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Alternative method to prevent thermal runaway in case of error on operating conditions continuous reactor

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ABSTRACT

Thermal runaway was studied in a continuous tubular pilot reactor under steady-state regime. Different accident scenarii were conducted by making some errors on reactant concentrations and/or temperature feed. To prevent thermal runaway, control by direct contact by solvent injection was used at different reactor locations. This injection allowed controlling the maximum reaction temperature. A simplified analytical method to estimate the maximum reaction temperature along the reactor was used.

Benefit of this control method was the diminution of computational time. Furthermore, by injecting solvent to control maximum reaction temperature, there is no need to shut down the unit. The control method was validated experimentally.

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1. Introduction

Chemical reactors are the heart of chemical processes. In case of cooling failure or malfunction, heat released by chemical reactions cannot be absorbed by the cooling system. Consequently, thermal runaway phenomenon can occur (Stoessel, 2008).

Thermal safety of chemical processes can be complex to evaluate. One should have strong advanced of chemical reaction engineering and of thermal analysis. Besides, one should develop some global approach when kinetics and thermodynamics are unknown. The first step is to differentiate thermal risk assessment for batch or semi-batch than from tubular or continuous stirred tank reactor (CSTR). In case of batch system, there is a thermal accumulation, which could lead

to a thermal runaway accident (Leveneur et al., 2012). Preliminary approach to evaluate the thermal risk of a chemical reaction without the knowledge of its kinetics was described in different articles or books (Stoessel, 2008; Leveneur et al., 2012, 2014; Guinand et al., 2014). The first step is to determine the total energy released by the chemical system. For that, Differential Scanning Calorimeter (DSC) is used to calculate the adiabatic temperature rise. Then, Time-to-Maximum-Rate under adiabatic condition (TMR_{ad}) should be measured by using an adiabatic reactor. These two safety criteria represent the severity and probability of the thermal risks. This methodology is adequate for a preliminary approach, but it is for a particular kinetic. Is this methodology still correct if initial operating conditions like reactant concentration or temperature change?

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Nomenclature

A_0	frequency factor
C	concentration (mol/m ³)
C_p	specific heat (J/kg K)
D	diameter (m)
e	thickness (m)
E	activation energy (J/mol)
h	convective heat transfer coefficient (W/m ² K)
ΔH	reaction enthalpy (J/mol)
L	length (m)
Δl	portion of the reactor length (m)
Pe	Peclet number
Q	mass flow rate (kg/s)
Q	volumic flow rate (m ³ /s)
r	Reaction rate (mol/m ³ s)
R	perfect gas constant (J/(mol K))
Re	Reynolds number
S_l	side surface (m)
T	temperature (°C)
V	mean velocity (m/s)
Vol	volume (m ³)
x	axial position (m)
U	global heat transfer coefficient (W/m ² K)

Greek symbol

α	reaction parameter
β	reaction parameter
Φ	heat-flow rate (J/s)
λ	thermal conductivity (W/m K)
ν	stoichiometric coefficient
ρ	density (kg/m ³)

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A	hydrogen peroxide
B	sodium thiosulfate
c	cooling fluid
in	inlet
inj	injection
out	output
max	maximum
r	reaction mixture
w	wall

Thermal runaway could occur in case of operator error due to wrong reactant concentrations or feeding temperature (Saada et al., 2015). This error could increase reaction rate, thus, the heat-flow rate released by chemical reaction, and making the initial scale-up non-adequate. The consequences of such error are the increase of the reaction temperature leading to thermal runaway and/or to product degradation. The maximum temperature value is a parameter to take into account during a thermal risk assessment.

Literature concerning the thermal safety assessment for batch reactor is vast. Different authors have developed some simplified methods to determine the thermal risk of a reaction or have developed some safety criteria (Westerterp and Molga, 2004; Bosch et al., 2004; Maestri and Rota, 2006). Literature review for thermal risk assessment for continuous reactor is rare (Morais et al., 2004; Schweitzer et al., 2010; Vernières-Hassimi et al., 2008, 2012, 2014a; Théron et al., 2014).

One should keep in mind that the coupling of mass and energy balance involves the resolution of complex differential equations. This resolution is time consuming and could increase the time to action in case of runaway. Thus, it is important to simplify this energy balance equation to an analytical expression mostly in case of fast reaction rate.

In this manuscript, a pilot was used to carry out some experiments with operator errors on inlet conditions. These errors have led to an increase of the reaction temperature. To control the temperature, solvent, i.e., water, was directly injected in the reaction mixture. To avoid long numerical resolution, the injected solvent flow rate was calculated based on a simplified analytical expression (Vernières-Hassimi et al., 2014b). This analytical expression proposed a linear relation between the maximum reaction temperature and the inlet/outlet conditions. This approach was validated experimentally in the case of errors on inlet condition.

2. Experimental set-up description and mathematical modelling

2.1. Experimental set-up description

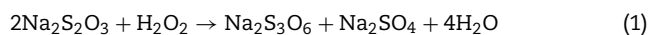
Fig. 1 illustrates the reactor setup. The total length of the tubular reactor is of 8.6 m, divided into sections of 0.8 m each. The internal diameter is of 1.84×10^{-2} m and the external diameter is of 3×10^{-2} m (Table 1). Reaction mixture circulates in the tubular part of the reactor, and heat carrier (cooling fluid) circulates in the opposite direction in the annular section (countercurrent-flow).

The solvent injection system can inject water at the inlet or at 1.5 m of the reactor.

2.2. Mathematical modelling

2.2.1. Chemical system

Oxidation of sodium thiosulfate by hydrogen peroxide was the reaction model because of its fast reaction rate and high reaction enthalpy, making it a model for safety studies (Cohen and Spencer, 1962; Lo and Cholette, 1972; Brungs et al., 1988; Aimé, 1991; Grau et al., 2000; Grau et al., 2002). When the ratio hydrogen peroxide concentration C_A on sodium thiosulfate concentration C_B is higher or equal to 1.96 (Aimé, 1991), chemical equation is



Kinetic expression of this reaction is

$$r = A_0 \exp\left(-\frac{E}{RT}\right) C_A^\alpha C_B^\beta \quad (2)$$

Kinetic data and reaction enthalpy ΔH determined by (Aimé, 1991) when the ratio of hydrogen peroxide concentration C_A on sodium thiosulfate C_B concentration is higher or equal to 1.96 are shown in Table 2:

Reaction orders are 1.5 in hydrogen peroxide and 0.6 in sodium thiosulfate. The overall reaction order is 2.1.

2.2.2. Mathematical modelling of the chemical reactor

To establish the chemical reactor model, the following assumptions were done:

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