Fault detection and isolation for industrial risk prevention

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ABSTRACT. *The main tool for the development of hazardous chemical syntheses remains the batch reactor. Nevertheless, even if it offers the required flexibility and versatility, this reactor presents technological limitations. In particular, poor transfer of the heat generated by exothermic chemical reactions is a serious problem with regard to safety. In this context, a simple failure is considered as prejudicial. So, fault detection and diagnosis are studied with a particular attention in the scientific and industrial community. This work presents a fault detection and isolation methodology. The developed methodology rests on a mixed approach which combines a model-based method for the fault detection and a pattern matching approach for the identification. It is integrated within a hybrid dynamic simulator. In this paper, the approach is tested during the operation of an exothermic reaction.*

RÉSUMÉ. *L'outil principal pour le développement de synthèses chimiques dangereuses demeure le réacteur discontinu. Malgré sa flexibilité et sa polyvalence, ce réacteur présente des limitations technologiques, notamment son manque de performance en transfert de la chaleur. Dans le cas d'une réaction exothermique, le risque d'emballement thermique est à craindre et une simple défaillance est alors considérée néfaste. Ainsi, la détection et de diagnostic sont étudiés avec une attention particulière dans la communauté scientifique et industrielle. La méthodologie développée dans ce travail repose sur une approche mixte qui combine une méthode à base de modèles pour la détection de défaut et une approche de reconnaissance de formes pour l'identification. Il est intégré dans un simulateur dynamique hybride. Dans cet article, l'approche est testée lors de l'opération d'une réaction exothermique.*

KEYWORDS: *fault detection and isolation, extended Kalman filter, dynamic hybrid simulation, model based method, risk assessment.*

MOTS-CLÉS : *détection et diagnostic de fautes, filtre de Kalman étendu, simulation dynamique hybride, méthode à base de modèle, analyse des risques.*

1. Introduction

Batch and semi-batch processes play an important role in the pharmaceutical and fine chemical process industry. They are the prevalent production modes for low volumes of high added value products. Such systems are characterized not only by a small scale flexible production, but also by complex chemical reaction (Grau *et al.,* 2000). Generally, these processes are complex and not entirely known. For this reason, batch or fed-batch reactors are frequently used for the development of hazardous chemical synthesis. Even if it offers the required flexibility and versatility, this reactor has technological limitations. Particularly, during the exothermic chemical reactions, the poor heat transfer can become a serious safety problem (Stoessel, 2008).

Numerous methods of risk analysis such as HAZOP (International standard IEC 61882) can be exploited for the identification of the main events or potentially dangerous deviations. Then, corrective measures can be taken in order to improve the process safety. However, these studies cannot prevent the occurrence of failures during the chemical reaction. For instance, these failures can be due to the malfunction of a sensor or of an actuator. There are also the structural changes, which refer to changes in the process itself (Venkatasubramanian *et al.,* 2003). These failures occur due to hard failures in equipment, such as a stuck valve, a broken or leaking pipe.

Consequently, fault detection and diagnosis are the purpose of a particular attention in the scientific and industrial community. The major idea is that the defect must not be undergone but must be controlled. Nowadays, these subjects remain a large research field. The literature quotes as many fault detection and diagnosis methods as many domains of application (Chang and Chen, 2011; Venkatasubramanian *et al.,* 2003). A notable number of works has been devoted to fault detection and isolation, and the techniques are generally classified as:

– methods without models such as quantitative process history based methods (neural networks (Venkatasubramanian *et al.,* 2003), statistical classifiers (Anderson, 1984)), or qualitative process history based methods (expert systems (Venkatasubramanian, et al., 2003)),

– model-based methods which are composed of quantitative model-based methods (such as analytical redundancy (Chow and Willsky, 1984), parity space (Gertler and Singer, 1990), state estimation (Willsky, 1976), or fault detection filter (Franck, 1990)) and qualitative model-based methods (such as causal methods: digraphs (Shih and Lee, 1995), or fault tree (Venkatasubramanian *et al.,* 2003)).

In this paper, the proposed approach to fault detection and isolation is a model-based approach, applied to chemical processes. This article is organized as follows. The first section focuses on the main fundamental concepts in the field of a batch reactor and of runaway scenarios. The second part presents the studied reaction. This is the oxidation of sodium thiosulfate with hydrogen peroxide. Then, its protocol is underlined. In section 3, illustrates the proposed model-based diagnosis approach. It uses the extended Kalman Filter, in order to generate a fault indicator. The high

robustness and real-time ability of observer is well-known for industrial applications (Ding, 2014). Next, its implementation is underlined and a general overview of the simulation environment and of the modelling within PrODHyS. Then, this approach is enlightened exploited through the simulation of an exothermic reaction monitoring. A fault in the functioning of the energy system is introduced at an unknown moment. The energy system does not provide enough heat quantity. Finally, Section 5 summarizes the contributions and achievements of the paper and some future research works are suggested.

2. Exothermic chemical reaction

The implementation of chemical reactions in batch or semi-batch reactors is strongly limited by the constraints linked to the dissipation of the heat generated by the reactions. Thus, this implementation can be made only after a complete risk analysis has been conducted to guarantee its safety, and the quality of its products (Chetouani et al., 2003). Consequently, we must firstly identify the risk in a chemical reactor. The main risk consists of a runaway scenario.
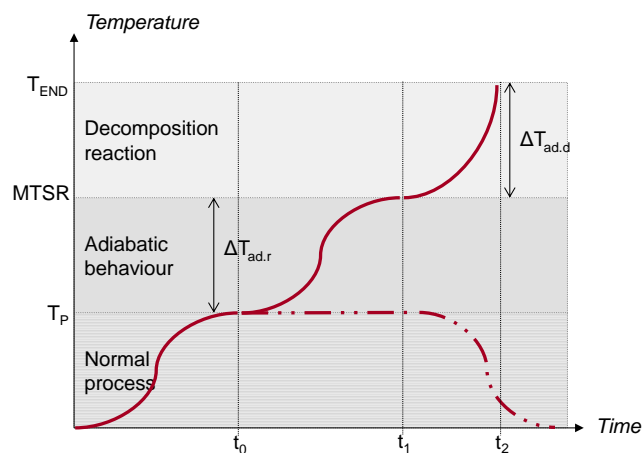
2.1. Runaway scenario

Figure 1. Runaway scenario

Gygax (1988) proposed the following runaway scenario. He considers the case where a complete cooling failure occurs (figure 1):

– The reactor is initially at the process temperature T_P.

– Next, at t=t0, the cooling stops and so the temperature increases due to the completion of the reaction. This temperature increase depends on the process conditions (Stoessel, 2008).

– A level called the Maximum Temperature of the Synthesis Reaction (MTSR) can be reached. At this temperature, a secondary decomposition reaction may be initiated, and the temperature of the mixture increases to reach the final temperature T_{END}.

The runaway reaction can be induced by an increase of the reaction rate (in other words, of the heat production) or by a decrease of the cooling capacity. A heat increase may be initiated either by an increase of the concentrations (causes: feeding, evaporation…), either by a catalytic acceleration (origins: reactive specificities, autocatalysis…), or by a temperature rise (reasons: reaction heat, energy adding).

A cooling capacity decrease may come from a decrease of the mixing efficiency (stopping of the stirring motor…), from a decrease of the heat transfer (causes: viscosity increase, etc.), from a quantity or potential increase (for example, due to the use of important quantities), or by a decrease of the power of the cooling system (origins: failures on the cooling system). The objective of this research work is so to prevent the reaction runaway by detecting the abnormal behaviour and diagnosing the causes of this behaviour.

2.2. Choice of the reaction

The chosen reaction is a very exothermic oxido-reduction one, the oxidation reaction of sodium thiosulfate $Na_2S_2O_3$ by hydrogen peroxide H_2O_2 (Chetouani, 2004). Because of its characteristics, this reaction is particularly exploited for safety study (Xaumier *et al.,* 2002; Chetouani *et al.,* 2003; Prat *et al.,* 2005; Benaissa *et al.,* 2008; Benkouider *et al.,* 2012). The stoechiometric scheme is:

$Na_2S_2O_3 + 2\ H_2O_2 \rightarrow$ ½ $Na_2S_3O_6 +$ ½ $Na_2SO_4 + 2\ H_2O$

This liquid homogeneous reaction is irreversible, fast, and highly exothermic (Lo and Cholette, 1972). The kinetics can be described by:

$$v = k \cdot [Na_2S_2O_3] \cdot [H_2O_2] \quad with\ k = k_0 \cdot \exp\left[\frac{-E_a}{R \cdot T}\right] \tag{1}$$

$k_0 = 2 \cdot 10^{10}\,\text{L.mol}^{-1}.\text{s}^{-1}$ and $E_a = 6.82 \cdot 10^4\,\text{J.mol}^{-1}$.

The reaction heat is ΔH_r = -586.2 kJ.mol^{-1} $[Na_2S_2O_3]$. These thermal characteristics underline the hazard of the reaction. This irreversible reaction is very fast and highly exothermic. A safety problem may occur.

For this work, some hypotheses are stated:

- The reactor is perfectly mixed with a homogeneous temperature in the reaction mixture;

- The feeding of the reactive does not induce volume contraction;

- The chemical reaction is performed in a pseudo-homogeneous medium;

- The thermal losses between the reaction mixture and the ambient surroundings can be neglected.

The process consists of a standard batch reactor of 100L, connected with a cooling system. The water utility fluid is provided at a temperature of 288.15K. The tank is filled by two material feeds. The aqueous sodium thiosulfate is introduced through the main inlet process fluid. In order to avoid a possible side reaction, H_2O_2 is in excess. The weight percent in the inlet flows are 15% for $Na_2S_2O_3$ and 30% for H_2O_2. The instrumentation of the process is composed of two temperature sensors (mixture and utility fluid), two flow sensors (the reactor feeding flow rate and the utility fluid flow rate). In normal functioning, the operating conditions of a typical oxidation reaction are summarized in Table 1.

Table 1. Operating Conditions

		Material feed 1	Material feed 2
Weight percent	$Na_2S_2O_3$	15%	-
	H_2O_2	-	30%
	H_2O	75%	70%
Temperature (K)		293.15	333.15
Flowrate (L.h^{-1})		120.9	31.2

In this work, only one exothermic reaction is considered. In this case, the thermal runaway risk can be evaluated according to two values:

– The adiabatic temperature rise $\Delta T_{ad,r}$ which corresponds to the temperature rise at total conversion rate in adiabatic conditions:

$$\Delta T_{ad,r} = \frac{-\Delta H_r F^0_{Na_2S_2O_3}}{\sum_{i=1}^{NC} F^0_i C_{P,i} M_i}$$

(2)

– The time to maximum rate (TMR) is evaluated (at the maximum temperature of the synthesis reaction (MTSR). This temperature corresponds to the temperature that will be reached in result of runaway of the reaction, assuming adiabatic conditions:

$$MTSR = T_p + \Delta T_{ad}$$

(3)

The simulation of the process in adiabatic conditions allows the evaluation of these values. These values are essential for the selection of appropriate safety barriers. Particularly, these can determine the required response time for the barriers.

2.3. Protocol

A typical operation of a batch or semi-batch reactor consists of the tracking of an a priori defined temperature profile. In the case of an exothermic reaction, three distinct steps are involved: preheating, reaction, and cooling (Xaumier *et al.,* 2002). The reactor temperature is controlled by manipulating the flowrate of the utility fluid. Figure 2 represents the experimental set up used in this study.

The process recipe is composed of several steps:

– A feeding step of $Na_2S_2O_3$ during 1200 seconds,

– A heating step to increase the temperature of the mixture from 293.15 K to 333.15 K,

– An other feeding step of H_2O_2 during 1200 seconds, with a constant temperature 333.15 K,

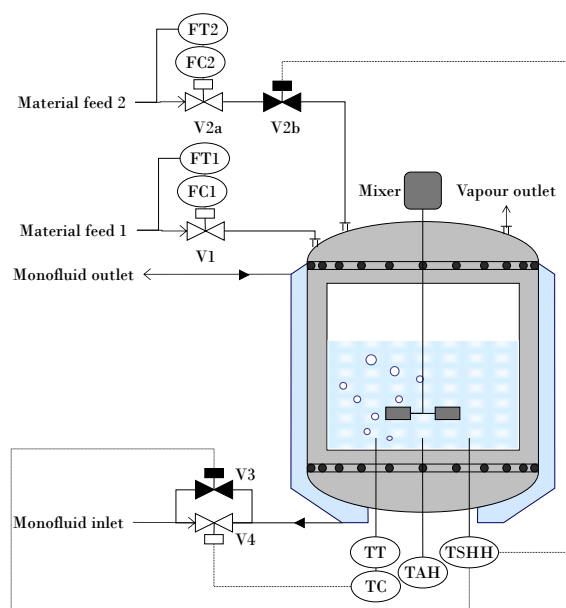– A constant temperature step (the reaction step) at 333.15 K during 4800 seconds.

Figure 2. Semi-batch process

2.4. Risk analysis

The risk management is a major requirement in the industrial context: a major accident is unacceptable. Identifying risks is essential for ensuring the safe design and operation of a process. Several techniques are available to analyse hazardous situations (Marhavilas *et al.,* 2011). Among them, the HAZard and OPerability study (HAZOP) is a well-known technique for studying the hazards of a system (Benaïssa *et al.,* 2008) and its operability problems (International Standard IEC 61882.2001).

Table 2. HAZOP data sheet sample

Section	Guide word	Parameter	Nb	Causes	Consequences
Reactor	More	Flow rate	1	Malfunction of the valves V1a or V2a (for example they remains totally opened)	• Accumulation of reagents • Increase of the reaction speed • Increase of the reactor temperature • Runaway
			2	Malfunction of the controller FC1 or FC2 (for example detection of a wrong value more important)	• Accumulation of reagents • Increase of the reaction speed • Increase of the reactor temperature • Runaway
	None	Flow rate	3	Malfunction of the valves V1 or V2a : they are closed	• Decrease of the conversion rate • Production loss
			4	Plug in pipe	• Decrease of the conversion rate • Production loss
			5	Pipe rupture	• Decrease of the conversion rate • Production loss • Big leaks of hazardous product
			6	Malfunction of FC1 or FC2: detection of a wrong value (less important)	• Decrease of the conversion rate • Production loss

The HAZOP team studied each element for deviations and considered their undesirable consequences. The identification of deviations is obtained by combining process parameters (temperature, flowrate, pressure…) with "guide words" (no, more, less, as well as, reverse, etc.). Each deviation is studied and particularly we search how undesirable events can occur in order to have this deviation. For each deviation, the possible causes are listed, the consequences of these causes are studied and the actions required are suggested. The conclusions are resumed in a

table, (Freeman *et al.,* 1992). Among all the potential hazards highlighted by the HAZOP method, numerous scenarios could potentially lead to a thermal runaway. So, these scenarios were identified as the most hazardous ones. Table 2 illustrates the result data sheet. It gives a short sample of the analysis carried out on the reactor part.

The interest of dynamic simulation in safety analysis of chemical processes is well-known (Ruiz *et al.,* 2001; Chiappetta *et al.,* 2006; Eisenberg *et al.,* 2006; Lou *et al.,* 2006). Moreover, the use of dynamic simulation as a complement in the HAZOP study of chemical reactors has been underlined (Svandova *et al.,* 2005; Di Miceli Raimondi *et al.,* 2015). In this research work, simulations are carried out to estimate the value of the temperature rise and the temperature increasing rate in case of failure.

3. Fault detection and isolation methodology

Nowadays, for reasons of safety and performance, monitoring and supervision play an important role in process control. The complexity and the size of industrial systems induce an increasing number of process variables and make difficult the work of operators. In this context, a computer aided decision-making tool seems to be wise. Nevertheless the implementation of fault detection and diagnosis for stochastic system remains a challenging task. Various methods have been proposed in different industrial contexts (Venkatasubramanian *et al.,* 2003). The proposed approach is a model-based approach involving the use of Hybrid Dynamic System simulation. As defined in (De Kleer *et al.,* 1984), our approach is based on the hypothesis that the reference model is assumed to be correct.

3.1. Architecture

The global principle of this system is shown in Figure 3, where the sequence of the different operations is underlined (Olivier-Maget *et al.,* 2008). Moreover, a distinction between the in-line and off-line operations is made. Our approach is composed of three parts: the generation of the residuals, the generation of the signatures and the generation of the fault indicators.

This methodology has been the subject of previous studies (Olivier-Maget *et al.,* 2009a, 2009b). Nevertheless, it must be validated with other systems and other fault types, in order to ensure its genericity. Current work aims at extending the application areas of process safety. So, the studied case concerns he implementation of an exothermic reaction.

3.2. Residual generation

The first part concerns the generation of the residuals (waved pattern in the Figure 3). In order to obtain an observer of the physical system, a real-time

simulation is done in parallel. So, a complete state of the system will be available at any time. Thus, it is based on the comparison between the predicted behavior obtained thanks to the simulation of the reference model (values of state variables) and the real observed behavior (measurements from the process correlated thanks to the Extended Kalman Filter).
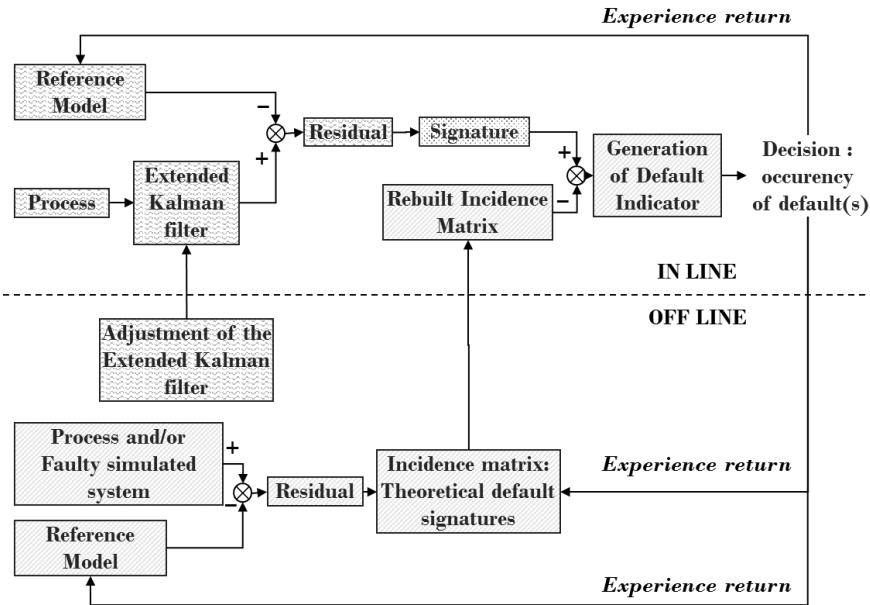

Figure 3. Supervision Architecture

The main idea is to reconstruct the outputs of the system from the measurement and to use the residuals for fault detection (Mehra and Peschon, 1971, Welch and Bishop, 1995, Simani and Fantuzzi, 2006). A lot of state estimators are developed for nonlinear systems (Banerjee and Jana, 2014). Kalman filtering based observer (Li *et al.,* 2004; Qu et Ahn, 2009), extended Luenberger observer (Zeitz, 1987 ; Gundale Mangesh and Jana, 2008), sliding nonlinear observer (Biagiola and Figueroa, 2004; Mezouar *et al.,* 2008), particle filtering (Farza *et al.,* 2004; Olivier *et al.,* 2012) are the most important estimators. In practice, the extended Kalman filter and its derived methods are widely exploited (Jazwinski,1970; Reif and Unbehauen, 1999; Einicke and White, 1999). Kalman filter variants have found widespread applications due to their simplicity and ability to handle reasonable uncertainties and nonlinearities (Salahshour *et al.,* 2008). The extended Kalman filter doesn't use a lot of CPU times and provides good results for systems with a moderate non linearity (Reif and Unbehauen, 1999; Einicke and White, 1999). Consequently, the current work focuses on the design of an extended Kalman Filter.

A description of the extended Kalman filter can be found in (Olivier-Maget et al., 2008). Besides the residual is defined according to the following equation:

$$r_i^r(t) = \frac{\hat{X}_i(t) - X_i(t)}{X_i(t)} \quad with \ i \in \{1, n\} \tag{4}$$

where X_i is the state variable, $\hat{X}_i$ is the estimated state variable with the extended Kalman Filter and n is the number of state variables.

Note that the generated residual $r_i^r(t)$ is relative. As a matter of fact, this allows the comparison of residuals of different variables, since the residual becomes independent of the physical size of the variable.

3.3. Signature generation

The second part is the generation of the signatures (doted pattern in the Figure 3). This is the detection stage. It determines the presence or not of a fault. This is made by a simple threshold $\varepsilon_i(t)$. The generated structure $S_i^{rN}(t)$ is denoted by the following equation:

$$S_i^{rN}(t) = \frac{Max\left[\left(\left|r_i^r(t)\right| - \varepsilon'_i(t)\right); 0\right]}{\sum\limits_{k=1}^{n} Max\left[\left(\left|r_k^r(t)\right| - \varepsilon'_k(t)\right); 0\right]} \quad with \quad i \in \{1, n\} \tag{5}$$

with $\varepsilon'_i(t) = \frac{\varepsilon_i(t)}{X_i(t)}$, where ε_i is the detection threshold.

The value of ε_i is chosen according to the model error covariance matrix of the Extended Kalman Filter (Olivier-Maget, 2007).

3.4. Generation of fault indicator

The last part deals with the diagnosis of the fault (hatched pattern in the Figure 3). The signature obtained in the previous part is compared with the theoretical fault signatures by means of a distance. A theoretical signature $T._j$ of a particular fault j is obtained by experiment or in our case, by simulations of the process with different occurence dates of this fault. Then, a fault indicator is generated. For this, two distances have been created: the relative Manhattan distance and the improved Manhattan distance (Olivier-Maget, 2007). The first distance is denoted by the following expression:

$$D_j^{Mr}(t) = \frac{\sum_{i=1}^{n}\left|S_i^{rN}(t) - T_{ij}\right|}{n} \tag{6}$$

The second distance, which allows the diagnosis of many simultaneous faults, is denoted by the following expression:

$$D_j^{Ma}(t) = \frac{\sum_{i=1}^{n}\left|S_i^{rN}(t) \times m' - T_{ij} \times n'\right| \cdot T_{ij}}{n'} \tag{7}$$

where n' is the number of non-zero elements of the theoretical fault signature $T_{\cdot j}$ and m' is the number of non-zero elements of the fault signature $S^{rN}(t)$.

Since both distances are defined in the space interval [0;1], the fault indicators are defined as the complement to 1 of these distances. An indicator can be viewed as the probability of the occurrence of a particular fault. These indicators follow a normal distribution. The well-known statistical test of Shapiro-Wilk confirms the veracity of this hypothesis. This is used to test normality. According to the test value, we can accept or reject the hypothesis that the corresponding distribution is normal. The Shapiro -Wilk W test is the most widely used normality test because it is a powerful test compared to many alternative tests (Shapiro and Wilk, 1968).

Next, the generated fault indicators are exploited to take a diagnosis of the system. For this, we suppose that:

– The minimum value of the indicator, for which the fault can be considered, is 0.68 – chosen according to the normal distribution–. This threshold corresponds to the probability at the standard deviation.

– The number of faults, which can simultaneously take place, is limited to three.

4. Results

The advantage of the simulation of the chemical reaction thermal effect is to prevent runaway problems, which can occur experimentally, particularly for such an exothermic reaction (Xaumier *et al.,* 2002). Then, to demonstrate the good performance of the diagnosis methodology, different simulations have been carried out on the hybrid dynamic simulator PrODHyS. Two faults have been introduced:

– Increase of the flow rate (fault 1): In this case, 6500 s after the beginning of the reaction, the feed rate of the sodium thiosulfate is multiplied by 2.

– Increase of the jacket temperature (fault 2): 7500 s after the beginning of the reaction, the cooling temperature is increased to 10 °C.

4.1. Adjustments

To perform a monitoring of a process, some off-line adjustments must be made. On the one hand, we need to determine the covariance matrices of the model and measurement disturbances. While the measurement noises are supposed to be well-known by experiments or by the sensor manufacturer, the model disturbances is estimated by an "ensemble method". This method is based on the following hypothesis: the sampling average corresponds to the best estimate of the system state (our model is supposed to be unbiased), and the dispersion around this average corresponds to a measurement of the mistake of this estimate (Evensen, 2003; Maget, 2007). Thus, numerous simulations have been performed during which a model parameter has been disturbed. This allowed the estimation of statistic distribution of the model mistakes. Then, if the behaviour of the system goes beyond this distribution, its behaviour is abnormal. So, the detection thresholds are determined according to the model disturbances.

On the other hand, the second adjustment is the learning of the incidence matrix. It is based on the same "ensemble" theory. For this, we perform a set of simulations, during which a fault is introduced at different occurrence dates, for each potential state of the hybrid dynamic system (Figure 4).
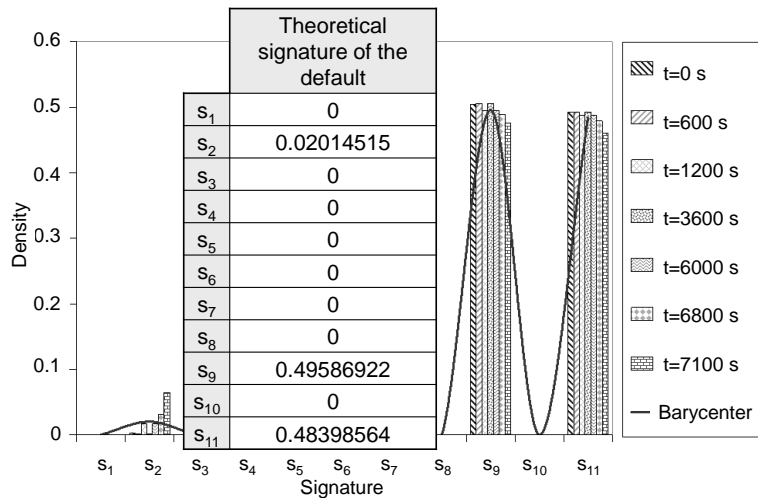

	Theoretical signature of the default
s_1	0
s_2	0.02014515
s_3	0
s_4	0
s_5	0
s_6	0
s_7	0
s_8	0
s_9	0.49586922
s_{10}	0
s_{11}	0.48398564

Figure 4. Learning of the incidence matrix

For this study we consider six faults. These faults are underlined by the HAZOP study. These faults could lead to a thermal runaway:

– Fault 1: The *material feed 1* provides material with a damaged flow rate (superior to the normal rate);

– Fault 2: The *material feed 1* provides material with a damaged composition (superior to the normal rate);

– Fault 3: The *material feed 2* provides material with a damaged flow rate (superior to the normal rate);

– Fault 4: The *material feed 2* provides material with a damaged composition (superior to the normal rate);

– Fault 5: The *energy feed* has a damaged temperature (superior to the normal temperature);

– Fault 6: The temperature regulation is damaged.

For example, figure 4 represents the results obtained for the fault 3. The signatures of this fault are presented for different occurence dates. They have the same pattern. The barycenter is estimated and we obtain the theoretic signature of the fault 3. Next, we perform this preliminary study for all the considered faults of the system and the incidence matrix is presented in table 3.

Table 3. Incidence Matrix

	Fault 1	Fault 2	Fault 3	Fault 4	Fault 5	Fault 6
Temperature (K)	0	0	0	0	0	0
Holdup (mol)	1	0.030239	0.020145	0	0	0
X[Sodium Thiosulfate]	0	0.950786	0	0	0	0
X[Hydrogen Peroxide]	0	0	0	0.289313	0	0.10448
X[Sodium Trithionate]	0	0	0	0	0	0
X[Sodium Sulfate]	0	0	0	0	0	0
X[Water]	0	0.018975	0	0.00103	0	0.000208
Reaction conversion	0	0	0	0	0	0
Cooling power (W)	0	0	0.495869	0.404899	0.009761	0.681392
Fluid Utility Temperature	0	0	0	0	0.049209	0
Fluid Utility Flowrate	0	0	0.483986	0.304758	0.94103	0.21392

Notice that the faults 4 and 6 would be confused, if we work with binary values. The work with real and non-binary values aims at distinguishing the importance of the symptoms between a fault with another one. Therefore, for a particular fault, the normalization allows that the great symptoms are underlined.

4.2. Normal operation

Figure 5 represents the simulated results for a normal operation. It gives the temperature profile of the process mixture, the evolution of the liquid holdup in the reactor, the conversion rate and the characteristics (flow rate and power) of the fluid utility. It must be noticed that the utility fluid flow rate increases during the feeding

step of H_2O_2. It points out that this oxidation reaction is fast and highly exothermic. We also notice that the conversion is not total (87%).
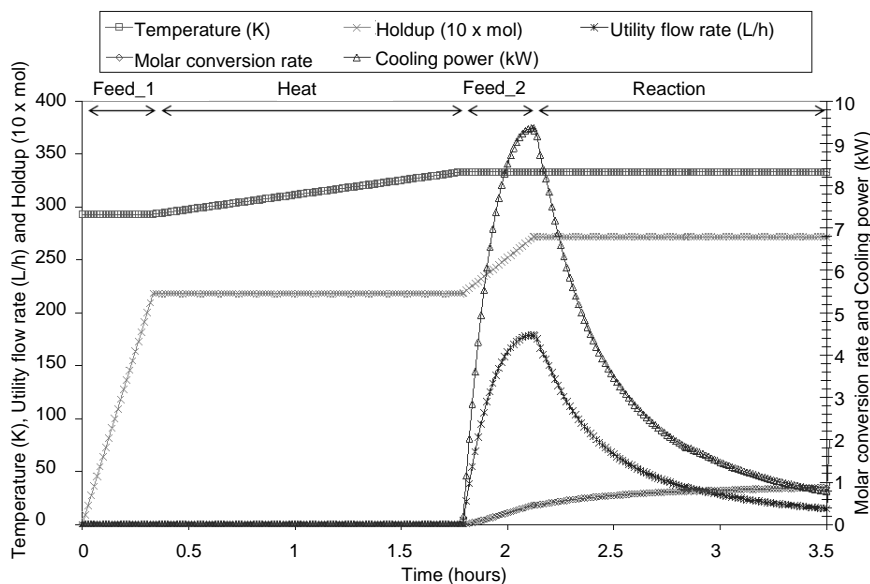

Figure 5. Normal operation

4.3. Simulation of the Adiabatic Reactor

The reaction of sodium thiosulfate with hydrogen peroxide, conducted in liquid solution, is very fast and strongly exothermic. The evacuation of heat released by the reaction can generate safety problem. The simulation of this reaction in the adiabatic reactor is studied. The results of this simulation allows the estimation of the adiabatic temperature rise $\Delta Tad,r$ and the time to maximum rate (TMR). This temperature corresponds to the temperature that will be reached in result of runaway of the reaction, assuming adiabatic conditions. For this simulation, we obtain, in an adiabatic condition, a temperature rise of 77 K. The obtained Time to Maximal Rate, TMR, is higher than 30 minutes. These both values parameters provide information about the kinetics of thermal runaway.

4.4. Detection

Let us remind that the thresholds for the detection correspond to the model uncertainties obtained by the adjustment of the Extended Kalman filter. Figure 6 shows the detection step. It illustrates the evolution of the residual linked to the utility fluid flow rate. In order to avoid a false diagnosis, the diagnosis step is not

launched as far as the symptoms of the abnormal behaviour appear. The abnormal behaviour is only confirmed after it has occurred three times.

Example 1: A fault of the material feed 2 is introduced at t = 6500 s. The material feed 2 provides material with a damaged flow rate (multiplied by 2). From t = 6800 s, the values of the residual underline the abnormal behaviour of the process. The diagnosis is launched at t = 6900 s. So, in less than 7 minutes, a diagnosis is made.

Example 2: A fault of the energy feed is introduced at t = 7500 s. The energy feed has a damaged temperature (increasing of 10 °C). From t = 7600 s, the values of the residual underline the abnormal behaviour of the process. The diagnosis is launched at t = 7700 s. So, in less than 4 minutes, a diagnosis is made.
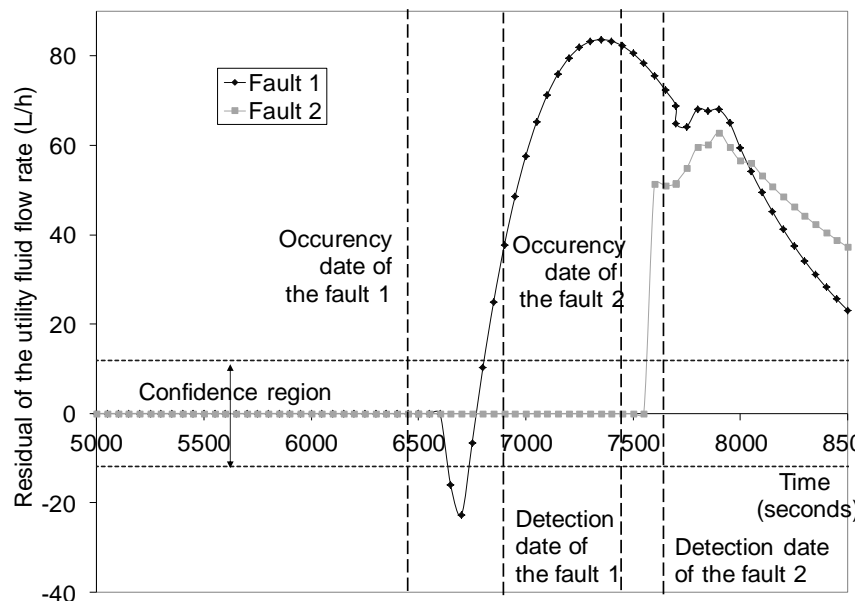

Figure 6. Residual evolution of the utility fluid flow rate

4.5. Diagnosis

Let us notice that the exploited signature in this approach is non binary, in order to quantify the deviation due to the fault.

The residual is then estimated and we obtain the corresponding instantaneous fault signature (Table 4). We compare the instantaneous fault signature (Table 4) with the theoretical fault signatures, by calculating the relative and improved

Manhattan distances ((6) and (7)). Then, the fault indicators are generated (Table 5). They correspond to the complement to 1 of these distances.

Table 4. Instantaneous fault signatures of both examples

	Example 1	Example 2
Temperature (K)	0	0
Holdup (mol)	0.0210	0
X[Sodium Thiosulfate]	0	0
X[Hydrogen Peroxide]	0	0
X[Sodium Trithionate]	0	0
X[Sodium Sulfate]	0	0
X[Water]	0	0
Reaction conversion	0	0
Cooling power (W)	0.5094	0.0007
Fluid Utility Temperature	0	0.1144
Fluid Utility Flowrate	0.4637	0.8849

Table 5. Fault indicators

		Fault 1	Fault 2	Fault 3	Fault 4	Fault 5	Fault 6
	Manhattan relative indicator	0.8225	0.8225	0.9968	0.9477	0.9048	0.9502
	Manhattan improved indicator	0.0630	0.0954	0.9835	0.8939	0.5435	0.7565
	Manhattan relative indicator	0.8182	0.8182	0.9063	0.8737	0.9881	0.8572
	Manhattan improved indicator	0	0.0947	0.5600	0.6432	0.9439	0.4289

Example 1: The relative Manhattan indicator detects the presence of the fault 3 with a probability of 99.68%. Nevertheless, the other faults are not eliminated, since their indicators are higher than 0.68 (cf. the point 3.4). In the opposite, with the improved Manhattan indicator, the faults 1, 2, and 5 are eliminated, since their indicators are lower than 0.68. The three remaining possibilities are the faults 3, 4, and 6. This example underlines the importance of using both indicators to be able to

conclude. So, by combining the results of the both indicators, we can rule on the presence of the fault 3, since their indicators are the maximums. For this reason, this fault is the most probable. So, the fault is located on the material feed 2. Furthermore, it has been identified: the material feed 2 provides material with a damaged flowrate.

Example 2: The relative Manhattan indicator detects the presence of the fault 5 with a probability of 98.81%. However, none fault is discriminated (all indicators higher than 0,68). With the improved Manhattan indicator, the presence of the fault 5 is distinguished, since the other indicators are lower than 0.68. So, by combining the results of both indicators, we can rule on the presence of the fault 5. Then, the fault is located on the energy feed of the reactor. Furthermore, it has been identified: the cooling energy feed of the reactor has a damaged temperature.

5. Conclusion

In this research work, the feasibility of using the simulation as a tool for fault detection and diagnosis is demonstrated. The fault detection and diagnosis approach, developed here, lies on the hybrid dynamic simulator PrODHyS. It is a general method for the detection and isolation of a fault occurrence. Besides, this approach allows the detection of numerous types of fault and has the ability to underline the simultaneous occurrence of many faults.

The works in progress aim at integrating this hybrid dynamic system simulation within a model-based supervision system. The goal is to define a recovery solution following the diagnosis of a default. For this, the results of signatures will be exploited in order to generate qualitative information. For example, with these results, we have the ability to distinguish a simple degradation and a failure. Next, this method will be combined with other diagnosis approaches: the set of the potential faults will be generated according to such methods as classification or case-based reasoning, restricting the search domain (to a part of the process or to a set of potential faults yielding similar symptoms) examined with the model-based technique. Moreover, the signature and residual results provide pertinent information to characterize the system observability and diagnosability. Our work could contribute to the placement of the most relevant sensors for the process diagnosis.

Relating to the operation of exothermic reactions, a novel concept of heat exchangers reactors offers enhanced thermal performances in continuously operating reactors. The experiment results emphasize a significant thermal efficiency: the reactant concentrations and therefore the heat generation can be increased without risk of thermal runaway (Di Miceli Raimondi et al., 2015). The Polysafe ANR project (N° ANR-2012-CDII-0007-01) focuses on the modelling of these reactors in normal and degraded mode, to study the process drift and demonstrate the inherently safer nature of these devices.

References

Anderson T.W. (1984). *An introduction to multivariate statistical analysis*, New York, Wiley.

Banerjee S., Jana A. K. (2014). High gain observer based extended generic model control with application to a reactive distillation column, *Journal of Process Control*, 24(4), p. 235-248.

Benaissa W., Gabas N., Cabassud M., Carson D., Elgue S., Demissy M. (2008). Evaluation of an intensified continuous heat-exchanger reactor for inherently safer characteristics, *Journal of Loss Prevention in the Process Industries*, 21(5), p. 528-536.

Benkouider A.M., Kessas R., Yahiaoui A., Buvat J.C., Guella S. (2012). A hybrid approach to faults detection and diagnosis in batch and semi-batch reactors by using EKF and neural network classifier, *Journal of Loss Prevention in the Process Industries,* 25(4), p. 694-702.

Biagiola S.I., Figueroa J.L. (2004). A high gain nonlinear observer: application to the control of an unstable nonlinear process, *Computers & Chemical Engineering*, 28, p. 1881-1898.

Chang C.T., Chen C.Y. (2011). Fault diagnosis with automata generated languages, *Computers and Chemical Engineering*, 35, p. 329-341.

Chetouani Y., Mouhab N., Cosmao J.M. and Estel L. (2003). Dynamic model-based technique for detecting faults in a chemical reaction, *Process Safety Progress*, 22(3), p. 183-190.

Chetouani Y. (2004). Fault detection by using the innovation signal: application to an exothermic reaction, *Chemical Engineering and Processing*, 43, p. 1579-1585.

Chiappetta G., Clarizia G. and Drioli E. (2006). Analysis of safety aspects in a membrane reactor, *Desalination*, 193 (1–3), p. 267-279.

Chow E.Y. and Willsky A.S. (1984). Analytical redundancy and the design of robust failure detection systems*, IEEE Transactions on Automatic Control*, 29(7), p. 603-614.

De Kleer J. and Williams B.C. (1987). Diagnosing multiple faults, *Artificial Intelligence*, 32, p. 97-130.

Di Miceli Raimondi N., Olivier-Maget N., Gabas N., Cabassud M., Gourdon C. (2015). Safety enhancement by transposition of the nitration of toluene from semi-batch reactor to continuous intensified heat exchanger, *Chemical Engineering Research and Design,* 94, p. 182-193.

Ding S.X. (2014). Data-driven design of monitoring and diagnosis systems for dynamic processes: A review of subspace technique based schemes and some recent results, *Journal of Process Control,* 24(2), p. 431-449.

Einicke G.A., White L.B. (1999). Robust Extended Kalman Filtering, *IEEE Trans. Signal Process.*, 47(9), p. 2596-2599.

Eizenberg S., Shachama M. and Brauner N. (2006). Combining HAZOP with dynamic simulation - Applications for safety education, *Journal of Loss Prevention in the Process Industries,* 19(6), p. 754-761.

Evensen G. (2003). The Ensemble Kalman Filter: Theoretical Formulation and Practical Implementation, *Ocean Dynamics*, 53, p. 343-360.

Farza M., M'Saad M., Rossignol L. (2004). Observer design for a class of MIMO nonlinear systems, *Automatica*, 40, p. 135-143.

Frank P.M. (1990). Fault diagnosis in dynamic systems using analytical and knowledge-based redundancy – a survey and some new results, *Automatica*, 26, p. 459-474.

Freeman R.A., Lee R., McNamara T.P. (1992). Plan HAZOP studies with an expert system, *Chemical Engineering Progress*, p. 28-32.

Gertler J. and Singer D. (1990). A new structural framework for parity equation-based failure detection and isolation, *Automatica*, 26, p.381-388.

Grau M.D., Nouguès J.M. and Puigjaner L. (2000). Batch and semibatch reactor performance for an exothermic reaction, *Chemical Engineering and Processing,* 39, p. 141-148.

Gundale Mangesh M., Jana A.K. (2008). A comparison of three sets of DSP algorithms for monitoring the production of ethanol in a fed-batch baker's yeast fermenter, *Measurement,* 41, p. 970-985.

Gygax R. (1988). Chemical reaction engineering for safety, Chemical reaction engineering for safety, *Chemical Engineering Science*, 43(8), p. 1759-1771.

Hétreux G., Théry R., Olivier-Maget N., Le Lann J.M. (2007). Exploitation de la simulation dynamique hybride pour la conduite de procédés semi-continus, *Journal Européen des Systèmes Automatisés*, 41(5), p. 585-616.

Jazwinski A.H.. (1970). *Stochastic Processes and Filtering Theory*, Libri, p. 376.

Li R., Corripio A.B., Henson M.A., Kurtz M.J. (2004). On-line state and parameter estimation of EPDM polymerization reactors using a hierarchical extended Kalman filter, *Journal of Process Control*, 14, p. 837-852.

Lo S.N. and Cholette A. (1972). Experimental study on the optimum performance of an adiabatic MT reactor, *The Canadian Journal of Chemical Engineering,* 50, p. 71-80.

Lou H.H., Chandrasekaran J., Smith R.A. (2006). Large-scale dynamic simulation for security assessment of an ethylene oxide manufacturing process, *Computers and Chemical Engineering*, 30, p. 1102-1118.

Maget V. (2007). *Développement et comparaison de méthodes d'assimilation de données appliqués à la restitution de la dynamique des ceintures de radiation de la Terre.* PhD-Thèse, université de Toulouse (École Nationale Supérieure de l'Aéronautique et de l'Espace), France, p. 223.

Marhavilas P.K., Koulouriotis D., Gemeni V. (2011). Risk analysis and assessment methodologies in the work sites: On a review, classification and comparative study of the scientific literature of the period 2000-2009, *Journal of Loss Prevention in the Process Industries*, 24(5), p. 477-523.

Mehra R.K. andPeschon J. (1971). An Introduction approach to fault detection and diagnosis in dynamic systems, *Automatica*, 5, p. 637-640.

Mezouar A., Fellah M.K., Hadjeri S. (2008). Adaptive sliding-mode-observer for sensorless induction motor drive using two-time-scale approach, *Simulation Modelling Practice and Theory*, 16, p. 1323-1336.

Olivier B., Huang I., Craig K. (2012). Dual particle filters for state and parameter estimation with application to a run-of-mine ore mill, *Journal of Process Control*, 22, p. 710-717.

Olivier-Maget N. (2007). *Surveillance des Systèmes Dynamiques Hybrides : Application aux procédés*, PhD Thesis of the Toulouse University (INSA).

Olivier-Maget N., Hétreux G., Le Lann J.M. and Le Lann M.V. (2008). Integration of a failure monitoring within a hybrid dynamic simulation environment, *Chemical Engineering and Processing*, 47(11), p.1942-1952.

Olivier-Maget N., Hétreux G., Le Lann J.M. and Le Lann M.V. (2009a). Model-Based Fault Diagnosis Using a Hybrid Dynamic Simulator: Application to a Chemical Process, *Computer Aided Chemical Engineering,* 27, p. 1641-1646.

Olivier-Maget N., Hétreux G., Le Lann J.M. and Le Lann M.V. (2009b). Fault Detection and Diagnosis Based on a Hybrid Dynamic Simulator, published in *The 7th IFAC International Symposium on Fault Detection, Supervision and Safety of Technical Processes,* Spain, p. 223-228.

Perret J., Hétreux G. and Le Lann J.M. (2004). Integration of an object formalism within a hybrid dynamic simulation environment, *Control Engineering Practice*, 12(10), p. 1211-1223.

Prat L., Devatine A., Cognet P., Cabassud M., Gourdon C., Elgue S. and Chopard F. (2005). Performance Evaluation of a Novel Concept "Open Plate Reactor" Applied to Highly Exothermic Reaction, *Chemical Engineering Technology*, 28(9), p. 1028-1034.

Qu C.C., Hahn J. (2009). Process monitoring and parameter estimation via unscented Kalman filtering, *Journal of Loss Prevention in Process Industries*, 22, p. 703-709.

Rawlings J.B., Bakshi B.R. (2006). Particle filtering and moving horizon estimation, *Computers & Chemical Engineering*, 30, p. 1529-1541.

Reif K., Unbehauen R. (1999). The extended Kalman filter as an exponential observer for nonlinear systems, *IEEE Trans. Signal Process.*, 47(8), p. 2324-2328.

Ruiz D., Canton J., Nougues J.M., Espuna A., Puigjaner L. (2001). On-line fault diagnosis system support for reactive scheduling in multipurpose batch chemical plants, *Computers and Chemical Engineering,* 25 (4–6), p. 829-837.

Salahshour K., Mosallaei M., Bayat M. (2008). Centralized and decentralized process and sensor fault monitoring using data fusion based on adaptive Extended Kalman Filter algorithm, *Measurement*, 41, p. 1059-1076.

Simani S. and Fantuzzi C. (2006). Dynamic system identification and model-based fault diagnosis of an industrial gas turbine prototype, *Mechatronics*, 16, p. 341-363.

Shih R. and Lee L. (1995). Use of fuzzy cause-effect digraph for resolution fault diagnosis for process plants, *Industrial and Engineering Chemistry Research*, 34(5), p. 1688-1717.

Shapiro S. S. and Wilk M. B. (1965). An analysis of variance test for normality (complete samples), *Biometrika*, 52 (3-4), p. 591-611.

Stoessel F. (2008). *Thermal safety of chemical processes*, *Risk assessment and Process Design*, Wiley-VCH, p. 374.

Svandova Z., Jelemensky L., Markos J., Molnar A. (2005). Steady states analysis and dynamic simulation as a complement in the HAZOP study of chemical reactors, *Process Safety and Environmental Protection*, 83 (B5), p. 463-471.

Venkatasubramanian V., Rengaswamy R., Yin K. and Kavuri S. N. (2003). A review of process fault detection and diagnosis, *Computers & Chemical Engineering*, 27, p. 293-346.

Welch G. and Bishop G. (1995). *An introduction to the Kalman filter*, Technical Report TR 95-041, University of North Carolina.

Willsky A.S. (1976). A survey of design methods for failure detection in dynamic systems, *Automatica*, 12, p. 601-611.

Xaumier F., Le Lann M.V., Cabassud M. and Casamatta G. (2002). Experimental application of nonlinear model predictive control: temperature control of an industrial semi-batch pilot-plant reactor, *Journal of Process Control*, 12(6), p. 687-693.

Zeitz M. (1987). The extended Luenberger observer for nonlinear systems, *System & Control Letters*, 9, p. 149-156.