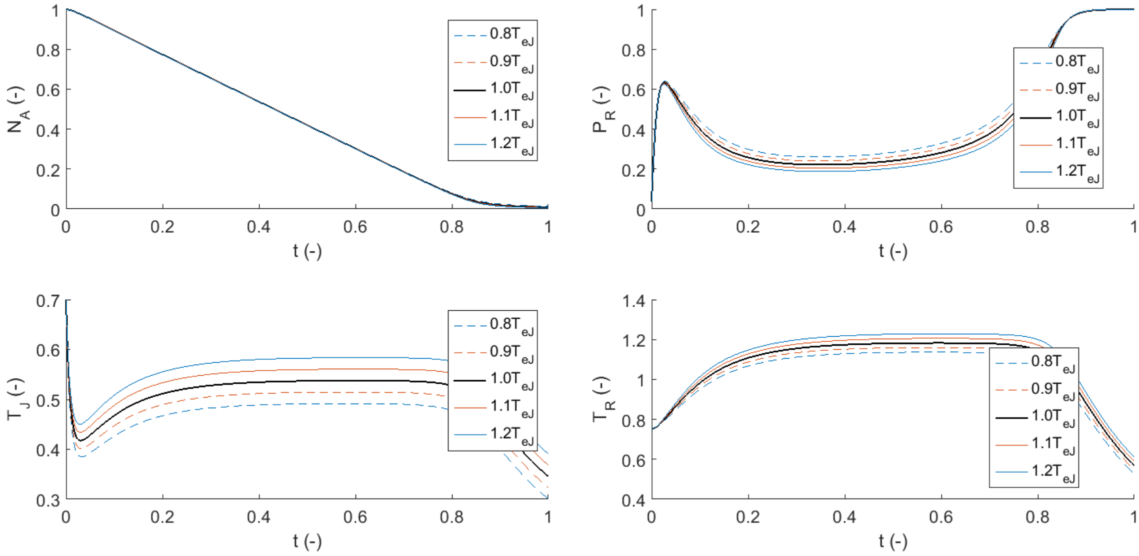


Source: Author

For the range assessed, the reactor initial pressure doesn´t have any significant effect in the process behavior as can be seen in Figure 21. The initial pressure determines the initial amount of hydrogen in the reactor headspace which affects the reaction rate, thus for a bigger initial pressure, one could expect a higher hydrogen consumption. However, in this case, the hydrogen consumption is limited by the catalyst amount and hydrogen flowrate. Therefore, only in the very beginning, a slight difference in the reaction rate can be noticed and then, very quickly the hydrogen consumption and the reaction rate for all cases converge to the same values. For that reason, no impact can be seen in any graph.

Figure 22 shows the model sensitivity against the cooling fluid inlet temperature.

Figure 22 – Model sensitivity: T_{eJ}

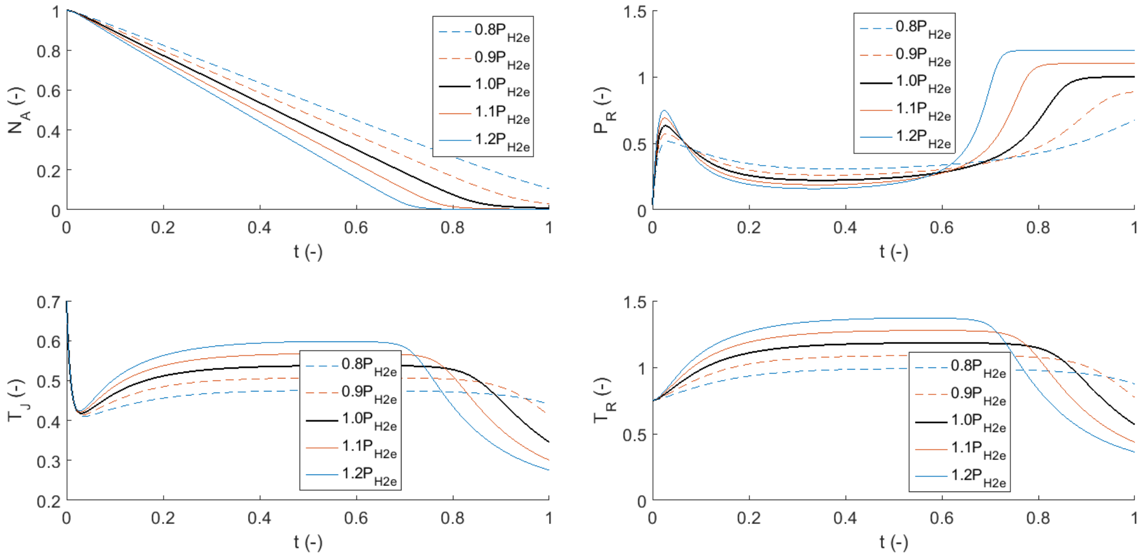


Source: Author

The cooling fluid inlet temperature has a direct and proportional impact in the reactor pressure and temperature, and in the jacket temperature. As expected, for cooler inlet temperatures, higher is the delta between reactor temperature and jacket temperature, therefore, more heat is removed and the temperature goes down. As a consequence, for lower temperatures, more hydrogen is accumulated in the reactor and the pressure stays at a higher level. However, the reaction time is not affected because the hydrogen flow rate is the same since the flow is choked in the major part of the reaction and the hydrogen valve is in a fixed position.

Figure 23 shows the model sensitivity against the hydrogen inlet pressure.

Figure 23 – Model sensitivity: $P_{H_{2,e}}$

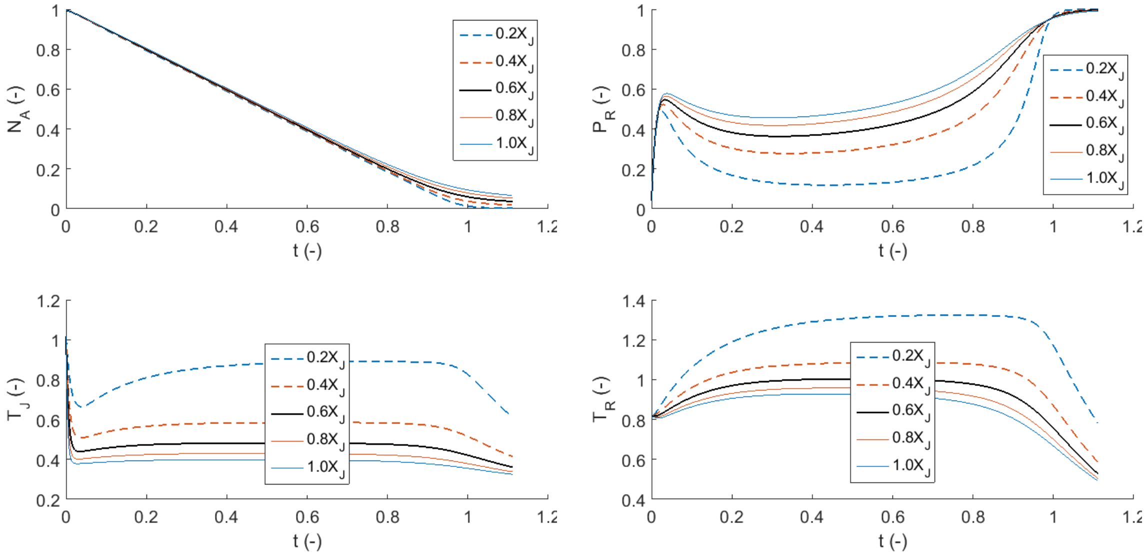


Source: Author

As one can see in Figure 23, a higher hydrogen inlet pressure causes a higher accumulation of hydrogen at the beginning of the reaction, however, as the pressure goes up, the reaction rate tends to increase and consumption of hydrogen starts to sharply occurs. As consequence, the pressure goes to a deeper level during the reaction. By the same chain of events, as the reaction rate increases, the temperature goes up as well as a result of the reaction heat release. With a higher reaction heat, the jacket struggles to remove the heat and its temperature increases as well. Finally, a faster reaction rate one can notice that the reaction time is strongly influenced by the hydrogen inlet pressure since the hydrogen flow rate is directly proportional to the inlet pressure.

Figure 24 shows the model sensitivity against cooling fluid valve opening. It can be seen at T_J graph that the jacket temperature is higher at lower cooling fluid valve openings since it increases the residence time inside the jacket and, hence, allows longer absorption of reaction heat. Consequently, reactor temperature increases since heat removal is worse for smaller temperature delta between the cooling fluid and reactor. As a result of higher reactor temperature, the hydrogen consumption increases which can be seen in P_R graph through the reactor pressure profile that is deeper for lower cooling fluid valve openings. Nevertheless, the hydrogen mass rate is still limited by the constant hydrogen valve opening, which explains the fact that the cooling fluid valve opening does not significantly influence the reaction time, as one can see at N_A graph.

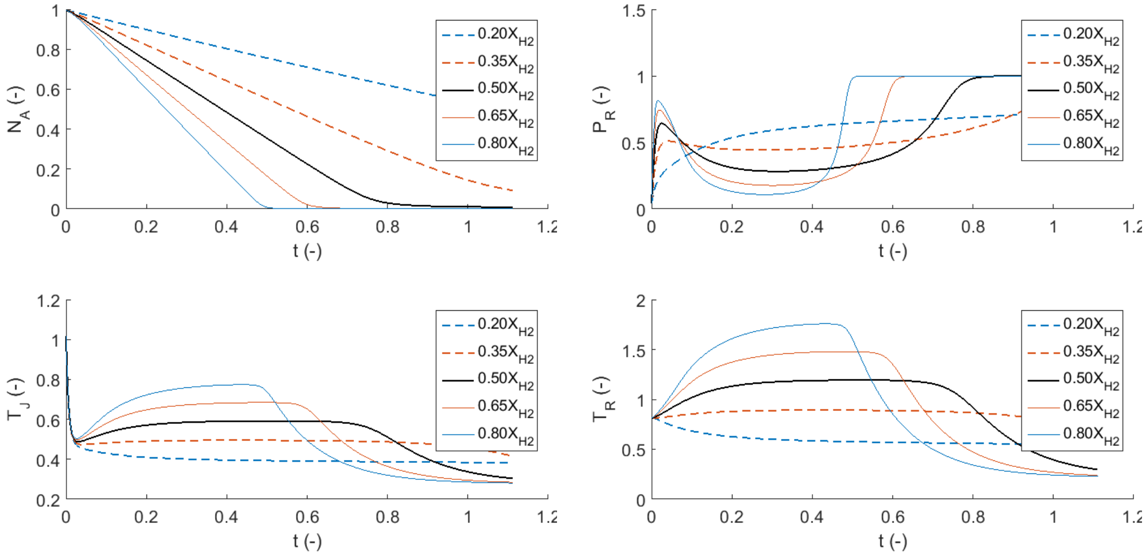
Figure 24 – Model sensitivity: Cooling fluid control valve opening (X_J)



Source: Author

Figure 25 shows the model sensitivity against the hydrogen valve opening.

Figure 25 – Model sensitivity: Hydrogen control valve opening (X_{H_2})



Source: Author

As one can see at N_A graph, the hydrogen valve opening directly affects the reaction time. For higher openings of the hydrogen valve, the reactant A total consumption is faster. This can be explained by the fact the hydrogen is also a reactant and, thus, limiting its mass rate (by limiting valve opening) can constrain the reaction rate. As a result, the reactor temperature is higher for higher hydrogen valve openings, which creates a self-sustaining increase of the reaction rate (Arrhenius Law) and contribute to the faster consumption of the hydrogen mass that enters the reactor gas phase. This results in the valley behavior of reactor pressure, that can be seen at P_R graph. Finally, at T_J graph, one could notice that the jacket temperature is directly influenced by reactor temperature when the cooling fluid flowrate to the jacket is kept constant.

4.3 Proposal of temperature control strategies

As explained in section 3.2, the current temperature control strategy is an ordinary PI feedback loop. It is known that feedback controllers may not be suitable when the process presents slow dynamics and/or is subject to disturbances and/or is subjected to a small operating ranges (CORRIPIO; SMITH, 2006). Since the temperature is a slow variable and the current process is subjected to different disturbances, the current controller strategy loses robustness in the presence of process disturbances. Besides, the feedback framework can suffer to maintain the controlled variable under strict constraints and is not designed to optimize the process.

Moreover, since the process is operated in a semi-batch mode, additional nonlinearities are introduced. As shown in section 2.4, the standard PID control struggles to perform in a highly nonlinear environment. However, PID controllers are still effective in cases where a highly nonlinear process is operated in a narrow range or if the nonlinearity level is not so high (SEBORG *et al.*, 2017). Nevertheless, the absence of a steady-state condition in semi-batch reactors makes the PID parameters tuning a challenge and in many cases, they have to be changed depending on batch step (SMITH, 2014).

In this context, it was proposed three new control techniques which were then compared between each other and with the current control strategy of the industrial hydrogenation reactor. These techniques were chosen due to its simplicity and feasibility to implement at the industrial plant.

For all the proposed strategies, an anti-reset windup method was applied in the controller’s implementation. The reset windup phenomenon is a well-known problem which occurs when the controller keeps integrating and building-up its integral term after its saturation and, as consequence, the controller response can be inadequate and cause large overshoots (CORRIPIO; SMITH, 2006). There are several methods to avoid this phenomenon but one of the most used for batch processes consists of the freezing of the integral term when the controller saturates (SEBORG *et al.*, 2017). Therefore, this approach was the one used in this work.

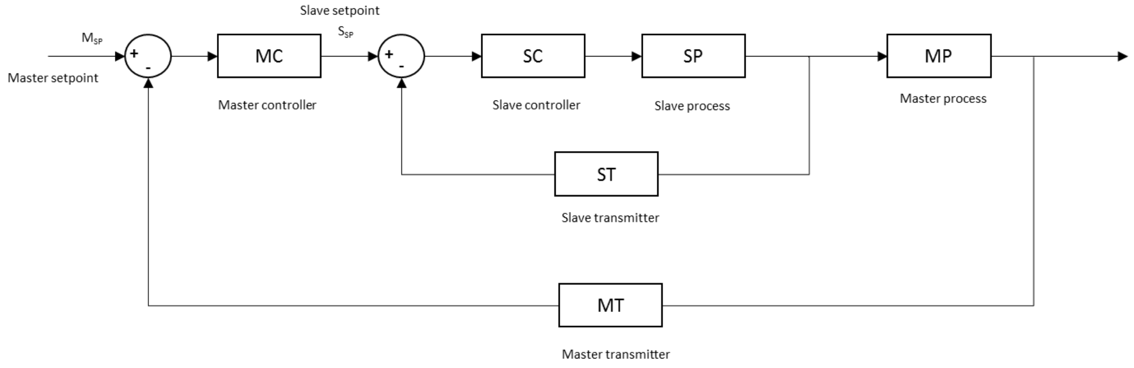
Although the derivative term of a PID controller can sometimes bring advantages, it was not used in the proposed strategies since all controllers of the industrial plant use only PI frameworks. Moreover, the majority of the industrial controllers uses only PI terms (CORRIPIO; SMITH, 2006; WADE, 2004). In addition, only PI controllers are often used for batch and semi-batch processes as can be seen in several works (GIL; VARGAS; CORRIOU, 2014; LI *et al.*, 2014; ŠTAMPAR; SOKOLIČ; KARER, 2013) even when self-tuning and adaptative frameworks are used (GRAICHEN; HAGENMEYER; ZEITZ, 2006; VASANTHI; PRANAVAMOORTHY; PAPPA, 2012).

4.4 Cascade control

It is usual to see cascade control schemes applied to temperature control of jacketed reactors (CORRIPIO; SMITH, 2006; SEBORG *et al.*, 2017; SMITH, 2014). These schemes consist of a primary controller for the reactor temperature which determines the set point of the secondary controller for the cooling system. This control framework usually has a better performance than standard feedback loops since it uses two feedback controllers and an intermediate setpoint (Shinskey,1996, apud. SEBORG *et al.*, 2017).

Figure 26 shows a generic block diagram for cascade control.

Figure 26 – Generic cascade control block diagram

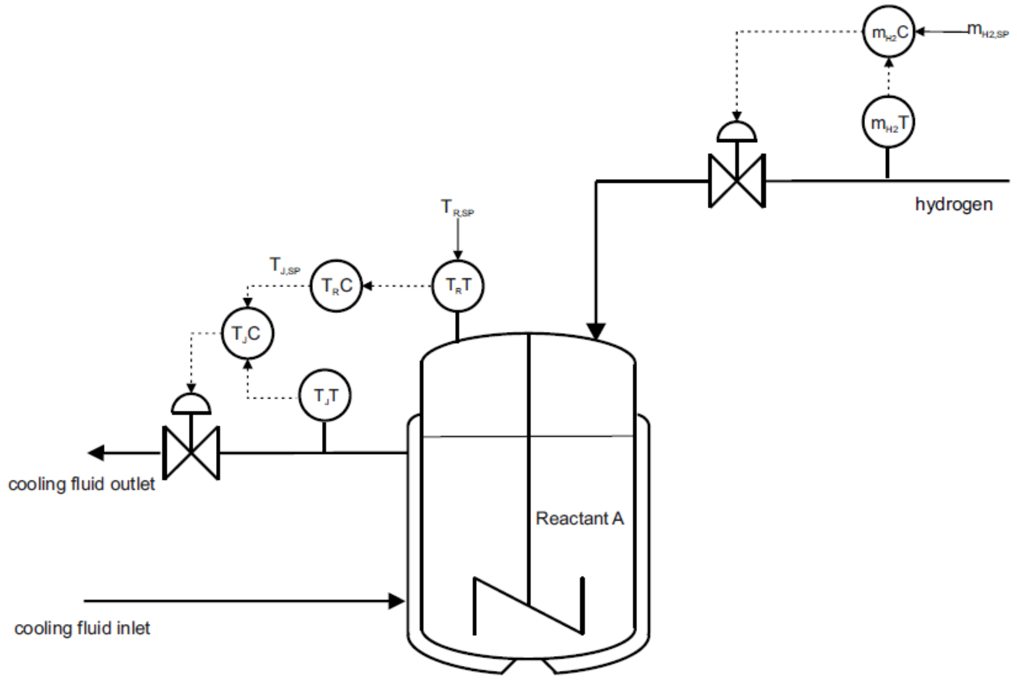


Source: Author

As shown in section 2.4, there are some applications of cascade control to batch reactors. Some studies show that that fixed PID parameters for this kind of control can sacrifices performance (CHYLLA; RANDALL HAASE, 1993) and the best solution would be to implement a self-tuning framework (VASANTHI; PRANAVAMOORTHY; PAPPA, 2012).

Nevertheless, due to the process characteristics, a standard PI cascade control was proposed as can be seen in Figure 27.

Figure 27 – Cascade control scheme



Source: Author

Table 4 shows the cascade controller's strategies and their constitutive equations:

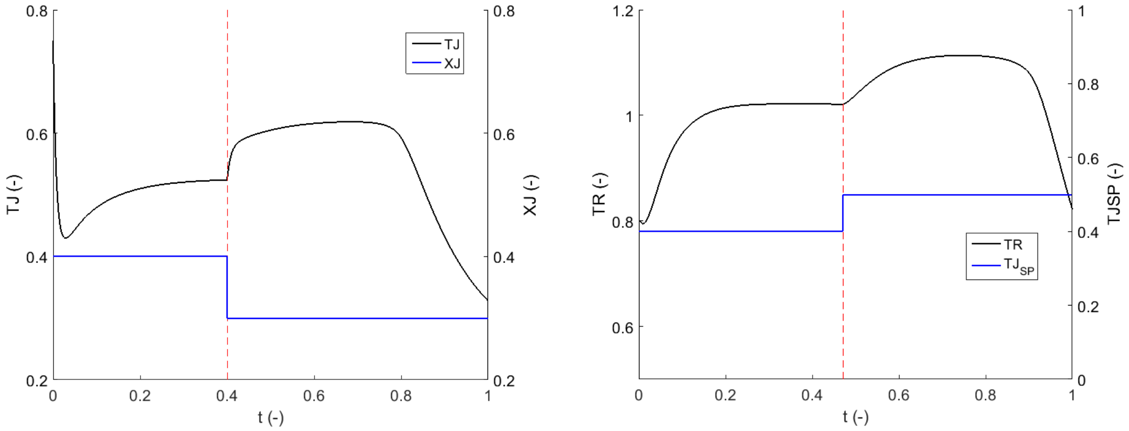
Table 4 - Cascade controller's constitutive equations

Controller	MV	CV	Controllers output	Equation
Jacket temperature (secondary)	X_J	T_J	$Out_{X_J} = bias_{X_J} + Kc_{T_J}\left(\beta_{T_J}.T_{J,SP} - T_J\right) + \frac{Kc_{T_J}}{\tau_{I,T_J}}\int\left(T_{J,SP} - T_J\right)dt$	(45)
Reactor temperature (primary)	$T_{J,SP}$	T_R	$Out_{T_{J,SP}} = bias_{T_{J,SP}} + Kc_{J_{SP}}\left(\beta_{T_{J,SP}}.T_{J,SP} - T_R\right) + \frac{Kc_{J_{SP}}}{\tau_{I,T_{SP}}}\int\left(T_{R,SP} - T_R\right)dt$	(46)

Source: Author

As the open-loop dynamics show a self-regulating dynamic for both jacket and reactor temperatures, the PI tuning parameters for the secondary and primary control were adjusted by an open-loop test applying a step disturbance on the inlet cooling fluid valve and jacket temperature setpoint, respectively, as can be seen in Figure 28.

Figure 28 – Open loop step test: Jacket and reactor temperatures response

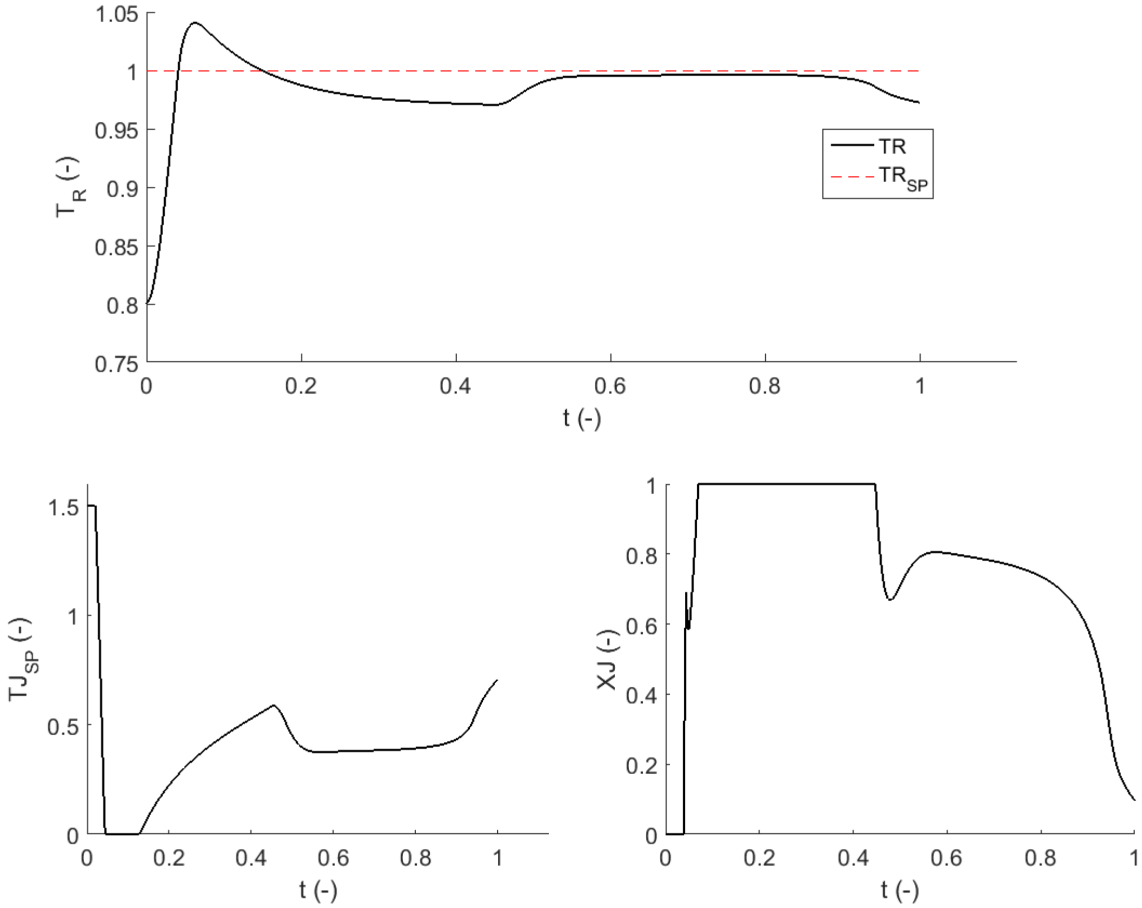


Source: Author

The jacket and reactor temperatures responses were then fitted to a first-order plus dead time (FOPDT) model and the controller's parameters were tuned using the 1/4 decay ratio method developed by Ziegler and Nichols (1942). The process response with the implemented strategy is shown in Figure 29.

The open-loop step tests were performed only when the process had already achieved a “steady-state” temperature. This was necessary since the selected tuning method is based on a steady-state response after a step disturbance. This is one of the main challenges of tuning batch processes since the temperature transient phase dynamics does not match the “steady-state” phase for which the tuning parameters were tuned and can cause overshoot.

Figure 29 – Cascade closed loop response with raw tuned parameters



Source: Author

Therefore, after the Ziegler and Nichols method, a trial-and-error approach was done to fine-tune the parameters which showed to be good enough to overcame this difficulty. Figure 30 shows the results of the closed-loop with the determined PI tuning parameters after the try-and-error fine-tuning. The values of the PI parameters can be found in Table 5.

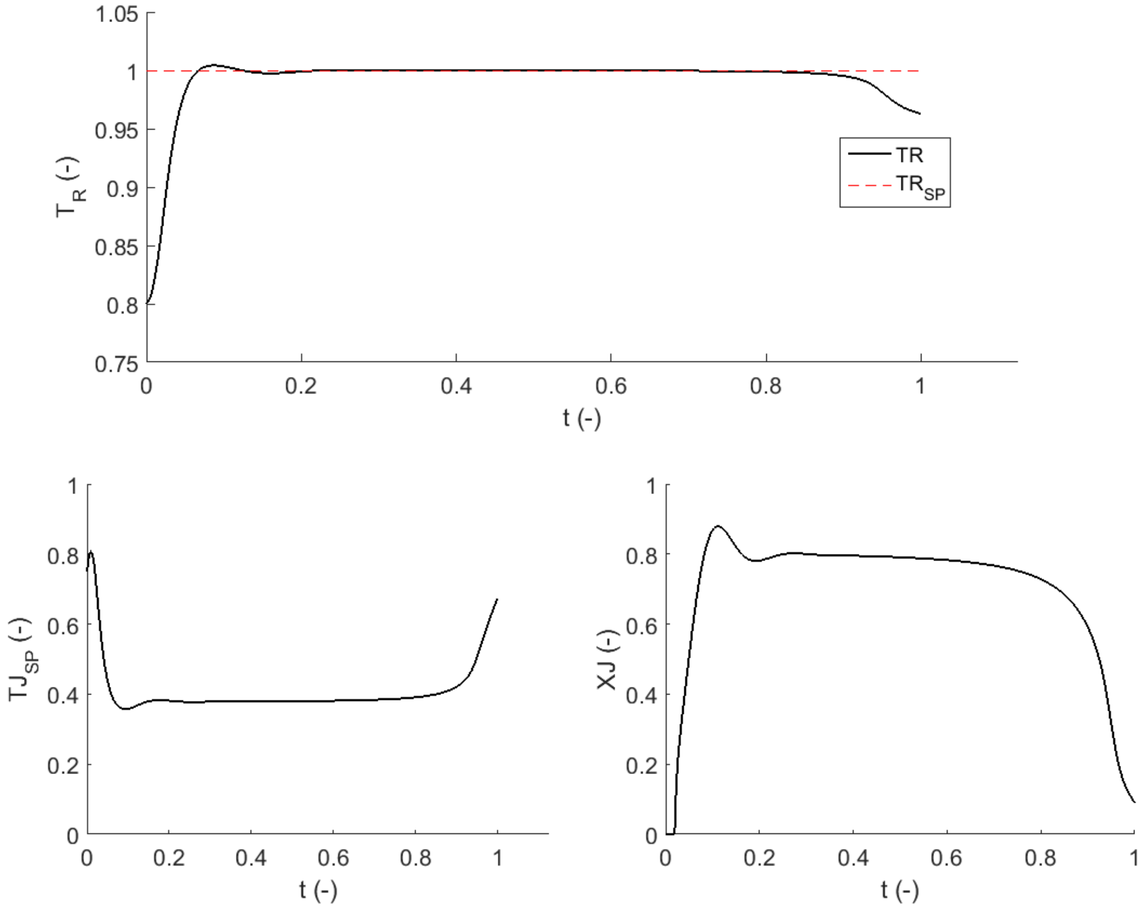
Table 5 - Cascade control parameters

Control parameters	Values
Kc_{T_J}	-10
τ_{I,T_J}	875
β_{T_J}	1
$Kc_{J_{SP}}$	5
$\tau_{I,T_{SP}}$	1800
$\beta_{T_{J,SP}}$	1

Source: Author

Note that the temperature drop at the end of the reaction is a normal behavior as one can see in Figure 13 and Figure 14 of section 4.1, as a result of the reaction rate reduction due to low concentration of reactant A and, thus, the equalization of pressures between the hydrogen feed and the reactor, reducing hydrogen flow into the reactor and indicating the reaction end. This fact is a natural behavior and doesn’t affect the quality or batch time and, thus, was not considered as a problem to be corrected by the controllers proposed.

Figure 30 – Cascade closed loop response after trial-and-error tuning parameters



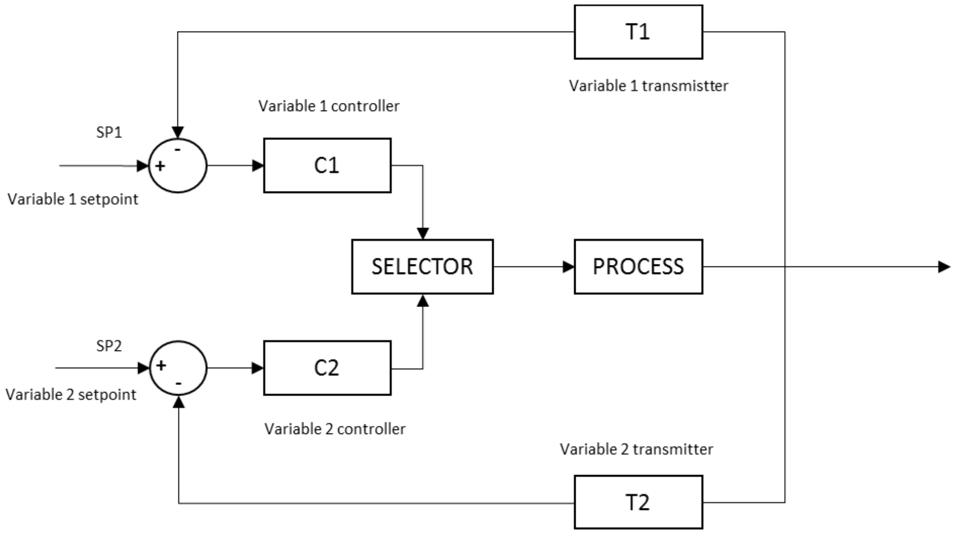
Source: Author

4.5 Override control

The override control is another commonly used control technique for temperature control of batch process. This framework is used as a simple optimization strategy that enables a soft transition between controllers to maximize/minimize a variable, and/or when there is a strict operating range whether due to quality or safety reasons (CORRIPIO; SMITH, 2006). This framework consists of the use of a selector which will switch the controller that will override and assume the control of the final element depending on the selector type chosen (e.g. maximum selector, minimum selector) (WADE, 2004).

In many cases, it comes with a valve position control embed which consider the position of a final control element as process variable. Smith (2010) shows many applications of override control and valve position control in batch reactors. There are many possible frameworks for this kind of control, Figure 31 shows a generic form of an override control based on two feedback loops which are activated depending on the selection.

Figure 31 – Generic override control block diagram

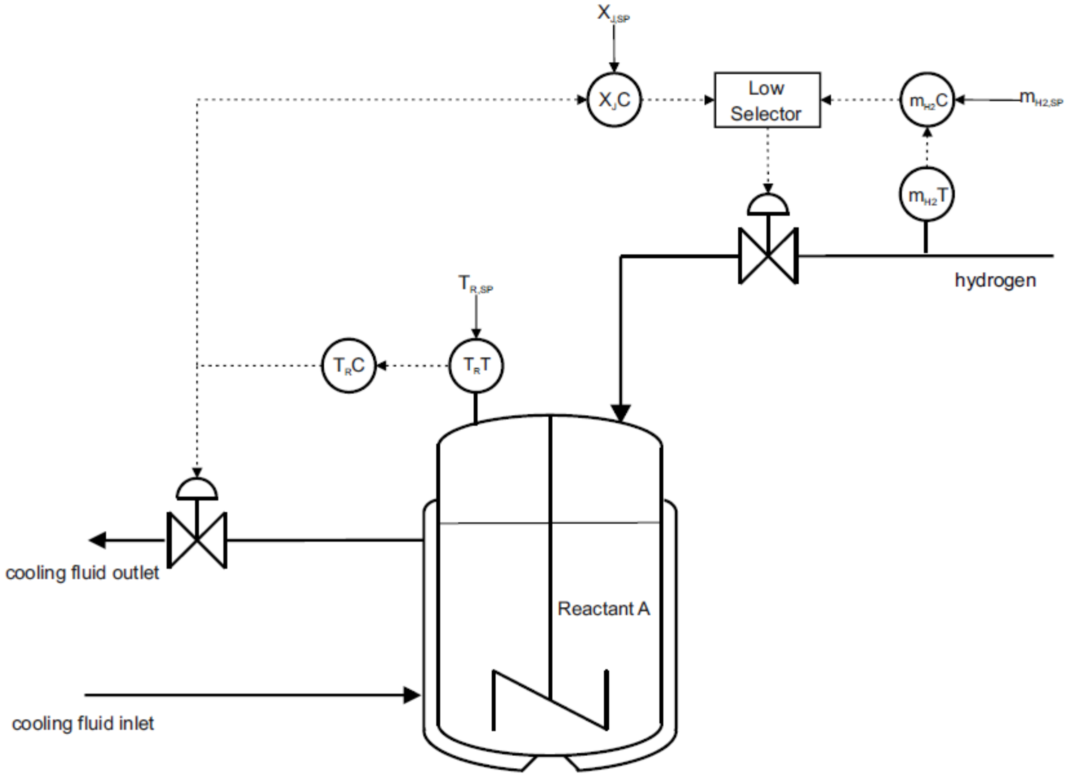


Source: Author

As can be seen in Figure 32, to this case study, it was proposed an override control with a minimum selector. The hydrogen valve is manipulated by two controllers, a hydrogen flow rate PI feedback controller and a PI feedback valve position controller (VPC). The first one is designed to keep the hydrogen flow rate to the reactor at a constant setpoint. The other one has the objective of keeping the cooling fluid valve in a pre-determined position.

The main goal of this control strategy is to maximize the hydrogen flow rate if there is enough cooling capacity to remove the reaction heat. If the cooling valve is below the determined position setpoint due to the reactor temperature controller output, the hydrogen flow rate can be increased. On the other hand, if the cooling valve is already above the determined position setpoint, the hydrogen flow rate needs to be reduced in order reduce reaction rate and thus, keep the reactor temperature under control. The selection of the controller is made by a minimum selector which ensures that always the critical controller overrides the control over the hydrogen inlet valve.

Figure 32 – Override control scheme



Source: Author

Table 6 shows the override controller's strategies and their constitutive equations:

Table 6 – Override controller's constitutive equations

Controller	MV	CV	Controllers output	Equation
Hydrogen flowrate	X_{H_2}	$\dot{m}_{H_{2,e}}$	$Out_{H_2} = bias_{H_2} + Kc_{H_2}\left(\dot{m}_{H_{2,e},SP} - \dot{m}_{H_{2,e}}\right) + \frac{Kc_{H_2}}{\tau_{I,H_2}} \int \left(\dot{m}_{H_{2,e},SP} - \dot{m}_{H_{2,e}}\right) dt$	(47)
Cooling fluid valve position	X_{H_2}	X_J	$Out_{H_2} = bias_{X_J} + Kc_{X_J}\left(\beta_{X_J}\,.X_{J,SP} - X_J\right) + \frac{Kc_{X_J}}{\tau_{I,X_J}} \int \left(X_{J,SP} - X_J\right) dt$	(48)
Reactor temperature	X_J	T_R	$Out_{T_R} = bias_{X_J} + Kc_{T_R}\left(\beta_{T_R}.T_{R,SP} - T_R\right) + \frac{Kc_{T_R}}{\tau_{I,T_R}} \int \left(T_{R,SP} - T_R\right) dt$	(49)

Source: Author

The PI parameters of the hydrogen flowrate controller were the same used in the real process control strategy. The PI parameters of the valve position controller were determined by a trial-and-error approach since its responses didn't fit to a first-order plus dead time (FOPDT) model as used for the cascade controller. The PID parameters tuning of a valve position control can be very challenging due to the lack of robust tuning procedures and the trial and error approach is often used (ALLISON; OGAWA, 2003). In addition, based on practical experience, the tuning for this kind of controller is difficult and can to introduce oscillations to the system.

The values of the PI parameters can be found in Table 7.

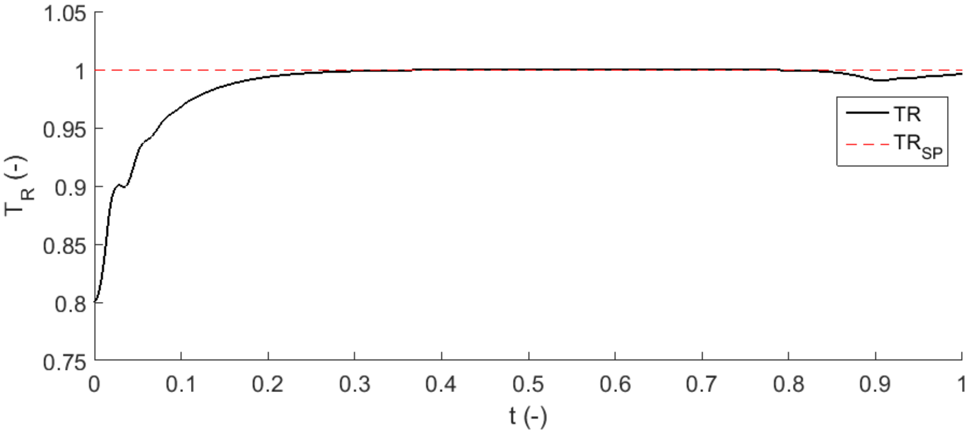
Table 7 - Override control parameters

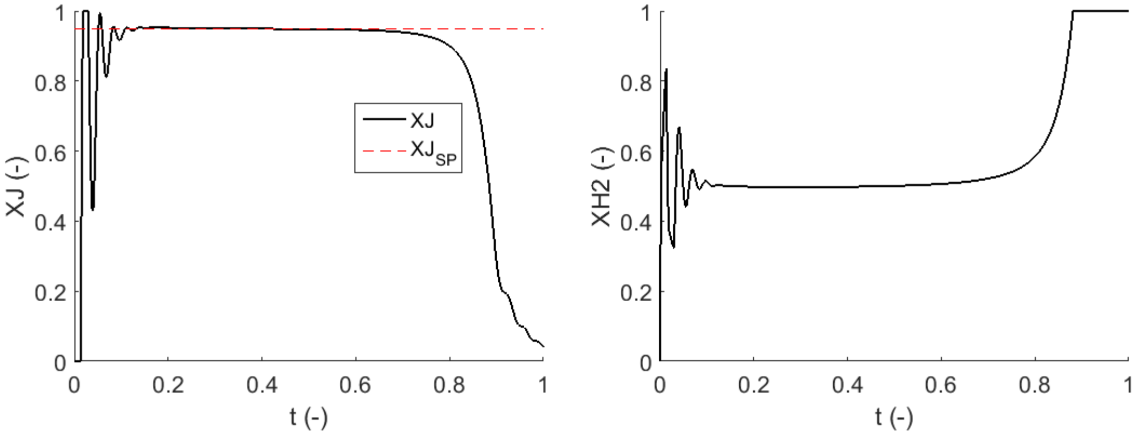
Control parameters	Values
Kc_{H_2}	0.1
τ_{I,H_2}	6
Kc_{X_J}	20
τ_{I,X_J}	800
β_{X_J}	1
Kc_{T_R}	-90
τ_{I,T_R}	1800
β_{T_R}	0.81

Source: Author

Figure 33 shows the results of the override control with the determined PI tuning parameters.

Figure 33 – Override closed loop response after try-and-error tuning parameters





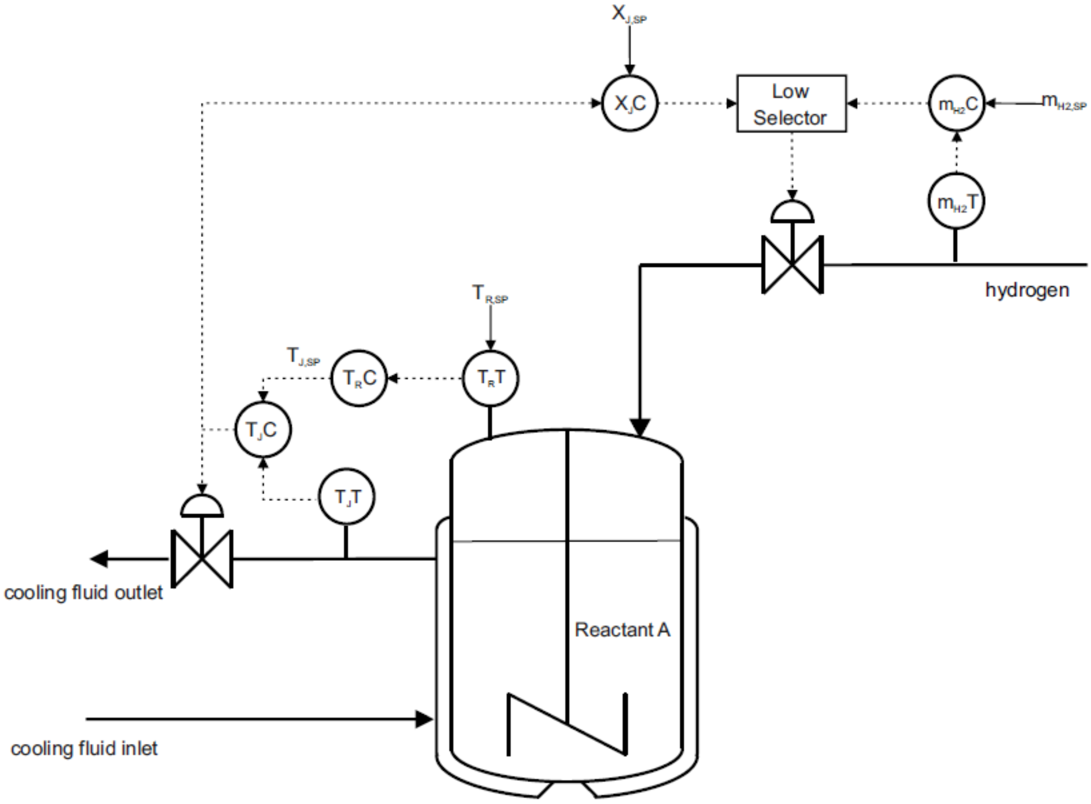
Source: Author

As mentioned before, the valve position control embed in the override controller can cause oscillation at the hydrogen valve as can be seen in Figure 33. Although the oscillation seems to be fast, it can be considered reasonable regarding the real time scale.

4.6 Cascade + override control

After a preliminary visual evaluation of the controlled and manipulated variables for the previous proposed control strategies, a combination of the cascade and override control is now proposed. The cascade control seems to have a faster response in terms of the temperature setpoint tracking. On the other hand, the override control maximizes the hydrogen flowrate. This proposed framework can be better observed in Figure 34.

Figure 34 – Cascade+override control scheme



Source: Author

Table 8 shows the override controller's strategies and their constitutive equations:

Table 8 – Cascade+override controllers’ constitutive equations

Controller	MV	CV	Controllers output	Equation
Jacket temperature (secondary)	X_J	T_J	$Out_{X_J} = bias_{X_J} + Kc_{T_J}\left(\beta_{T_J}\,.T_{J,SP} - T_J\right) + \frac{Kc_{T_J}}{\tau_{I,T_J}}\int\left(T_{J,SP} - T_J\right)dt$	(50)
Reactor temperature (primary)	$T_{J,SP}$	T_R	$Out_{T_{J,SP}} = bias_{T_{J,SP}} + Kc_{T_{J,SP}}\left(\beta_{T_{J,SP}}.\,T_{R,SP} - T_R\right) + \frac{Kc_{T_{J,SP}}}{\tau_{I,T_{J,SP}}}\int\left(T_{R,SP} - T_R\right)dt$	(51)
Hydrogen flowrate	X_{H_2}	$\dot{m}_{H_{2,e}}$	$Out_{H_2} = bias_{H_2} + Kc_{H_2}\left(\dot{m}_{H_{2,e},SP} - \dot{m}_{H_{2,e}}\right) + \frac{Kc_{H_2}}{\tau_{I,H_2}}\int\left(\dot{m}_{H_{2,e},SP} - \dot{m}_{H_{2,e}}\right)dt$	(52)
Valve position	X_{H_2}	X_J	$Out_{H_2} = bias_{X_J} + Kc_{X_J}\left(\beta_{X_J}\,.X_{J,SP} - X_J\right) + \frac{Kc_{X_J}}{\tau_{I,X_J}}\int\left(X_{J,SP} - X_J\right)dt$	(53)

Source: Author

The PI parameters of the cascade control were the same determined in section 4.4. The PI parameters of the valve position controller were determined by a trial-and-error approach. Combining both control techniques made it more difficult to tune the PI parameters. Therefore, further studies in PID tuning techniques should be made to improve controller performance. The values of the PI parameters can be found in Table 9.

Table 9 - Override control parameters

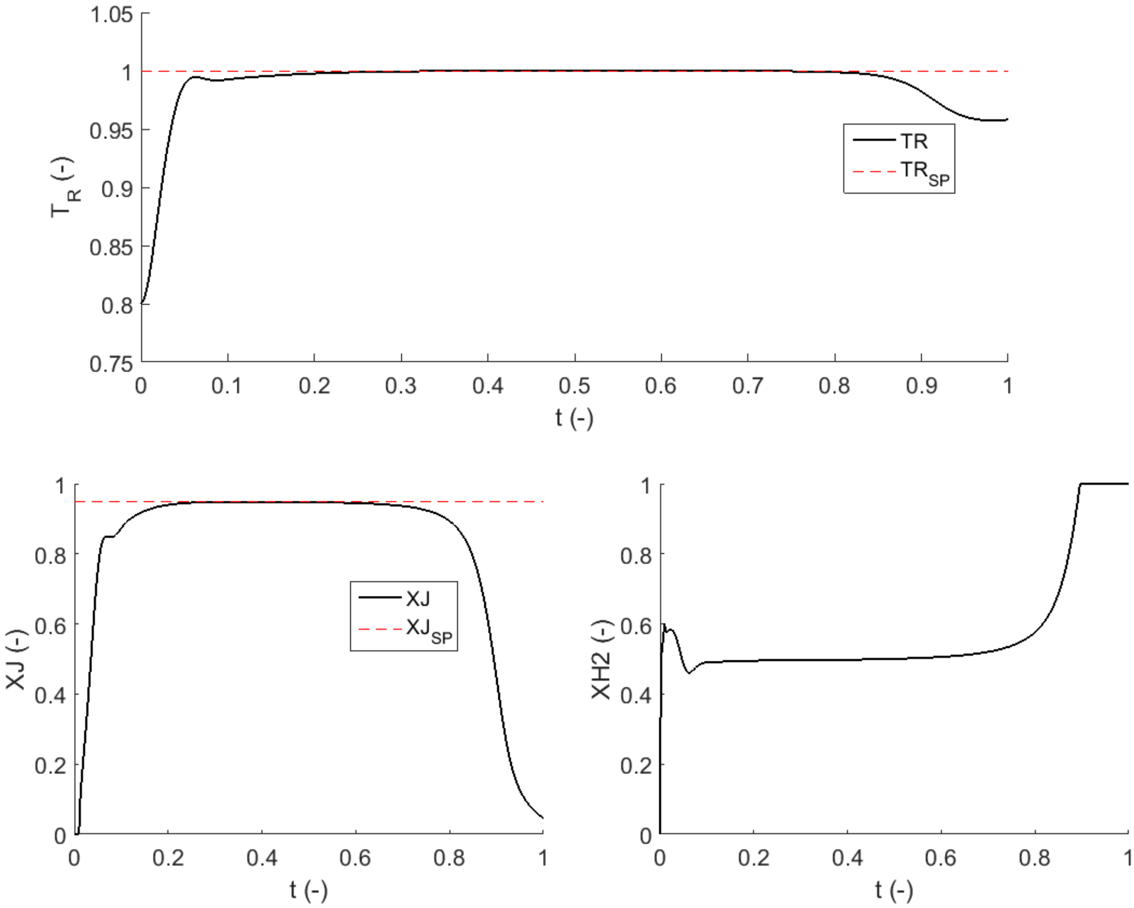
Control parameters	Values
Kc_{T_J}	-10
τ_{I,T_J}	875
β_{T_J}	1
$Kc_{J_{SP}}$	5
$\tau_{I,T_{SP}}$	1800
$\beta_{T_{J,SP}}$	1
Kc_{T_J}	-10
Kc_{H_2}	0.1
τ_{I,H_2}	6
Kc_{X_J}	100
τ_{I,X_J}	800
β_{X_J}	0.7

Source: Author

Figure 35 shows the results of the cascade + override control. The hydrogen valve shows an oscillatory behavior but less significant when observed with solely override framework. Regarding X_J graph, the valve position controller is not so fast to reach the setpoint of X_J. This fact is related to the tuning parameters. When more aggressive tuning parameters were chosen, X_J achieved faster its setpoint. However, X_{H_2} oscillated more, and as a consequence, interfered in reactor temperature control, exceeding the quality limits.

Comparing to the previous strategies, for this case, in the beginning of the batch the temperature showed a faster increase and, suddenly, a sharp attenuation. It can be observed, that in the very beginning of reaction, the hydrogen valve is being controlled by the hydrogen flowrate controller since the cooling fluid valve is still too far from its setpoint. As long as the cooling fluid valve position gets closer to its setpoint, the valve position controller overrides the hydrogen flowrate controller and closes the hydrogen valve. In this moment, the reaction rate decreases and, as a consequence, the temperature increase rate is reduced.

Figure 35 – Cascade + Override closed loop response after try-and-error tuning parameters



Source: Author

4.7 Temperature control strategies comparison

4.7.1.1 Assessment of controller performance in standard operation

The integral of the value of the absolute error (IAE), the integral of the squared error (ISE) and the integral of the time-weighted absolute error (ITAE) criteria are often used to optimize and evaluate the performance of controllers tuning (SEBORG *et al.*, 2017). Besides, this approach can also be used to compare different control strategies as can be seen in several papers (CANCELIER *et al.*, 2016; HOSEN *et al.*, 2013; HUSSAIN; KERSHENBAUM, 2000; VASANTHI; PRANAVAMOORTHY; PAPPA, 2012).

The criteria aforementioned can be written as the following:

$$IAE = \int_0^t |error| \, dt \tag{54}$$

$$ISE = \int_0^t error^2 \, dt \tag{55}$$

$$ITAE = \int_0^t t \, . \, |error| \, dt \tag{56}$$

Therefore, the IAE, ISE and ITAE criteria were used to compare the performance of the current control strategy and the three new control strategies proposed.

Table 10 shows the IAE, ISE, and ITAE values calculated for the temperature responses of the four control strategies during standard operation.

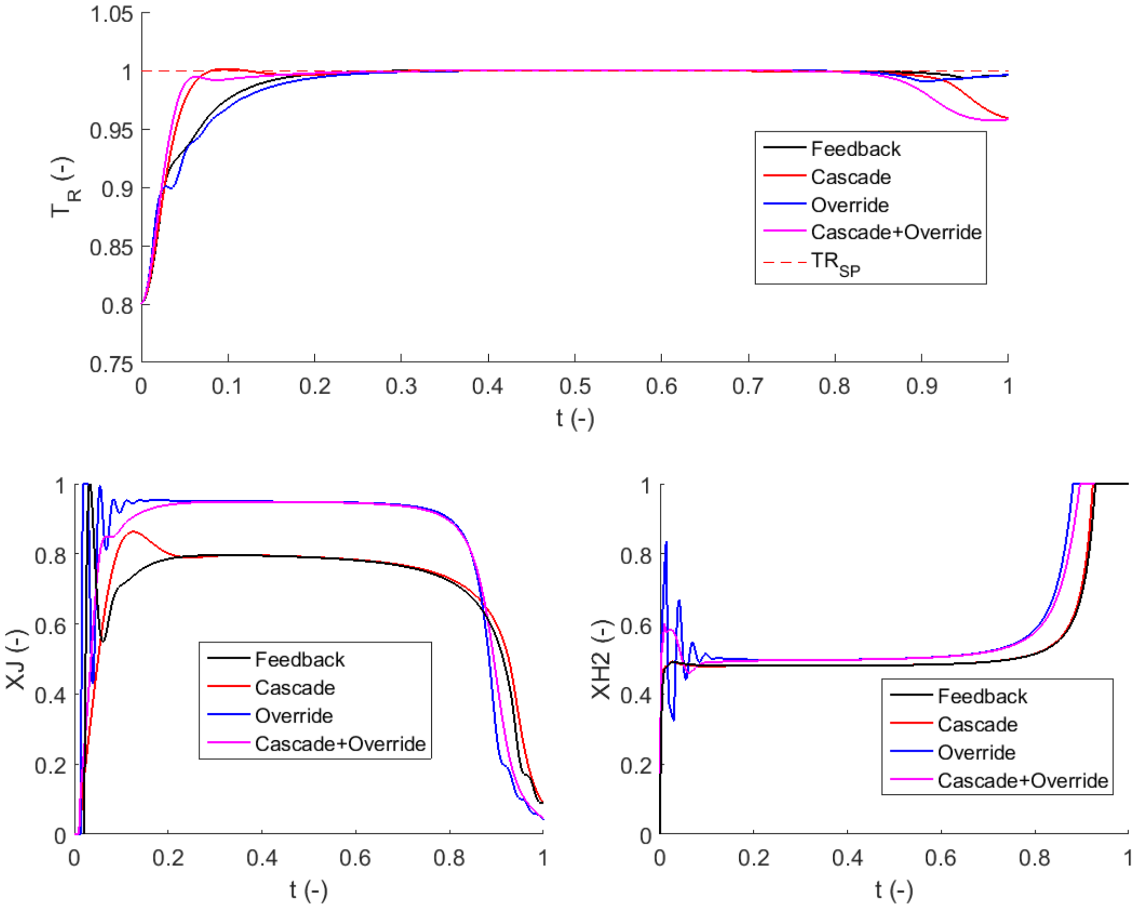
Table 10 – Comparison of IAE, ISE and ITAE values for temperature response during normal operation

Controller Strategy	**IAE**	**ISE**	**ITAE**
Feedback	7.60	78.69	2.90
Cascade	5.12	64.10	1.45
Override	8.45	79.14	3.98
Cascade + Override	**4.80**	**56.27**	**1.41**

Source: Author

As one can see in Table 10, the cascade and the cascade+override controllers showed the lowest IAE, ISE, and ITAE values. The cascade+override was slightly better which indicates that this is the best control strategy during standard operation. These results can be also seen in Figure 36. One can notice that the cascade and the cascade+override achieved the temperature setpoint faster than the other control strategies.

Figure 36 – Control strategies response during normal operation



Source: Author

Under standard conditions and according to the selected criteria, the cascade and cascade+overide controller seems the best choice to control the reactor temperature. However, would be very simplistic to take any conclusion of this first assessment since there are many other factors that can affect the temperature control. In that manner, it is fundamental to evaluate the controllers' performance against non-standard conditions and disturbances.

4.7.1.2 Assessment of controller performance under non-standard operational conditions

As mentioned in section 4.2, the initial mass fraction of A is not a fixed value due to downstream process oscillations, and hence, can directly affect the temperature control, since more or less substance A will be available to react. Similarly, the catalyst activity, represented by the kinetic factor, is an uncontrolled parameter that can disturb the reactor pressure and temperature. Therefore, the controllers' performance was evaluated varying those input conditions parameters.

Table 11 shows the IAE, ISE and ITAE values calculated for the temperature responses of the four control strategies under non-standard operational conditions.

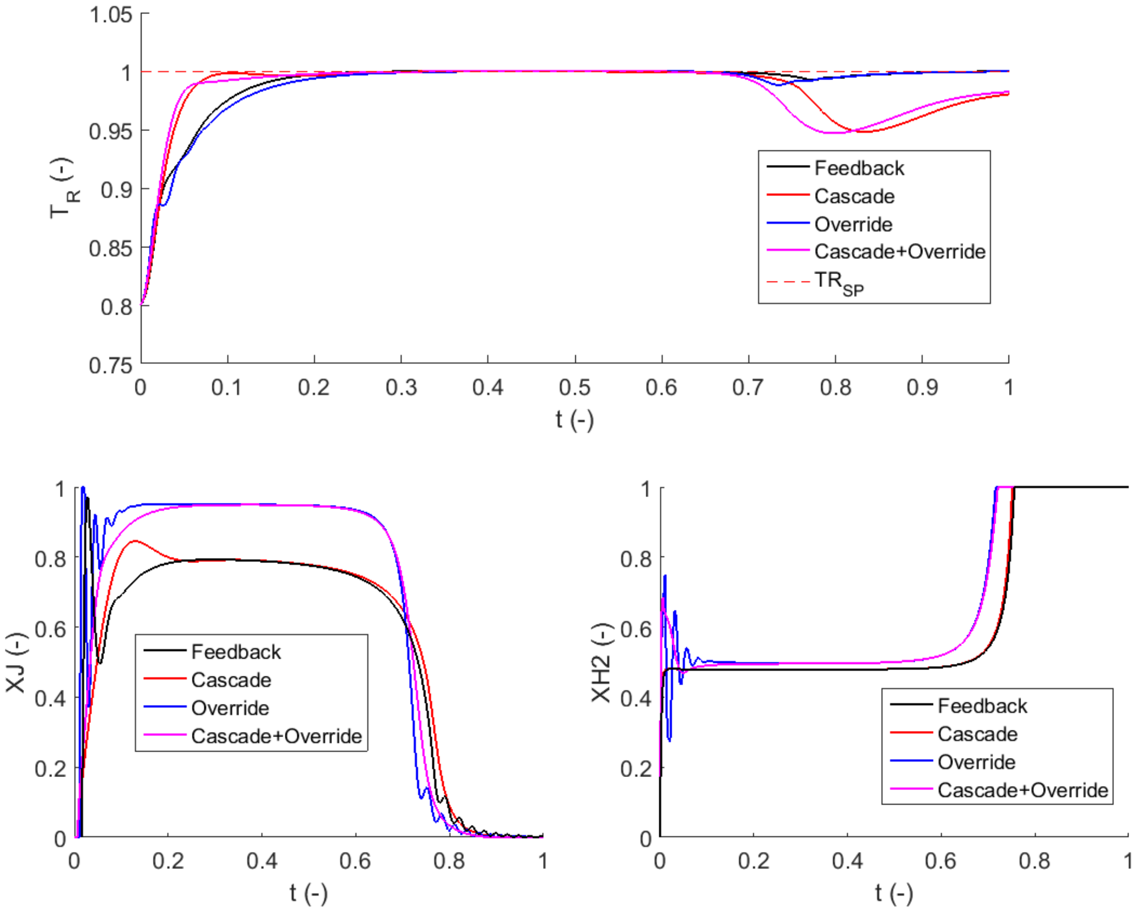
Table 11 – Comparison of IAE, ISE and ITAE values for temperature response under different initial mass fraction of A ($\boldsymbol{x_{A0}}$)

Controller Strategy	**IAE**		**ISE**		**ITAE**	
	$0.8\ x_{A0}$	$1.2\ x_{A0}$	$0.8\ x_{A0}$	$1.2\ x_{A0}$	$0.8\ x_{A0}$	$1.2\ x_{A0}$
Feedback	7.63	7.60	76.67	81.52	3.02	2.83
Cascade	5.12	5.47	59.00	70.25	1.75	1.51
Override	8.45	8.45	78.48	80.18	4.01	3.98
Cascade + Override	**4.80**	**4.87**	**52.34**	**61.45**	**1.55**	**1.32**

Source: Author

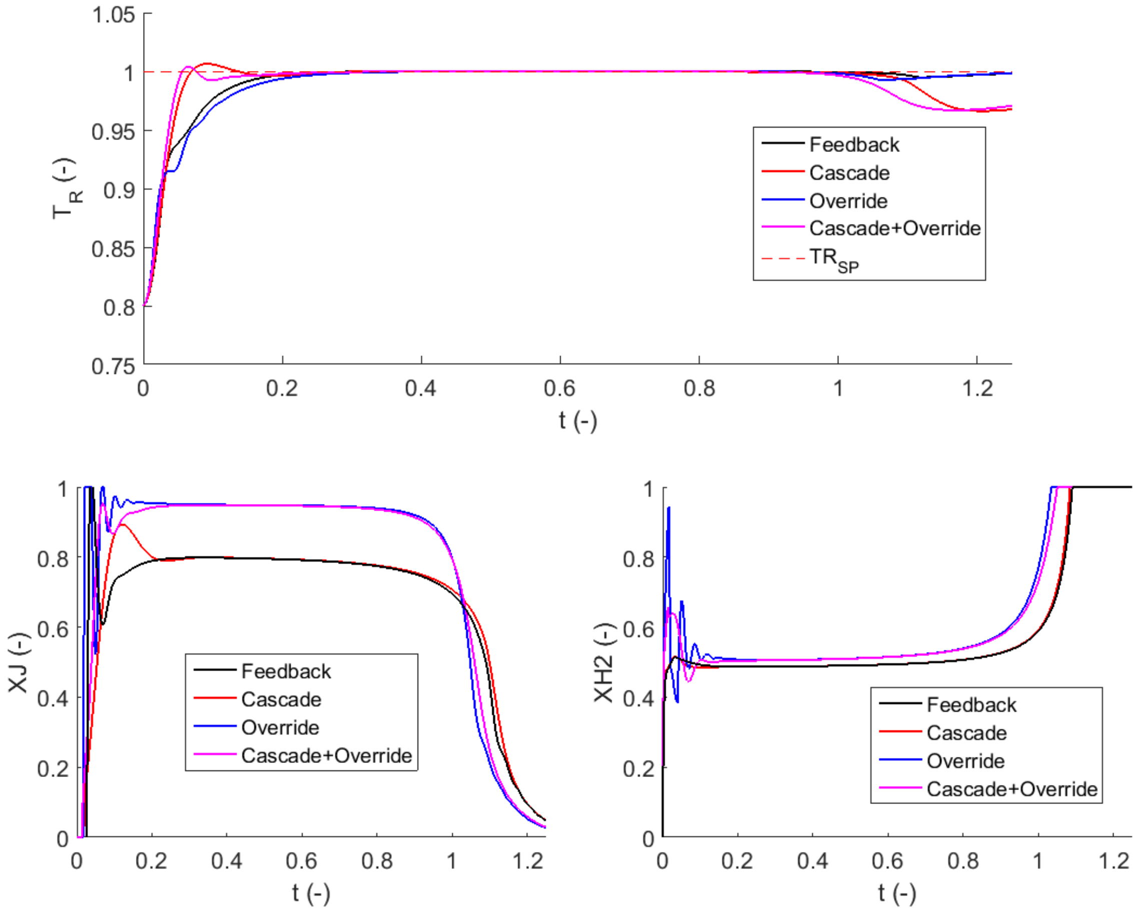
It can be seen that the results obtained when comparing the controllers under different initial mass fraction of A are very similar to the ones for standard operation. According to the error criteria defined, the cascade+override control presents a better temperature tracking than other investigated controllers. The cascade controller also performed satisfactorily. Figure 37 and Figure 38 show the simulated temperature profiles and manipulated variables behavior. It is important to highlight that the temperature drop at the end of the reaction is a natural phenomenon and is not significant for the process. Therefore, as explained in section 4.4, was not considered as a problem during the controllers' design.

Figure 37 – Control strategies response under lower initial mass fraction of A $(0.8 * x_{A0})$



Source: Author

Figure 38 – Control strategies response under higher initial mass fraction of A $(1.2 * x_{A0})$



Source: Author

Table 12 shows the IAE, ISE and ITAE values calculated for the temperature responses of the four control strategies under non-standard operational conditions.

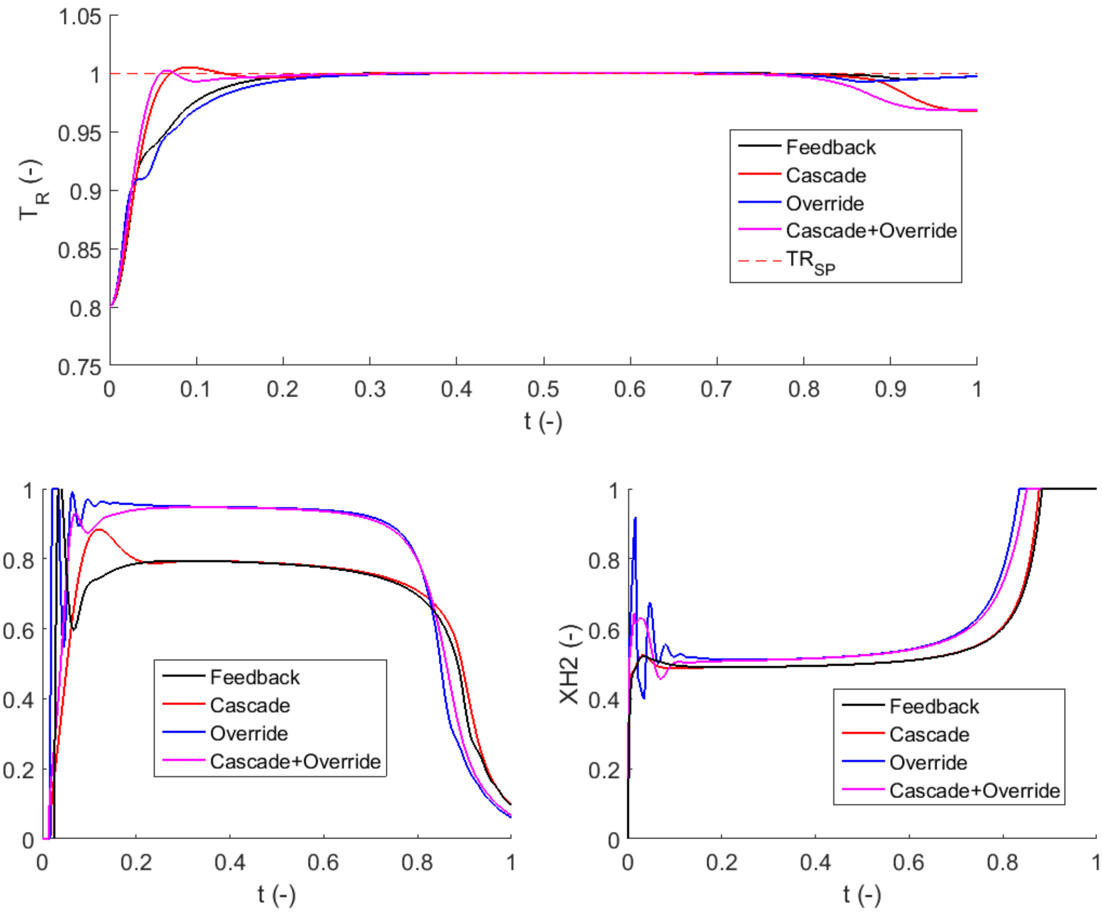
Table 12 – Comparison of IAE, ISE and ITAE values for temperature response under different kinetic factor ($\boldsymbol{f_{kat}}$)

Controller Strategy	**IAE**		**ISE**		**ITAE**	
	$0.8\ f_{kat}$	$1.2\ f_{kat}$	$0.8\ f_{kat}$	$1.2\ f_{kat}$	$0.8\ f_{kat}$	$1.2\ f_{kat}$
Feedback	7.61	7.60	80.83	77.39	2.88	2.91
Cascade	5.37	5.06	68.65	61.06	1.54	1.47
Override	8.45	8.44	80.10	78.69	4.00	3.98
Cascade + Override	**4.84**	**4.78**	**60.50**	**53.80**	**1.36**	**1.45**

Source: Author

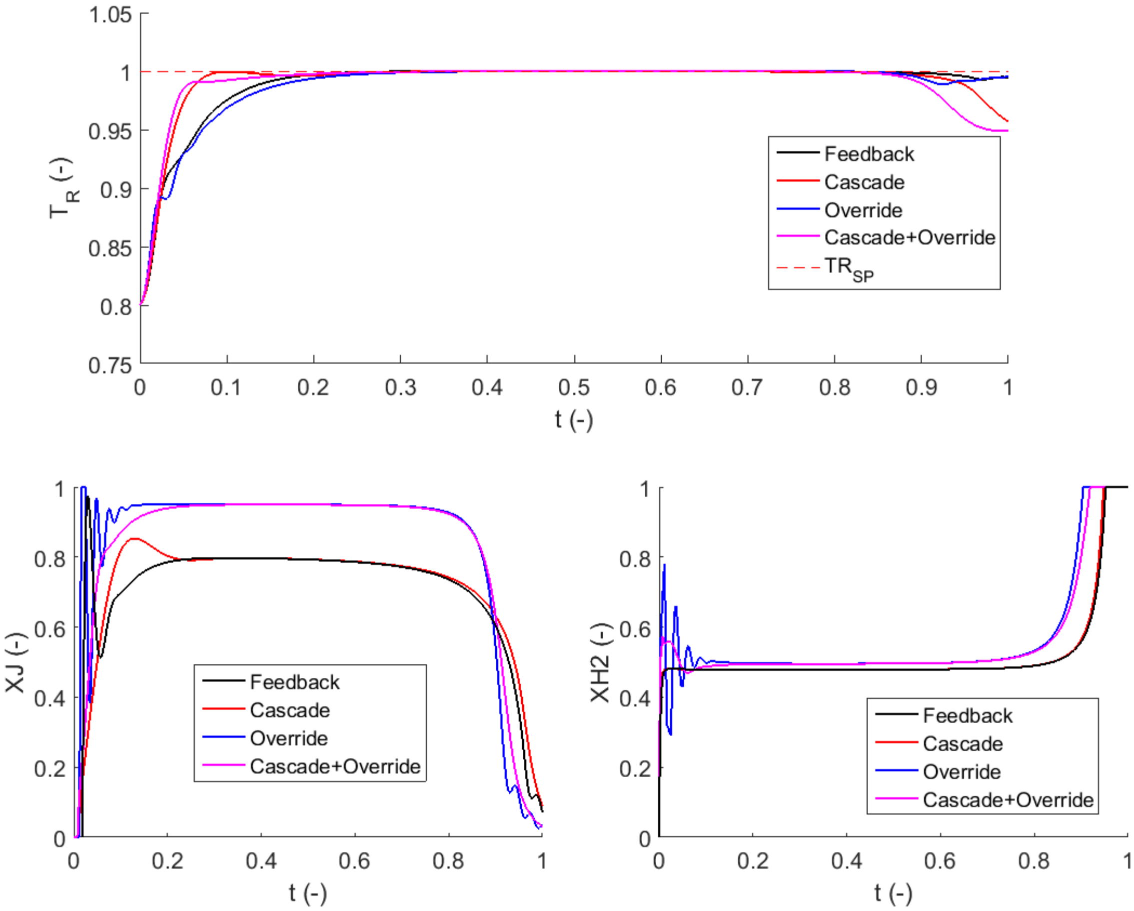
Again, the results obtained for different kinetic factors are similar to the ones for standard operation. The cascade+override control showed a better temperature tracking than the other investigated controllers. The cascade controller also showed a satisfactorily performance. Figure 39 and Figure 40 show the simulated temperature profiles and manipulated variables behavior.

Figure 39 – Control strategies response under lower kinetic factor (f_{kat})



Source: Author

Figure 40 – Control strategies response under higher kinetic factor (f_{kat})



Source: Author

For the non-standard conditions and regarding the criteria values analyzed, the cascade and cascade+overide controllers were the best choices to control the reactor temperature. Nevertheless, the controller's performance was also evaluated under disturbances during the batch.

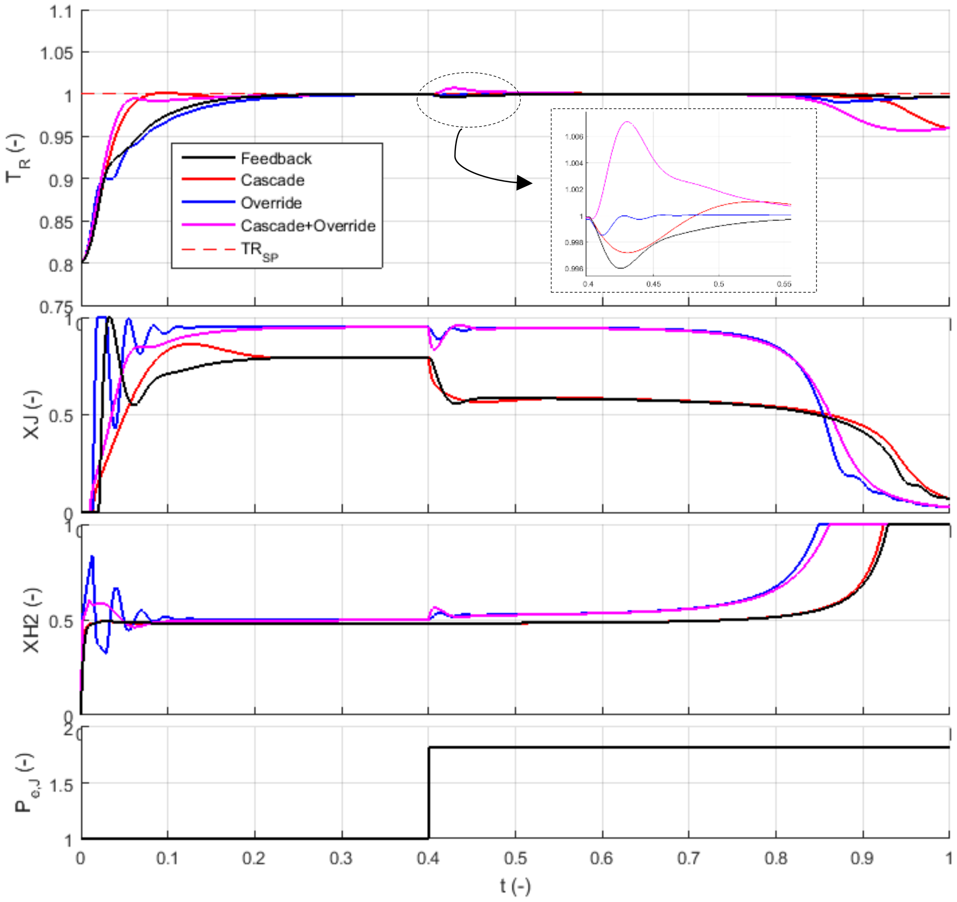
4.7.1.3 Assessment of controller performance under disturbances

As mentioned in the previous sections, different disturbances can affect the process under study. In the industrial plant, the cooling fluid inlet pressure and hydrogen inlet pressure can oscillate during batch. These disturbances can be represented as step and pulse variations in the model input conditions. In that context, the controller's performance was evaluated against disturbances on the cooling fluid inlet pressure and hydrogen inlet pressure. The cooling fluid inlet temperature also fluctuates during the process, however, it doesn't have a standard behavior since it depends mainly on weather conditions and, hence, would not be feasible to simulated a realistic behavior of this disturbance.

Therefore, two step and two pulse disturbances were applied to the cooling fluid inlet pressure and hydrogen inlet pressure. The pulse disturbances represent short changes in the input conditions while step disturbances represent continuous changes. These continuous and short disturbances are both observed in the real plant and the magnitude of them were determined based on plant historic data.

Figure 41 shows a step disturbance of magnitude 170% of cooling fluid inlet nominal pressure at t = 0.4. The feedback, cascade, and override strategies were able to handle the disturbance since the temperature oscillated less than ±0.5% in these cases. For the feedback and cascade controllers, the cooling fluid valve has a similar behavior: after the disturbance, the valve closes to compensate the cooling fluid pressure increase, and consequently, to reduce cooling fluid flowrate to reactor jacket in order to keep T_R at its setpoint. Regarding the hydrogen valve, since it is controlled by an independent flow control, its position is not affected by the disturbance. On the other hand, since the override frameworks includes a valve position control, the controller actuates to maintain the cooling fluid valve at a maximum and constant position and actuates opening the hydrogen valve to increase the reaction rate. Particularly, the cascade+override control actuates too fast causing a bigger oscillation at the hydrogen valve which entails higher oscillations in temperature.

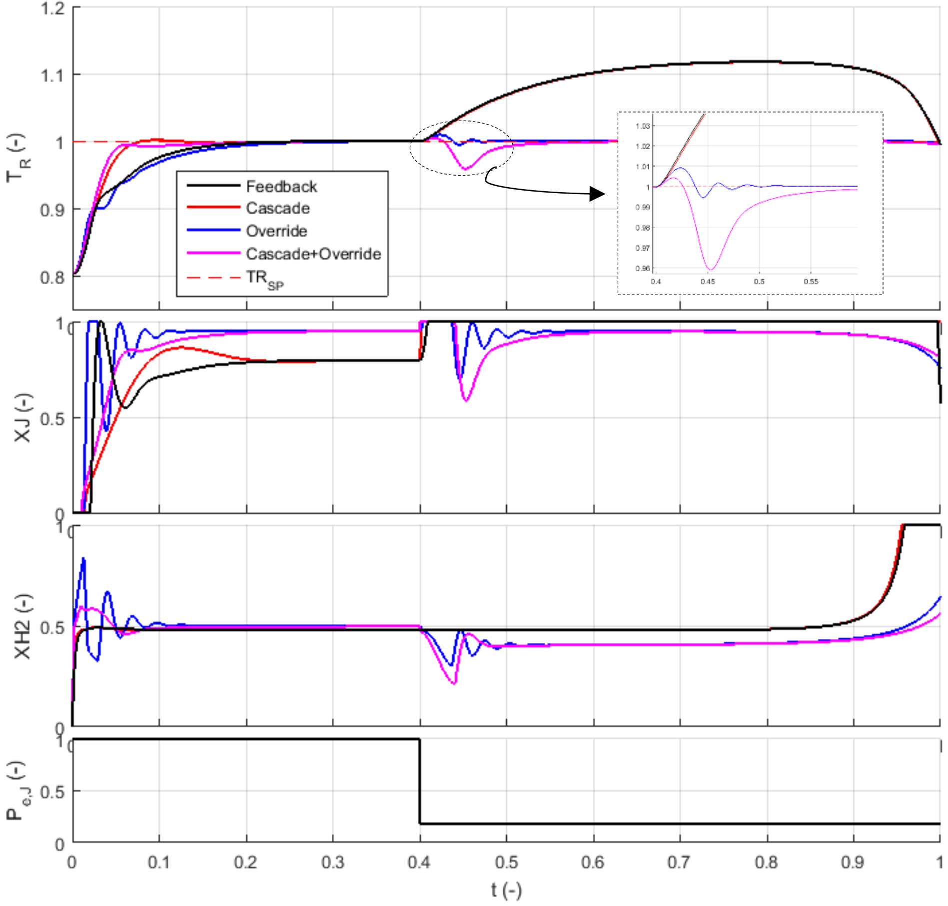
Figure 41 – Step disturbance at cooling fluid inlet pressure (P_{eJ}) – t = 0.4 and 170% of cooling fluid inlet nominal pressure



Source: Author

A 30% magnitude step disturbance on the cooling fluid inlet nominal pressure at t = 0.4 was also simulated, as can be seen in Figure 42. The feedback and cascade controllers were not able to handle satisfactorily the disturbance: after the disturbance, the cooling fluid valve opens to compensate the pressure reduction and guarantee a minimum cooling fluid flow to remove the reaction heat. However, as one can see in X_J graph, for these controllers, the controller's output saturated (100% of X_J) after the disturbance indicating that the cooling capacity of the system was lower than the released reaction heat. With the valve position control (VPC), embed in the override frameworks, the aforementioned problem was overcome since these controllers can indirectly measure the cooling capacity through the monitoring of the cooling fluid valve position. When the cooling fluid valve opening is above its set point, the controllers close the hydrogen feed valve, as it can be seen in the X_{H2} graph. The cascade+override actuates faster than the override causing temperature oscillations higher than 4%, which can cause quality problems.

Figure 42 – Step disturbance at cooling fluid inlet pressure (P_{eJ}) – t = 0.4 and 30% of cooling fluid inlet nominal pressure



Source: Author

Table 13 summarizes the controllers' performance through IAE, ISE and ITAE criteria.

Table 13 – Comparison of IAE, ISE and ITAE values for reactor temperature response under step disturbances at cooling fluid inlet pressure

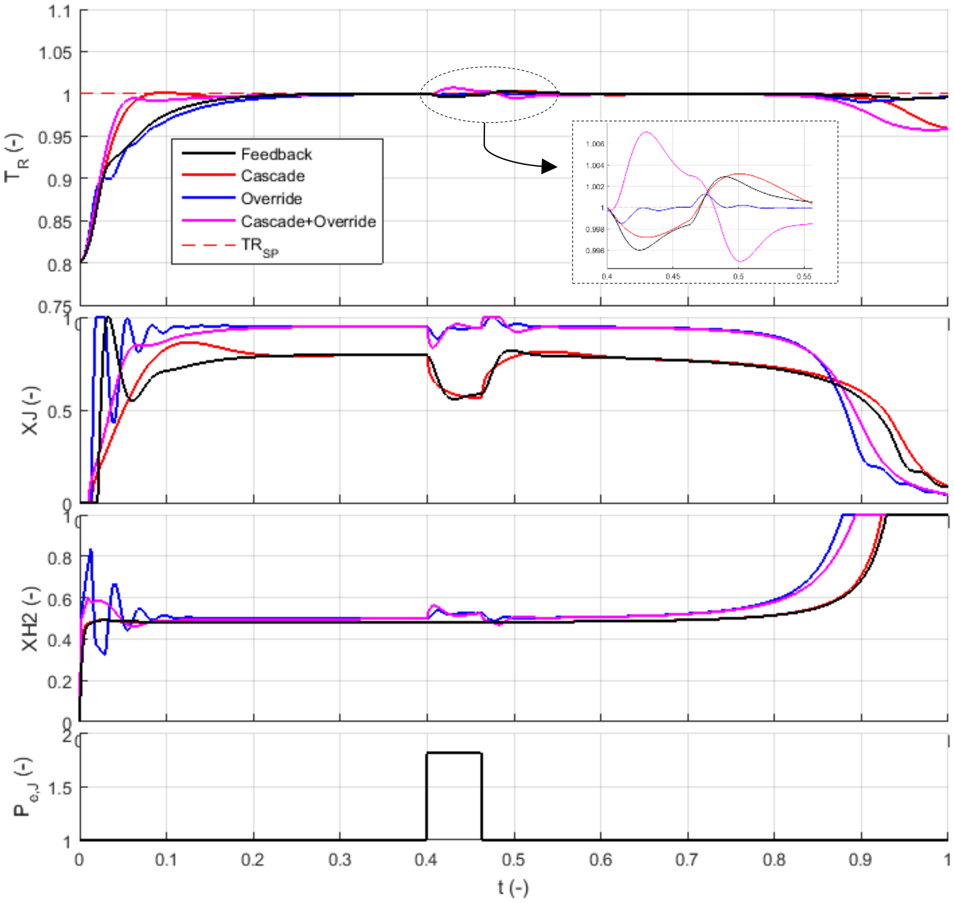
Controller Strategy	**IAE**		**ISE**		**ITAE**	
	170% of cooling fluid inlet pressure step disturbance	30% of cooling fluid inlet pressure step disturbance	170% of cooling fluid inlet pressure step disturbance	30% of cooling fluid inlet pressure step disturbance	170% of cooling fluid inlet pressure step disturbance	30% of cooling fluid inlet pressure step disturbance
Feedback	0.219	18.93	0.046	174.2	0.814	88.9
Cascade	0.218	18.65	0.030	170.8	0.839	87.6
Override	**0.073**	**0.301**	**0.003**	**0.136**	**0.250**	**1.058**
Cascade + Override	0.390	1.597	0.141	3.752	1.449	5.984

Source: Author

As one can see, the override control strategy presents the lowest values of all criteria, which indicates a better performance facing step disturbances at cooling fluid inlet pressure.

Similar results were obtained for pulse disturbances. Figure 43 shows a pulse disturbance of magnitude 170% of cooling fluid inlet nominal pressure at t = 0.4. The controllers' performance against the pulse disturbances was worse compared to the step disturbances since there are two changes at the cooling fluid inlet pressure in a relatively short period of time. The feedback, cascade, and override strategies were able to handle this disturbance since the temperature oscillation was lower than ±0.4% in all these cases. The cascade+override controller actuated too fast in the hydrogen valve, causing oscillation in temperature higher than 0.6%, which can cause quality problems.

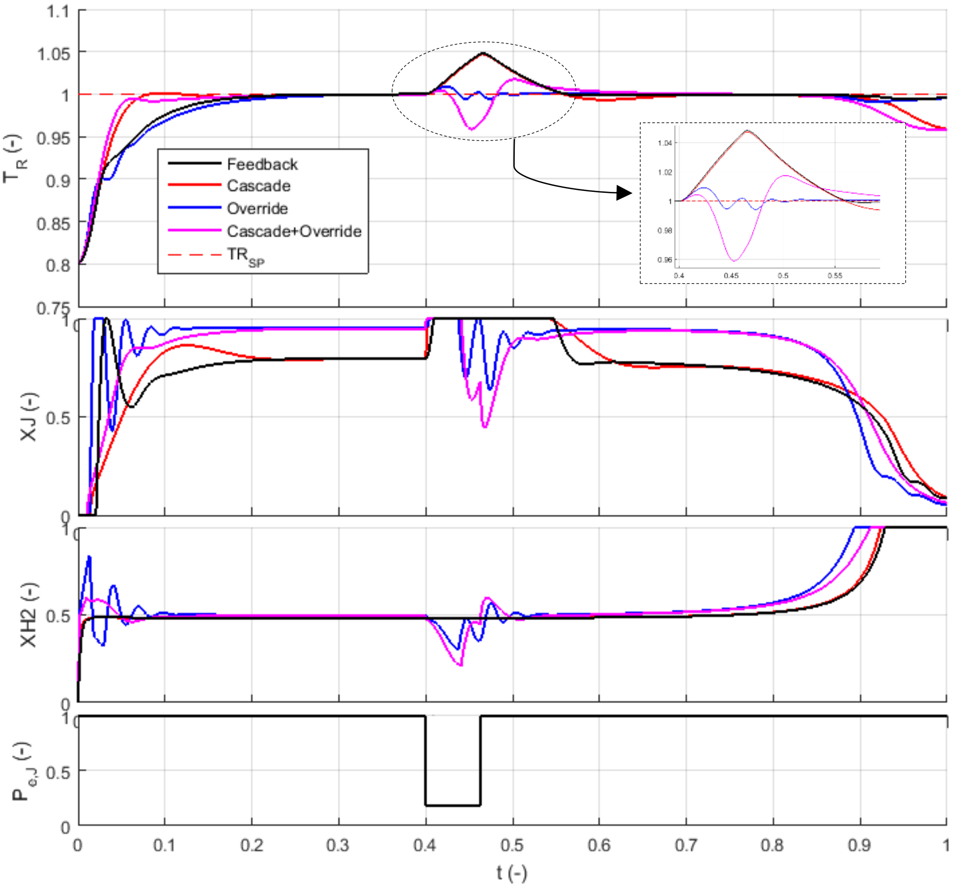
Figure 43 – Pulse disturbance at cooling fluid inlet pressure (P_{eJ}) – t = 0.4 and 170% of cooling fluid inlet nominal pressure



Source: Author

Figure 44 shows a pulse disturbance of magnitude 30% of cooling fluid inlet nominal pressure at t = 0.4. The feedback and cascade strategies were not able to satisfactorily control the reactor temperature against the disturbance. As one can see in X_J graph, for these controllers, the controller output saturated (100% of X_J) after the disturbance indicating that the cooling capacity of the system was lower than the released reaction heat. On the other hand, similarly to the case represented by Figure 42, the valve position control (VPC) embed in the override frameworks are able to deal with the lack of cooling capacity, since they actuate reducing hydrogen flowrate. The cascade+override controller actuates too aggressively in the hydrogen valve, causing oscillation in temperature higher than 4%, which can cause quality problems.

Figure 44 – Pulse disturbance at cooling fluid inlet pressure (P_{eJ}) – t = 0.4 and 30% of cooling fluid inlet nominal pressure



Source: Author

Table 14 summarizes the controllers' performance through IAE, ISE and ITAE criteria.

Table 14 – Comparison of IAE, ISE and ITAE values for reactor temperature response under pulse disturbances at cooling fluid inlet pressure

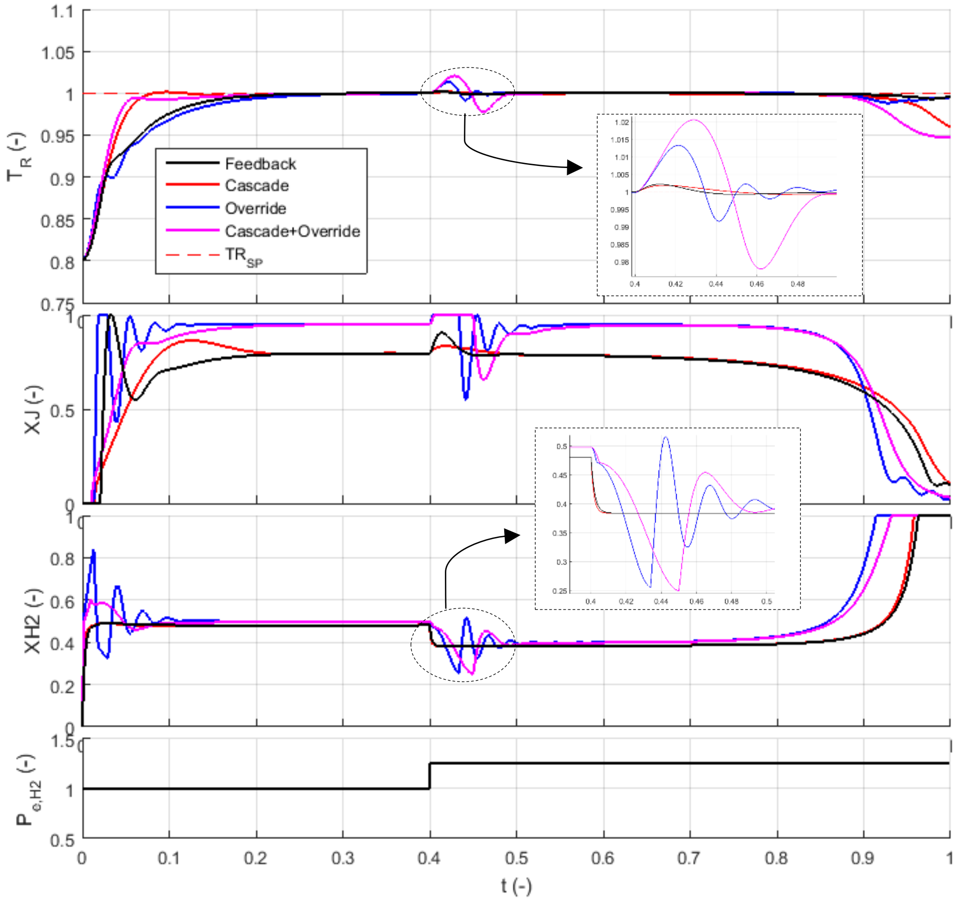
Controller Strategy	**IAE**		**ISE**		**ITAE**	
	170% of cooling fluid inlet pressure pulse disturbance	30% of cooling fluid inlet pressure pulse disturbance	170% of cooling fluid inlet pressure pulse disturbance	30% of cooling fluid inlet pressure pulse disturbance	170% of cooling fluid inlet pressure pulse disturbance	30% of cooling fluid inlet pressure pulse disturbance
Feedback	0.283	2.993	0.062	9.572	1.087	11.38
Cascade	0.360	3.321	0.066	9.266	1.471	13.12
Override	**0.092**	**0.387**	**0.005**	**0.163**	**0.327**	**1.417**
Cascade + Override	0.525	2.069	0.190	4.226	2.057	8.150

Source: Author

The override control strategy presents the lowest values of all criteria, which indicates a better performance facing pulse disturbances at cooling fluid inlet pressure.

In addition to the simulation for the disturbances performed on the cooling fluid inlet pressure, disturbances in the hydrogen inlet pressure were also studied. Figure 45 shows a step disturbance of magnitude 125% of hydrogen inlet nominal pressure at t = 0.4. In this case, the feedback and cascade controllers showed better performance since the temperature errors were less than ±0.5% whereas the override and cascade+override performed with errors greater than 1%. This disturbance affects directly the hydrogen feed rate and, as a consequence, the reaction heat and reactor temperature. The feedback and cascade strategies have an independent hydrogen flowrate control and a temperature controller which actuates to open the cooling fluid valve. On the other hand, the override and cascade+override strategies include a valve position control which actuates to maximize the cooling fluid valve opening and reduces the hydrogen valve opening to reduce the reaction rate. It can be seen that in the override and cascade+override controllers the hydrogen valve response is slower and more oscillating. This fact is related to the tuning challenges already discussed in previous sections.

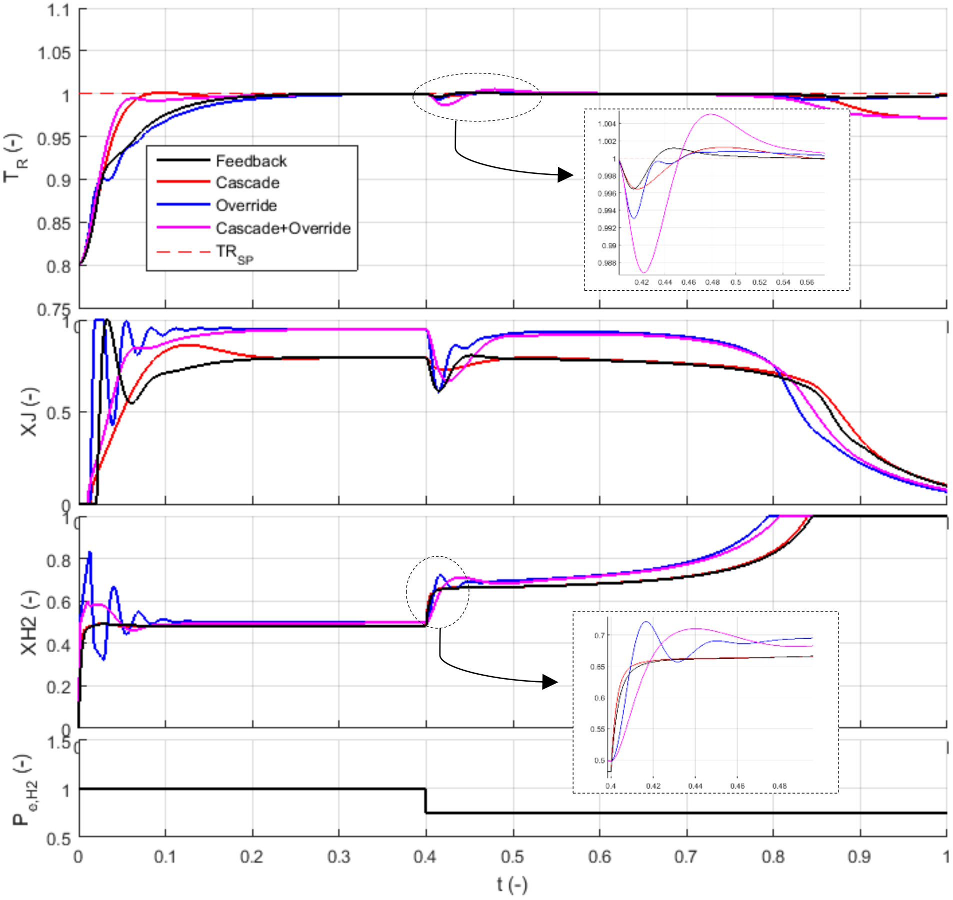
Figure 45 – Step disturbance at hydrogen inlet pressure (P_{e,H_2}) – t = 0.4 and 125% of hydrogen inlet nominal pressure



Source: Author

Figure 46 shows a step disturbance of magnitude 75% of hydrogen inlet nominal pressure at t = 0.4. The feedback and cascade strategies were able to handle this disturbance since the temperature oscillations were less than ±0.5%. The valve position control embedded on the override and cascade+override frameworks actuated to maximize the cooling fluid valve position by increasing the opening of the hydrogen valve. However, the hydrogen valve response was slower and more oscillating due to tuning challenges discussed in previous sections. This slow and oscillating response result on greater temperature errors compared to the feedback and cascade strategies.

Figure 46 – Step disturbance at hydrogen inlet pressure (P_{e,H_2}) – t = 0.4 and 75% of hydrogen inlet nominal pressure



Source: Author

Table 15 summarizes the controllers' performance through IAE, ISE and ITAE criteria.

Table 15 – Comparison of IAE, ISE and ITAE values for reactor temperature response under step disturbances at hydrogen inlet pressure

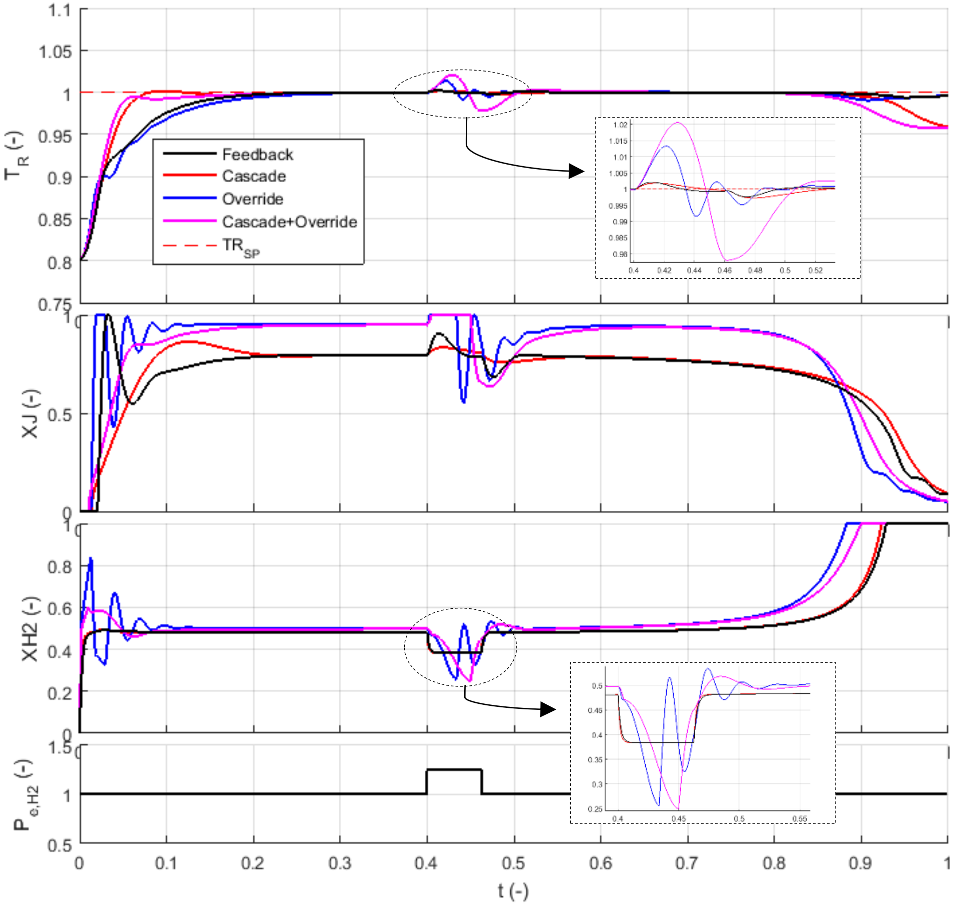
Controller Strategy	**IAE**		**ISE**		**ITAE**	
	125% of hydrogen inlet pressure step disturbance	75% of hydrogen inlet pressure step disturbance	125% of hydrogen inlet pressure step disturbance	75% of hydrogen inlet pressure step disturbance	125% of hydrogen inlet pressure step disturbance	75% of hydrogen inlet pressure step disturbance
Feedback	**0.094**	**0.116**	**0.007**	**0.017**	**0.355**	**0.428**
Cascade	0.152	0.216	0.011	0.031	0.596	0.838
Override	0.396	0.216	0.264	0.053	1.396	0.784
Cascade + Override	0.942	0.611	1.408	0.397	3.389	2.265

Source: Author

As one can see, the feedback control strategy presents the lowest values for all criteria, which indicates a better performance facing step disturbances at hydrogen inlet pressure. Although the cascade performance was not the best for this case, the observed control response and temperature errors were satisfactory.

A similar approach was performed for pulse disturbances. Figure 47 shows a pulse disturbance of magnitude 125% of hydrogen inlet nominal pressure at t = 0.4. The feedback and cascade controllers performed satisfactorily since the temperature errors were less than ±0.5% whereas the override and cascade+override performed with errors greater than 2%. It can be seen that in the override and cascade+override controllers the hydrogen valve response is slower and more oscillating. This fact is related to the tuning challenges already discussed in previous sections.

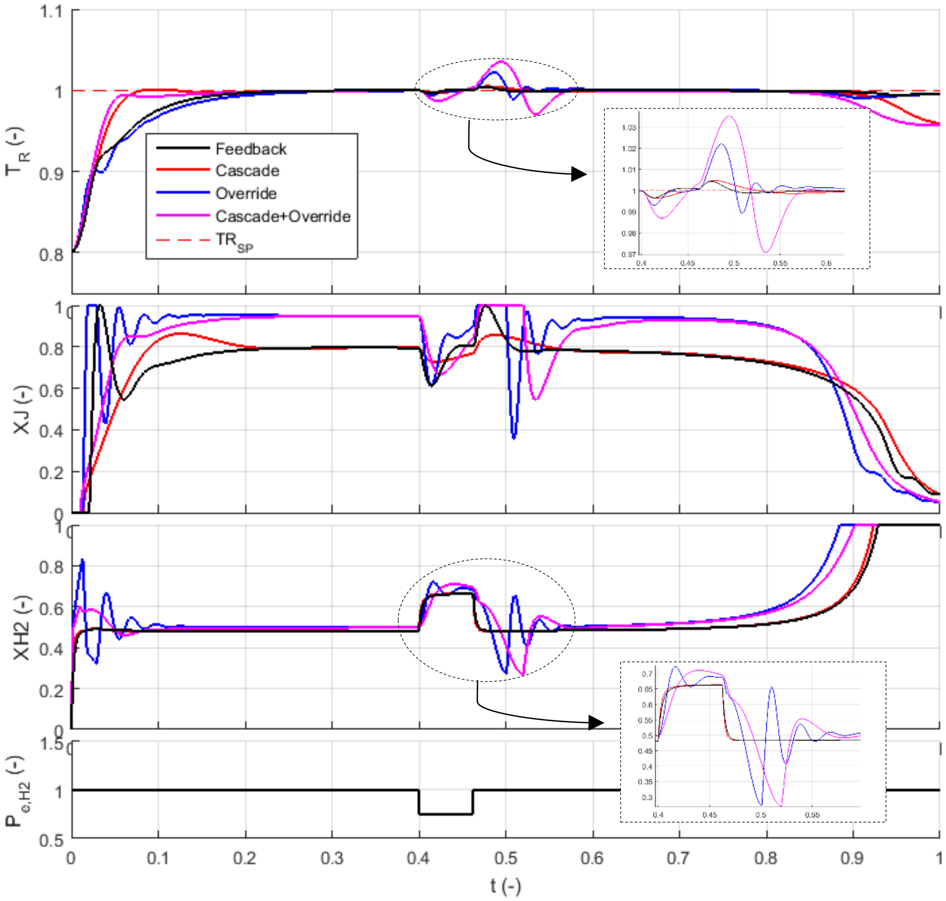
Figure 47 – Pulse disturbance at hydrogen inlet pressure (P_{e,H_2}) – t = 0.4 and 125% of hydrogen inlet nominal pressure



Source: Author

Finally, Figure 48 shows a pulse disturbance of magnitude 75% of nominal pressure at t = 0.4. The feedback and cascade strategies were able to handle this disturbance since the temperature oscillation was less than ±0.5% whereas the override and cascade+override performed with errors greater than 3%. It can be seen that in the override and cascade+override controllers the hydrogen valve response is slower and more oscillating. This fact is related to the tuning challenges already discussed in previous sections.

Figure 48 – Pulse disturbance at hydrogen inlet pressure (P_{e,H_2}) – t = 0.4 and 75% of nominal pressure



Source: Author

Table 16 summarizes the controllers' performance through IAE, ISE and ITAE criteria.

Table 16 – Comparison of IAE, ISE and ITAE values for reactor temperature response under pulse disturbances at hydrogen inlet pressure

Controller Strategy	**IAE**		**ISE**		**ITAE**	
	125% of hydrogen inlet pressure pulse disturbance	75% of hydrogen inlet pressure pulse disturbance	125% of hydrogen inlet pressure pulse disturbance	75% of hydrogen inlet pressure pulse disturbance	125% of hydrogen inlet pressure pulse disturbance	75% of hydrogen inlet pressure pulse disturbance
Feedback	**0.120**	**0.225**	**0.014**	**0.044**	**0.450**	**0.870**
Cascade	0.190	0.348	0.025	0.080	0.744	1.382
Override	0.468	0.796	0.284	0.837	1.705	3.146
Cascade + Override	1.231	2.052	1.796	4.465	4.563	8.193

Source: Author

As one can see, the feedback control strategy presents the lowest values for all criteria, which indicates a better performance facing step disturbances at hydrogen inlet pressure. Although the cascade performance was not the best for this case, the observed control response and the temperature errors were satisfactory for this application.

Finally, after all tests performed, the discussed pros and cons for the techniques evaluated regarding temperature control are summarized in Table 17.

Table 17 – Pros and cons of feedback, cascade, override and cascade+override control techniques regarding temperature control

Controller Strategy	Pros	Cons
Feedback	• Easier to implement • Easier to tune • More robust against hydrogen feed disturbances	• Not able to handle saturation of cooling capacity
Cascade	• Easier to implement • Faster control responses compared to feedback • Achieved satisfactory results in many cases	• Not able to handle saturation of cooling capacity
Override	• More robust against cooling fluid disturbances • Can handle cooling capacity variations • Faster control responses compared to feedback	• Harder to tune • Higher oscillation in the beginning
Cascade + Override	• Reaches setpoint faster • Lower error in normal operation and under different operational conditions • Can handle cooling capacity variations	• Harder to tune • Higher oscillation against disturbances

Source: Author

Regarding temperature control, the most relevant aspects were discussed. Nevertheless, the influence of the controller on batch time and quality are also important when evaluating the controllers' performance.

4.7.1.4 Batch time and quality parameters performance

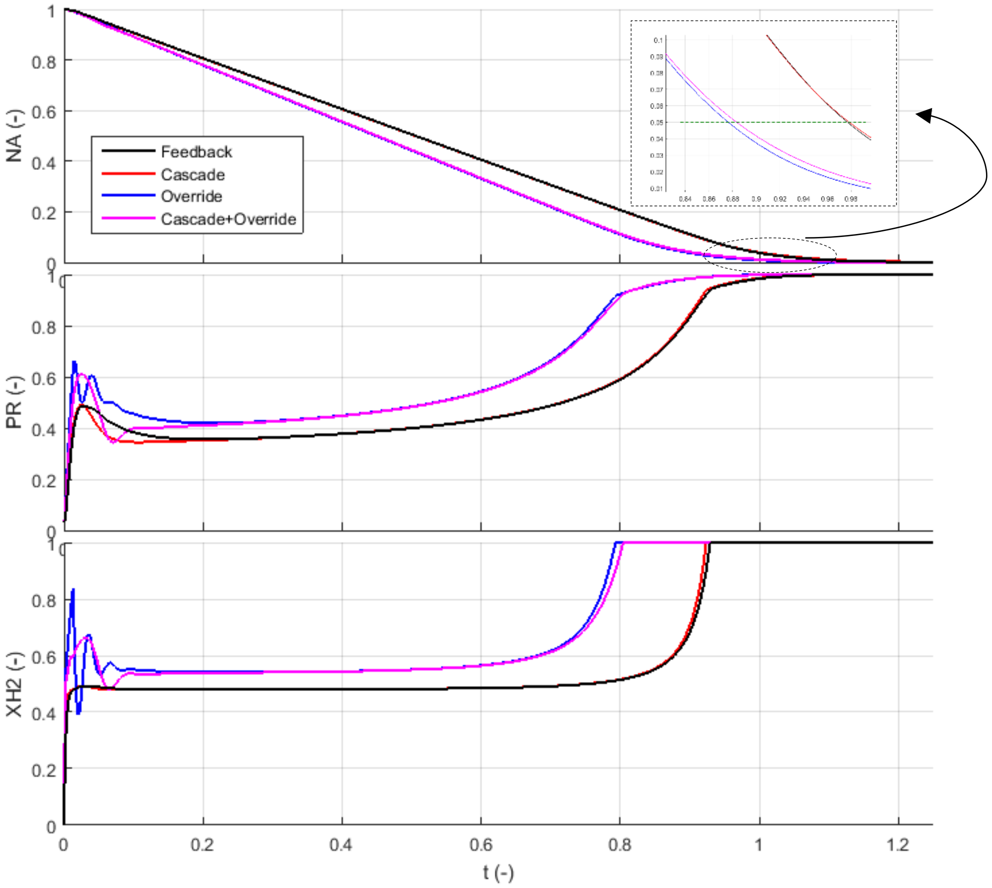
Batch time and product quality are fundamental parameters for batch processes. Therefore, the influence of the temperature control strategy on these parameters was evaluated.

The batch time can be defined as the time to get a consumption of 99.5% of $\mathrm{N_A}$. Regarding quality, for this specific process, it can be established direct correlation of product quality with reactor pressure which means that oscillations in the reactor pressure can cause side reactions and quality issues.

To explore the controller’s potential, this evaluation was done considering a hypothetical full cooling capacity availability. In other words, it was considered a cooling fluid flowrate sufficiently high and a cooling fluid temperature sufficiently low to avoid saturation of control actions.

Figure 49 shows the behavior of the number of mols of A, reactor pressure and hydrogen valve opening under normal operation and enough cooling capacity.

Figure 49 – $N_A, P_R \text{ and } X_{H_2}$ under normal operation – Batch time and quality analyses

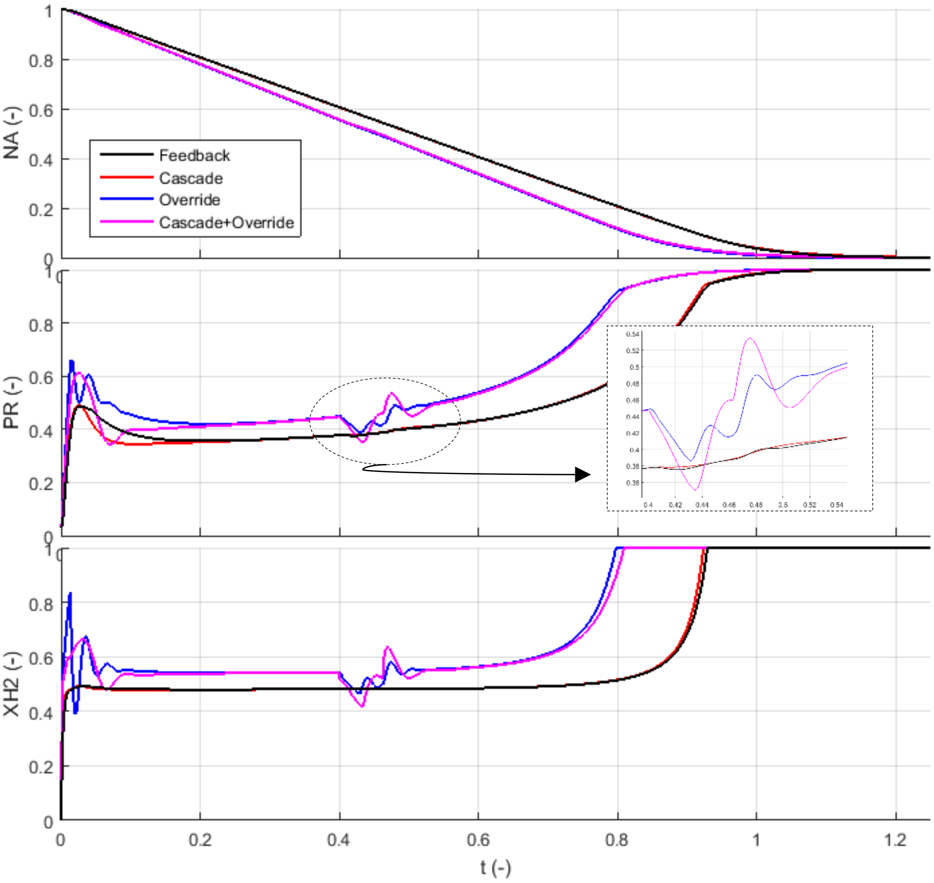


Source: Author

As one can see, the override and cascade+override controllers were able to reduce the batch time by almost 20%. Due to the valve position controller embed to their frameworks, the hydrogen flow rate can be maximized when there is cooling capacity available. However, the override strategy also entailed pressure oscillation during the batch. This could negatively impact the quality results. In the end, the cascade+override controller seems to be better since reduces the batch time and causes just a small oscillation in the pressure at the beginning of the batch.

Figure 50 shows the behavior of the same variables, but against a step disturbance at cooling fluid inlet pressure normal operation and enough cooling capacity.

Figure 50 – $N_A, P_R \; and \; X_{H_2}$ under a step disturbance at cooling fluid inlet pressure – Batch time and quality analyses



Source: Author

As expected, the override and cascade+override reduced the batch time, however, the disturbance caused oscillation in the response of these controllers. As explained above, the oscillation at pressure can negatively affect the quality of the final product. The feedback and cascade controllers' responses were more stable and showed less oscillations against process disturbances.

5 CONCLUSION

The present study developed a mathematical model to describe a semi-batch hydrogenation reactor of a real industrial plant. The model was validated through comparison with the plant real data and was able to satisfactorily represent the real process. For the reactor temperature and pressure simulations, the errors between the model and the real data were below 2% and 10%, respectively.

The process dynamics were discussed via model sensitivity analysis and three different temperature control strategies were proposed in order to evaluate alternatives for the plant current control approach.

The sensitivity analyses showed to be very helpful to understand the process dynamics contributing to the development of the proposed control techniques and enhancing the process knowledge of plant personnel.

During the development of the proposed control techniques, the lack of tuning methods for batch and semi-batch (no steady-state) processes represented significant challenges on their implementation. In the case of the override frameworks, the valve position controller introduced oscillations and difficulties to define the tuning parameters. For real application of the proposed strategies, a fine-tuning should be performed at the plant either by experienced personnel and/or DCS vendor autotuning tools.

The use of an anti-reset windup method to avoid controller's inadequate responses was fundamental for the controllers' satisfactory performance observed in the simulations. It is known that the majority of DCS vendors already provide anti-reset windup algorithms embed to their controllers, however, it is fundamental to emphasize that this fact should also be taken into account for academic implementation.

The proposed control strategies were compared under different conditions. The current feedback controller showed good results under hydrogen feed disturbances and caused less pressure oscillation. However, the controller saturated with high disturbances at cooling fluid leading to loss of temperature control. On the other hand, the overrides frameworks demonstrated very good results when subjected to cooling fluid disturbances and can reduce the batch time if the system cooling fluid capacity is improved. Nevertheless, they caused much more oscillation at hydrogen valve, impacting the reactor pressure during normal operation and in case of disturbances.

Finally, it was interesting to observe that even a simple technique, as the cascade strategy, brought satisfactory results regarding temperature control and product quality for the majority of the cases. Although is not able to reduce batch time or detect cooling capacity saturation, it is the best control strategy for this specific case study considering the disturbance and operational ranges assessed since it showed fast and stable responses against disturbances that will not jeopardize quality.

The lack of cooling capacity introduces a limitation to the feedback and cascade controllers that is not related to the controller itself. For higher magnitude disturbance values, the system is limited by the available cooling capacity. In that manner, ordinary control strategies such as feedback or cascade easily saturated while more advanced techniques as override frameworks could handle this problem by controlling the hydrogen feed when necessary. On the other hand, the valve position controller embedded to the override controllers introduced oscillation at hydrogen feed leading to unwanted collateral effects, such as temperature and pressure oscillation.

In sum, the definition of a control strategy strongly depends on the type of process since each process has its characteristics and needs. Factors, such as, but not limited to, process constraints, quality constraints, installation constraints, production bottleneck, cost-benefit of implementation, should be part of the assessment. It is important to evaluate also, how the new control strategy can interfere in other variables of the process to evaluate whether the unwanted collateral effects can be accepted or not. The change of a control strategy should be done with caution and in a tailor-made approach. In conclusion, the choice of the better controller technique will be always a tradeoff between pros and cons.

6 FUTURE WORKS PROPOSAL

During the development of this work other possibilities of studies were found as the following:

a) Use and investigation of uncertainty handling and parameter estimation tools to improve the fit between the model and real data;
b) Use of the model to develop operational training;
c) Use of the model to perform safety analysis;
d) Comparison with other controller techniques such as:
    - Feedforward controller: this controller could help the prediction of disturbances and improve performance;
    - Nonlinear controller: the process has inherent non-linear characteristics, therefore would be interesting to investigate how non-linear controller would better address this fact;
    - Adaptative and self-tuning: as mention before, some authors recommend these techniques to get better results during heat-up and reaction phase in batch and semi-batch reactor;
    - Nonlinear model predictive control: literature has shown many applications of NMPC to temperature control in batch and semi-batch reactors;
    - Neural networks: literature has shown many applications of neural networks to temperature control in batch and semi-batch reactors;
e) Comparison of other anti-reset windup methods since this phenomenon strongly affects the performance of the controller;
f) Evaluate the performance of the controllers at different moments in time since the process is operated in semi-batch mode, different moments can show different behaviors;
g) Use of other performance measures to compare the different control techniques, such as, rise time, settling time, maximum overshoot, manipulation effort etc;

h) Comparison and investigation of tuning methods since there is a lack of tuning tools when there is no steady-state, typical characteristic of batch and semi-batch processes;
i) Implementation of soft sensors to evaluate quality issues;

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