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# Directed Hypergraph model-based FDI of chemical reaction

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***ABSTRACT:*** *Different graphical tools (Bond Graph, Signed diercted graph, Temporal Causal Garaph, ...) have been developed to perform diagnosis of chemical processes. As hypergraphs generalize graphs in which an edge can connect any number of vertices, this paper aims to generate a hypergraph model-based fault detection and isolation. The directed hypergraph model of chemical reaction kinetics have been used to generate redundancy relations. The proposed fault indicator scheme is illustrated through a reaction which has a second-order reaction rate*
***Keywords:*** *chemical reaction, directed hypergraph, FDI*

## I. INTRODUCTION

In general, many of chemical and biochemical reactions used in industry are quite dangerous and compounds are formed from elements which can react violently, burn so rapidly or even explode [1] ,[2]. When a very wide amount of the chemical reactions can relatively cause damage, an early Fault Detection and Isolation ($FDI$) in such cases will contribute to decrease the process upsets and retain it reliable and effective with respect to dynamic behaviors in healthy operating conditions.

In this context, several methodologies relied on the analytical redundancy principle have been developed to address the fault diagnosis challenges. Among them, observers [3], [4], [5] and Kalman filters [6], [7] which require a lot of computing and numerical values of the chemical process parameters. These latter are not always available. In addition, fault diagnosis of chemical processes is relatively difficult due to the incomplete prior-knowledge and unpredictable production changes. Graphical methods can therefore offer appropriate solutions to many difficulties arising in this field. For instance the Bond Graph (BG) and the Signed Directed Graph (SDG) have been widely used for both modeling and monitoring purposes. In fact, several authors [8], [9], [10] have proposed to use the bond graph formalism where the set of quantitative equations can be derived and can be after that formulated for model-based fault diagnosis by finding the fault indicators which are easily calculated (for more details see [11], [12]) Nevertheless, when the process becomes more complex, the number of quantitative equations increases and the testing process gets inefficient. The SDG, as a kind of qualitative graphical models, is whereas well suited for this situation. Due to its causal and structural properties, relationships between a cause-effect pair in subsystems are graphically described without any need of numerical calculations [13], [14], [15], [16] and references therein for more details).

On the other side, hypergraphs are further considered as a generalization of graphs allowing representing relationships explicitly. These techniques are involved in many areas of sciences ([17], [18], [19]) and even they are useful in chemical engineering applications. Recently, Directed Hypergraphs (DH) have attracted many researchers on modeling of chemical reaction networks. In [20] Temkin and al. look for a good representation of reaction behaviors by using weighted directed hyper-edges. Nodes are the chemical components whereas hyper-edges are the reactions. Moreover, the difference that may arise in modeling biological facts with graphs and hypergraphs where multimolecular reactions are modelled by using directed hypergraphs [21]. The availability of an accurate chemical kinetics model of the monitored chemical systems can significantly improve the performance of diagnostic tools, minimising the probability of false alarms. As underlined in [22],

A large amount of attention is paid to the chemical reaction modelling problem, with reference to nonlinear model structures. However, the chemical representation using directed hypergraphs has not been already introduced in literature. Thus, an alternative framework for enhancing FDI performances is required.

The novelty of this paper is to design a FDI algorithm based on the behavioral, structural and causal properties of our hypergraph model associated to chemical reaction kinetics proposed in [22]. In point of fact, the Directed Hypergraph description is automatically obtained from the bond graph modeling and applied later

for qualitative analysis of FDI tasks. Compared with the bond graph based residual signals [23], the developed framework is able to minimize the number of redundant relations taking into account the qualitative state of known variables. It aims to improve the monitoring time and the performances (including rapidity and accuracy) through this fault generator scheme without any need of numerical calculations. The case study is presented so as to illustrate the efficiency of the developed approach.

The paper proceeds as follows: Section **1** gives some notations and preliminaries necessary for the present work.The problem formulation is presented in Section 2. Afterwards, Section 3 describes the proposed FDI scheme for generating qualitative redundancy relations well-founded on directed hypergraph tool. Then, we compare between the developed algorithm and the classical BG procedure via a case study in Section V. Section 4 recapitulates some ultimate remarks. Section 5 concludes the paper by highlighting the strengths of the proposed method.

## II. NOTATIONS AND PRELIMINARIES*:*

We provide herein some definitions and properties involved thereafter for the Hypergraphs-based fault monitoring.

### 2.1 Graph :

A graph $G$ is defined as a dual pair $(V, A)$ where $V = (v_1, v_2, \dots, v_n)$ is a finite set whose elements are called nodes and $A = (a_1, a_2, \dots, a_m)$ a finite set of elements of V whose elements are called edges. An edge $a_i$ of the set $A$ is defined by an unordered pair of nodes, called the ends of $a_i$. If edge $a_i$ connects the vertices $a$ and $b$, we say that these vertices are adjacent, or incidents with $a_i$ or that edge $a_i$ is incident with vertices $a$ and $b$.

### 2.2 Bond Graph :

The Bond Graph (BG) is a graph $G = (V, A)$ where its nodes $V$ represent subsystems basic elements (junction) or components and edges $A$ show the instantaneous mutual power transfers between nodes (power bonds). BG is unified for all physical fields. For a detailed presentation of the BG method we refer the reader to [24], [25].

### 2.3 Hypergraph :

A hypergraph is a generalization of a graph in which an edge can be connected to any number of vertices. Formally a hypergraph $H$ is a pair $(V, E)$ of finite set $V = (v_1, v_2, \dots, v_n)$ and a set $E = (e_1, e_2, \dots, e_m)$ of subsets of $V$ such that $\${ e_i \neq \varnothing, \ \forall i$ and $\cup e_i = V$. The elements of $V$ are called vertices and elements of $E$ are called hyperedges.

### 2.4 Directed Hypergraph :

A directed hypergraph $H = (V, E)$ is a hypergraph with a directed hyperedges , such that every $e = (T(e), H(e)) \in E$ is an ordered pair of non-empty disjoint subsets $T(e)$ and $H(e)$. where $T(e)$ and $H(e)$ are the sets of vertices that appear respectively in the tail and the head of the hyperarc $e$. We say that $e$ is incident on each vertex in $T(e)$. For each $\upsilon \in T(e)$ , $e$ is outgoing hyperedge from $\upsilon$ and for each $\upsilon \in H(e)$, $e$ is incoming hyperedge to $\upsilon$.

### 2.5 Path in Hypergraph (hyperpath) :

In a hypergraph, a path $P_{st} = (v_1, e_1, v_2, e_2, \dots, v_q, e_q, v_{q+1})$ connecting vertices $s$ and $t$ is a sequences of vertices and hyperedges satisfying $\${ v_i, v_{i-1} \in e_i, 1 \leq i \leq q$ , $s = v_1$, $t = v_q$, all hyperedges are distinct and all vertices are distinct.

### 2.6 Directed hyperpath :

A directed hyperpath $P_{st}$ from vertex $s$ to vertex $t$ in a directed hypergraph $H$ is an alternating sequences of vertices and hyperedges $P_{st} = \left(v_1 = s, e_{i_1}, v_2, e_{i_2}, v_3, \ldots, v_q, e_{i_q}, v_{q+1} = t\right)$, where $v_j \in T\left(e_{i_j}\right)$, $v_{j+1} \in H\left(e_{i_j}\right)$ for each $1 \leq j \leq q$, all hyperedges are distinct and all vertices are distinct.

### 2.7 B-connection and B-hyperpath :

A hyperedge $e = \left(T(e), H(e)\right)$ with $\left|H(e)\right| = 1$ is called a Backward hyperedge, or simply B-hyperedge.

The B-connection in directed hypergraph is defined as follows:

- Vertex $v$ is connected to itself.
- If for $e_i \in E$ all the vertices in $T(e)$ are B-connected to $v$, then each vertex $u$ in $H(e)$ is B-connected to $v$.

The notion of B-hyperpath generalise the notion of a directed path in digraphs.

### 2.8 Forward and Backward star :

The Forward star FS and Backward star BS are defined respectively as follow:

$$FS(v) = \left\{e \in E, v \in T(e)\right\} \text{ and } BS(v) = \left\{e \in E, v \in H(e)\right\}.$$

## III. PROBLEM STATEMENT

Consider the following second-order reaction assumed to be reversible and generated in this form as

$$\upsilon_{A_1} A_1 + \cdots + \upsilon_{A_n} A_n \underset{A_r}{\overset{A_f}{\square}} \upsilon_{B_1} B_1 + \cdots + \upsilon_{B_m} B_m \qquad (1)$$

Where $E = \bigcup_{i=1}^{n} A_i$ the set of reactants and $P = \bigcup_{j=1}^{m} B_j$ represents the set of products whereas $v-{i}$ are the stoichiometric coefficients.

In accordance with the law of mass action, the rate of the chemical reaction is stated as follows

$$J = k_+ \prod_{l \in E} C_l^{|\upsilon_l|} - k_- \prod_{l \in P} C_l^{|\upsilon_l|} \qquad (2)$$

where $k_+$ and $k_-$ are the forward and reverse speed constants respectively.

From the fundamental equations of thermodynamics and the mole balances, the density of entropy production $\dot{S}_{gen}\left[JK^{-1}m^{-3}s^{-1}\right]$ of a reaction is given by:

$$A * J = T * \dot{S}_{gen} \qquad (3)$$

where $J\left[mol\, m^{-3} s^{-1}\right]$ is the reaction flux, $T[K]$ the thermodynamic temperature

and $A\left[J\, mol^{-1}\right]$ is the negative Gibbs reaction energy:

$$A = A_f - A_r \qquad (4)$$

$A_f$ and $A_r$ are linear combinations of the chemical potentials $\mu_i$:

$$A_f = \sum_{i \in E} |\upsilon_i| \mu_i \qquad (5)$$

$$A_r = \sum_{i \in P} |\upsilon_i| \mu_i \qquad (6)$$

For ideal mixtures, the chemical potentials $\mu_i$ are given by :

$$\mu_i = \mu_0^i + RT \ln\left(C_i / C_0\right) \qquad (7)$$

such that $C_i = \frac{n_i}{V} = \frac{1}{V}\int \dot{n}_l$, $V$ the volume of the reaction, $\mu_0^i$ the chemical standard potential of pure chemical species, $C_0$ a standard concentration (e.g. $C_0 = 1 mol/L$) and $R$ the universal gas constant.
Thus, we obtain:

$$\mu_i = \mu_0^i + RT \ln\left(\frac{1}{C_0 V}\int \dot{n}_l\right) \quad (8)$$

As we can write :

$$J = k_+ \prod_{i\in E}\left(\frac{\int \dot{n}_l}{V}\right)^{|\upsilon_i|} - k_- \prod_{i\in P}\left(\frac{\int \dot{n}_l}{V}\right)^{|\upsilon_i|} \quad (9)$$

The molar balances of different species are defined as the following:

$$\begin{cases} \dot{n}_l = \dot{n}_{li} - \dot{n}_{lo} - \dot{n}_{1l} & , \quad l = A_i, i = 1\cdots n \\ \dot{n}_l = -\dot{n}_{lo} + \dot{n}_{1l} & , \quad l = B_j, j = 1\cdots m \end{cases} \quad (10)$$

where $\dot{n}_{1l} = \upsilon_l J$ and $\dot{n}_{li}$ and $\dot{n}_{lo}$, respectively denote the inlet and outlet molar flow, are written by:

$$\begin{cases} \dot{n}_{li} = \dot{m}_l / M_l & , \quad l = A_i \\ \dot{n}_{lo} = \dot{n}_l\left(\dot{m}_l / (\rho V)\right) & , \quad l = A_i, B_j \end{cases} \quad (11)$$

$\rho$ : the mass density, $M_l$ : the molar mass.

In our previous work [21], we develop a novel model for a chemical reaction kinetics using hypergraph representation as graphical tool. This model can be derived directly from the basic energy and material balances where vertices represent space variable, input and output variables and hyperedges are the relationships between them. This model is able to rewrite the kinetic rate using power conjugate variables (chemical affinity, rate). Secondly, we present a Bond Graph-Hypergraph analogy to get the directed hypergraph model directly from the Bond Graph model.
We aim in this work for monitoring based hypergraphs where we will generate analytical redundancy relationship (ARR) from the developed directed hypergraph model based on the properties and characteristics of hypergraphs.

## IV. PROPOSED FAULT MONITORING FRAMEWORK

### 4.1 Redundancy Relations for fault monitoring :

A Redundancy Relationship (RR) is a model constraint that contains known terms of the system such as inputs, sensor measurements and model parameters. This RR is generated by eliminating unknown variables which is not always a trivial task especially for complex systems. For linear systems one may use for example the parity space technique to detect sensors and actuators faults. A model based FDI (Fault Detection and Isoltaion) method relies on the chosen system's model where the difficulty increases with the complexity of the system. Nonetheless, as the system becomes more complex (in particularly for large scale systems), the number of Analytical Redundancy Relations (ARRs) increases and the testing process becomes inefficient. To overcome these limitations, the proposed framework is developed well-founded on the merging of qualitative reasoning and bond graph modeling theory so as to benefit from both methods. In point of fact, the qualitative description is involved to obtain qualitative redundant relations (QRRs)deduced automatically from the hypergraph representation which is built by using the BG model.
The proposed framework will be considered in this paper so as to focus on improving the performance according to the fault monitoring procedure.

### 4.2 Bond Graph model-based FDI :

In this sequel we recall the BG-based method, called covering path method, to generate the ARRs.
The ARRs are derived by eliminating the unknowns variables using the causal paths of the BG model. The method can be stated as follows:

1) Assign a preferred derivative causality on the nominal BG.
2) Choose a junction in the model.
3) Find the corresponding ARR by writing its characteristic equation of the considered junction. Unknown variables are eliminated by covering causal paths in the Bond Graph model.
4) Consider the following junction.
5) If the corresponding ARR is different from the the other derived ARRs then keep it, else consider another junction.
6) Repeat point 4 until all junctions are considered and the independent signatures are obtained.

**4.3 Directed Hypergraph model-based FDI :**

This proposed method generates systematically ARRs from a hypergraph model. We determine the B-hyperpaths that relate known variables to generate ARRs. The known variables in the B-hyperpath are elements of the ARR. The method is presented algorithmically as follows:

1) $K = \{\text{unknown variables}\}$
2) for all variables $v_i$ in $K$ do :

   $P = \varnothing$

- Step 1 : define $FS(v_i)$

  for $e_j \subseteq FS(v_i)$

  if there is $v_k \in H(e_j)$ is a known variable

  then $P = \{P, v_k\}$ , go to Step 2

  else $v_i = v_k$ and return to step 1

- Step 2 : define $BS(v_i)$

  for $e_j \subseteq BS(v_i)$

  if there is $v_k \in T(e_j)$ is a known variable

  then $P = \{P, -v_k\}$

  else $v_i = v_k$ , return to step 2

3) Elements in $P$ with corresponding signs are variables of the selected ARR.

This algorithm determines all the B-hyperpath that connect two measurable variables. One may use this algorithm to prove whether two vertices are B-connected in a directed hypergraph.

## V. CASE STUDY: THE SECOND ORDER CHEMICAL REACTION

We present over here this case study to illustrate the performance of the proposed approach.

**5.1 Description of the chemical reaction:**

The case study deals with a reaction which has a second-order reaction rate between acid acetic $C_2H_4O_2$ and ethanol $C_2H_6O$ to produce ethyl acetate $C_4H_8O_2$ and water $H_2O$ . This reaction is represented by:

$$C_2H_4O_2 + C_2H_6O \underset{A_r}{\overset{A_f}{\rightleftharpoons}} C_4H_8O_2 + H_2O \qquad (12)$$

It is a widely used reaction in engineering laboratories because it illustrates the theory of reactor very easily. In the sequel $A$ , $B$ , $C$ and $D$ stand for acid acetic, ethanol,ethyl acetate and water [26]. This reaction can be written in the general form as:

$$\upsilon_A A + \upsilon_B B \underset{A_r}{\overset{A_f}{\rightleftharpoons}} \upsilon_C C + \upsilon_D D \qquad (13)$$

where $\upsilon_i$ (for $i = A, B, C, D$ ) are the stoichiometric coefficients. In our case these coefficients are equal to one because the esterification is one-to-one reaction.

**5.2 Chemical reaction representations:**

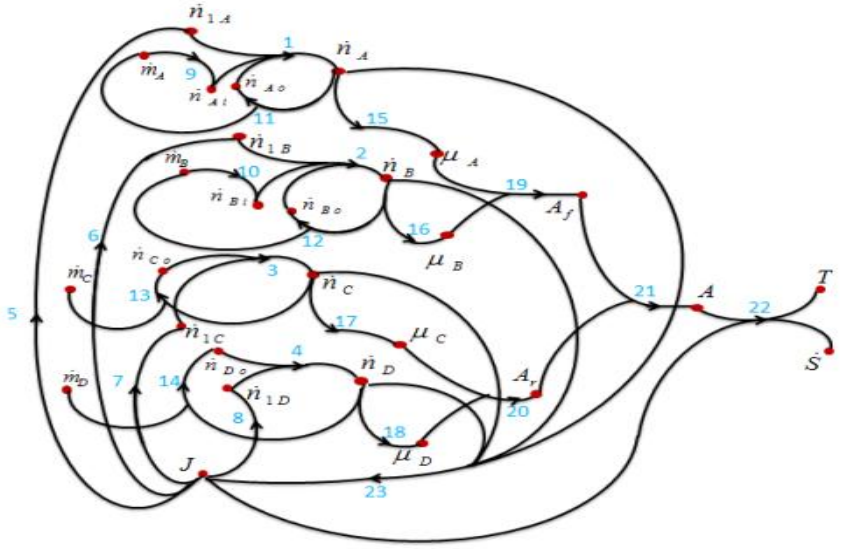

**Figure 1** directed hypergraph model $H$

Fig. (1) presents the directed hypergraph model $H = (V, E)$ where :

$$V = \{\dot{n}_{1A}, \dot{m}_A, \dot{n}_{Ai}, \dot{n}_{Ao}, \dot{n}_A, \dot{n}_{1B}, \dot{m}_B, \dot{n}_{Bi}, \dot{n}_{Bo}, \dot{n}_B, \dot{n}_{1C}, \dot{m}_C, \dot{n}_{Co}, \dot{n}_C,$$
$$\dot{n}_{1D}, \dot{m}_D, \dot{n}_{Do}, \dot{n}_D, \mu_A, \mu_B, \mu_C, \mu_D, A_f, A_r, A, J, T, \dot{S}\}$$

and $E = \{E_1, E_2, \ldots, E_{23}\}$ :

$$E_1 = (\{\dot{n}_{1A}, \dot{n}_{Ai}, \dot{n}_{Ao}\}, \{\dot{n}_A\}), E_2 = (\{\dot{n}_{1B}, \dot{n}_{Bi}, \dot{n}_{Bo}\}, \{\dot{n}_B\}), E_3 = (\{\dot{n}_{1C}, \dot{n}_{Co}\}, \{\dot{n}_C\})$$

$$E_4 = (\{\dot{n}_{1D}, \dot{n}_{Do}\}, \{\dot{n}_D\}), E_5 = (\{J\}, \{\dot{n}_{1A}\}), E_6 = (\{J\}, \{\dot{n}_{1B}\}), E_7 = (\{J\}, \{\dot{n}_{1C}\}),$$

$$E_8 = (\{J\}, \{\dot{n}_{1D}\}), E_9 = (\{\dot{m}_A\}, \{\dot{n}_{Ai}\}), E_{10} = (\{\dot{m}_B\}, \{\dot{n}_{Bi}\}), E_{11} = (\{\dot{m}_A, \dot{n}_A\}, \{\dot{n}_{Ao}\}),$$

$$E_{12} = (\{\dot{m}_B, \dot{n}_B\}, \{\dot{n}_{Bo}\}), E_{13} = (\{\dot{m}_C, \dot{n}_C\}, \{\dot{n}_{Co}\}), E_{14} = (\{\dot{m}_D, \dot{n}_D\}, \{\dot{n}_{Do}\}),$$

$$E_{15} = (\{\dot{n}_A\}, \{\mu_A\}), E_{16} = (\{\dot{n}_B\}, \{\mu_B\}), E_{17} = (\{\dot{n}_C\}, \{\mu_C\}), E_{18} = (\{\dot{n}_D\}, \{\mu_D\}),$$

$$E_{19} = (\{\mu_A, \mu_B\}, \{A_f\}), E_{20} = (\{\mu_C, \mu_D\}, \{A_r\}), E_{21} = (\{A_f, A_r\}, \{A\}),$$

$$E_{22} = (\{A, J\}, \{\dot{S}\}), E_{23} = (\{\dot{n}_A, \dot{n}_B, \dot{n}_C, \dot{n}_D\}, \{J\})$$

The corresponding incidence matrix $B$ of the generated dirhypergraph $H$ is defined as follows :

$$b_{ij} = \begin{cases} -1 & if \quad v_i \in T(E_j) \\ 1 & if \quad v_i \in H(E_j) \\ 0 & otherwise \end{cases} \qquad (14)$$

The BGs have been successfully used for modeling different kinds of systems involving multiphysical phenomena [24]. Hence, an easier understand of the overall system.
Fig. (2) presents the BG model of the considered esterification [26], [27].

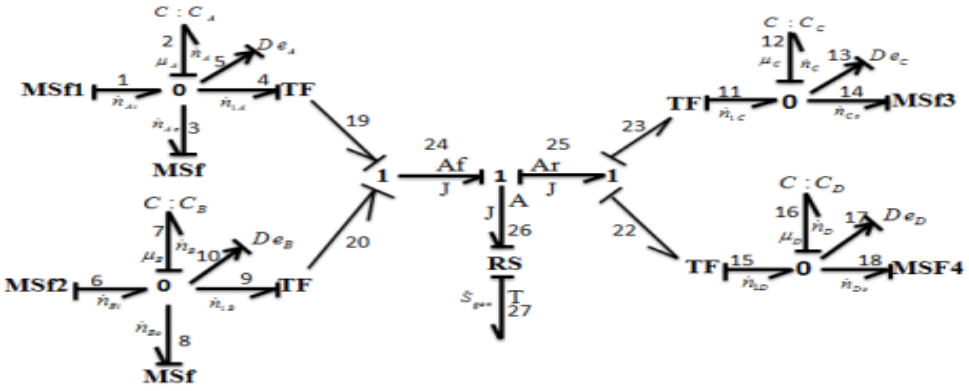


**Figure 2** bond graph model

Based on BG-hypergraph analog [21] the automatically generation of the hypergraph model $H_{BG}$ of the chemical kinetics is deduced directly from the Bond graph model as shown in Fig. (3).

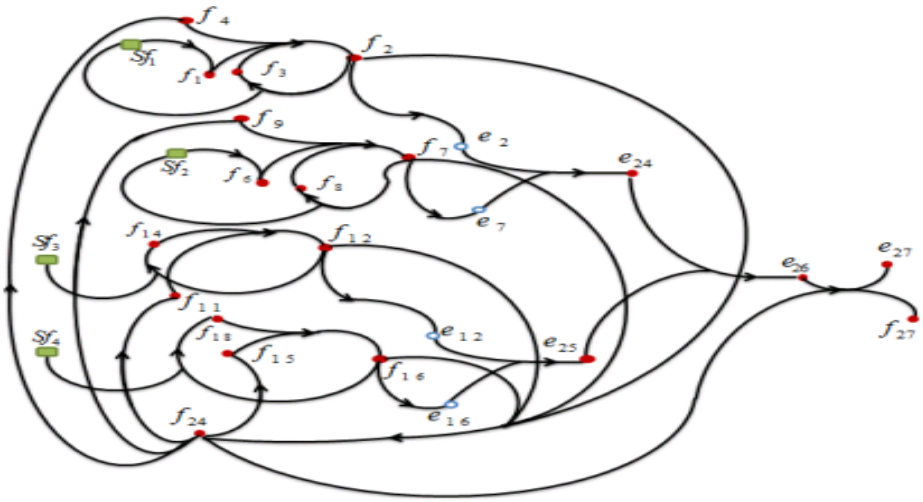

**Figure 3** deduced hypergraph model

One may easily verify even the hypergraph $H_{BG}$ presents the same incidence matrix. Thus, both of the hypergraphs model represents the same chemical reaction kinetics.

**5. 3 Fault Monitoring and main results:**

The chemical reaction Bond graph model in derivative causality is given (Fig. (4).

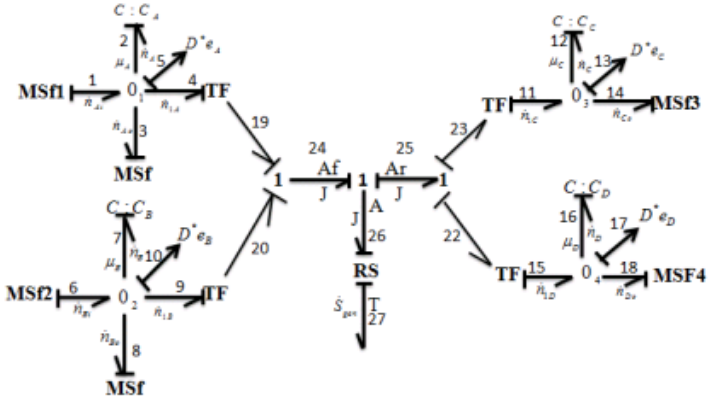


**Figure 4** chemical reaction BG model in derivative causality

There are four sensors elements, so four measurements constraints can be derived. First consider junction $0_1$, the corresponding constitutive relation is:

$$f_1 - f_2 - f_3 - f_4 = 0 \tag{15}$$

Where $f_1 = MSf = \dot{n}_{Ai} = \frac{\dot{m}_A}{M_A}$, $f_2 = C_A \frac{de_2}{dt}$, $f_3 = MSf = \dot{n}_{Ao} = n_A \frac{\dot{m}_A}{\rho V}$, $f_4 = \upsilon_A J$

The unknown variable $f_2$ is eliminated by covering the causal path:

$$f_2 \rightarrow \phi_{C_A} \rightarrow e_2 \rightarrow \tilde{D}e_A : \mu_A$$

Finally, the first ARR is then deduced substituting unknown variable in Eq.(\ref{ARR1}):

$$ARR_1 = \dot{n}_{Ai} - \frac{V}{RT_r}\exp\left(\frac{\mu_A - \mu_{A0}}{RT}\right) - n_A \frac{\dot{m}_A}{\rho V} - \upsilon_A J$$

$$= \frac{\dot{m}_{Ai}}{M_A} - \frac{V}{RT_r}\exp\left(\frac{\mu_A - \mu_{A0}}{RT_r}\right) - n_A \frac{\dot{m}_A}{\rho V} - \upsilon_A r_f \left(1 - \exp\left(\frac{A}{RT_r}\right)\right)V \tag{16}$$

$$= 0$$

$\mu_{A0}$ is the standard chemical potential.

From junction $0_2$, we obtain :

$$ARR_2 = \dot{n}_{Bi} - \frac{V}{RT_r}\exp\left(\frac{\mu_B - \mu_{B0}}{RT}\right) - n_B \frac{\dot{m}_B}{\rho V} - \upsilon_B J$$

$$= \frac{\dot{m}_{Bi}}{M_B} - \frac{V}{RT_r}\exp\left(\frac{\mu_B - \mu_{B0}}{RT_r}\right) - n_B \frac{\dot{m}_B}{\rho V} - \upsilon_B r_f \left(1 - \exp\left(\frac{A}{RT_r}\right)\right)V \tag{17}$$

$$= 0$$

From junction $0_3$, we have :

$$f_{11} - f_{12} - f_{14} = 0 \tag{18}$$

Where $f_{11} = \upsilon_C J = \upsilon_C r_f \left(1 - \exp\left(\frac{A}{RT_r}\right)\right)V$ , $f_{12} = C_C \frac{de_{12}}{dt}$ and $f_{14} = MSf3 = \dot{n}_{Co} = n_C \frac{\dot{m}_C}{\rho V}$

The unknown variable $f_{12}$ is deduced from the causal path:

$$f_{12} \rightarrow \phi_{C_C} \rightarrow e_{12} \rightarrow \tilde{D}e_C : \mu_C$$

Thus the third ARR can be obtained:

$$ARR_3 = \upsilon_C r_f \left(1 - \exp\left(\frac{A}{RT_r}\right)\right)V - n_C \frac{\dot{m}_C}{\rho V} - \frac{V}{RT_r}\exp\left(\frac{\mu_C - \mu_{C0}}{RT_r}\right) \tag{19}$$

$$= 0$$

And from junction $0_4$, we got :

$$ARR_4 = \upsilon_D r_f \left(1 - \exp\left(\frac{A}{RT_r}\right)\right)V - n_D \frac{\dot{m}_D}{\rho V} - \frac{V}{RT_r}\exp\left(\frac{\mu_D - \mu_{D0}}{RT_r}\right) \tag{20}$$

$$= 0$$

Table (I) gives the fault signature matrix of the studied chemical reaction:

**Tableau 1** FSM deduced from the BG model

| | ARR1 | ARR2 | ARR 3 | ARR4 | D | I |
|---|---|---|---|---|---|---|
| $\dot{m}_A$ | -1 | 0 | 0 | 0 | 1 | 1 |
| $\dot{m}_B$ | 0 | -1 | 0 | 0 | 1 | 1 |
| $\dot{m}_C$ | 0 | 0 | -1 | 0 | 1 | 1 |
| $\dot{m}_D$ | 0 | 0 | 0 | -1 | 1 | 1 |
| $\mu_A$ | 1 | 0 | 0 | 0 | 1 | 1 |
| $\mu_B$ | 0 | 1 | 0 | 0 | 1 | 1 |
| $\mu_C$ | 0 | 0 | 1 | 0 | 1 | 1 |
| $\mu_D$ | 0 | 0 | 0 | 1 | 1 | 1 |

The hypergraph model of the studied chemical reaction is given in Fig. (1) The set of unknown variables is $K = \{\dot{n}_A, \dot{n}_B, \dot{n}_C, \dot{n}_D\}$ .

Step 1: $FS(\dot{n}_A) = \{e_{15}, e_{23}\}$ , for $e_{15}$ , the chemical potential $\mu_A$ is the known variable in $H(e_{15})$ thus $P = \{\mu_A\}$

$BS(\dot{n}_A) = \{e_1\}$ , the known variable in $T(e_1)$ is the molar flow $\dot{m}_A$ and $P_1 = \{\mu_A, -\dot{m}_A\}$ . We proceed in the same manner for the others variables in $K$ . We obtain $P_2 = \{\mu_B, -\dot{m}_B\}$ , $\$P_3 = \{\mu_C, -\dot{m}_C\}$ , $P_4 = \{\mu_D, -\dot{m}_D\}$ .The obtained result is summarized in the following fault signature matrix (Table (II))

**Tableau 2** FSM deduced from the hypergraph model

| | QRR1 | QRR2 | QRR 3 | QRR4 | D | I |
|---|---|---|---|---|---|---|
| $\dot{m}_A$ | -1 | 0 | 0 | 0 | 1 | 1 |
| $\dot{m}_B$ | 0 | -1 | 0 | 0 | 1 | 1 |
| $\dot{m}_C$ | 0 | 0 | -1 | 0 | 1 | 1 |
| $\dot{m}_D$ | 0 | 0 | 0 | -1 | 1 | 1 |
| $\mu_A$ | 1 | 0 | 0 | 0 | 1 | 1 |
| $\mu_B$ | 0 | 1 | 0 | 0 | 1 | 1 |
| $\mu_C$ | 0 | 0 | 1 | 0 | 1 | 1 |
| $\mu_D$ | 0 | 0 | 0 | 1 | 1 | 1 |

## VI. DISCUSSION

This case study first highlights the efficiency of the proposed analogy BG-Hypergraph transformations. The hypergraph model is then built on the bases of the process knowledge using the Bond Graph model developed from deep physical understanding of the complex system, thus avoiding the complexity of numerical calculations.

It shows also that BG-based FDI method provides an intuitive and direct way to derive ARRs from the BG model by eliminating unknown variables of the constitutive relations of every junction using causal paths of the graph. However, it requires that all storage elements on the graph to be assigned in derivative causality. Also the BG model requires a detailed mathematical description of the process dynamic, which may not be always possible. We may derive the hypergraph model even from the BG model in integral causality. The proposed hypergraph-based FDI shows its effectiveness through the case study. We obtain the same signature matrix using both BG and hypergraph-based FDI approches. Moreover, in an other example, not shown here, the total number of ARRs derived by this method will be reduced compared by those generated usin the BG-based FDI.

## VII. CONCLUSION

This paper has applied directed hypergraph for diagnosis of chemical reactions kinetics and developed algorithm for ARR generation. As B-hyperpath in hypergraphs generalize shortest path in digraphs, the proposed algorithm can be summarized as follow: from a unknown variable in the hypergraph model, in a first stage we follow the B-hyperpath that relates it to a measurable variable then we use the back-propagation from the selected unknown variable to a measurable variable. Known variables in the path are elements of the ARR.
The effectiveness of the proposed algorithm has been fulfilled through the example of a reaction which has a second-order reaction rate between acid acetic ($C_2H_4O_2$) and ethanol ($C_2H_6O$) to produce ethyl acetate ($C_4H_8O_2$) and water ($H_2O$).

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