Simulation and control of water-gas shift packed bed reactor with inter-stage cooling

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Abstract. Water-Gas Shift Reaction (WGSR) has become one of the well-known pathways for $H_2$ production in industries. The issue with WGSR is that it is kinetically favored at high temperatures but thermodynamically favored at low temperatures, thus requiring careful consideration in the control design in order to ensure that the temperature used does not deactivate the catalyst. This paper studies the effect of a reactor arrangement with an inter-stage cooling implemented in the packed bed reactor to look at its effect on outlet temperature. A mathematical model is developed based on one-dimensional heat and mass transfers which incorporate the intra-particle effects. It is shown that the placement of the inter-stage cooling and the outlet temperature exiting the inter-stage cooling have strong influence on the reaction conversion. Several control strategies are explored for the process. It is shown that a feedback-feedforward control strategy using Multi-scale Control (MSC) is effective to regulate the reactor temperature profile which is critical to maintaining the catalysts activity.

1. Introduction
The potential of Hydrogen is being widely examined as a possible energy carrier because of its high energy content and environmental compatibility. Hydrogen can be produced from a wide range of primary energy sources and pathways. Presently, Water-Gas Shift Reaction (WGSR) represents an attractive pathway for H2 production and this reaction is currently used in many important industrial applications [1].

The WGSR is a well-known and extensively studied reaction, which is a mildly exothermic and equilibrium limited reaction [2] shown in equation (1):

$$CO + H_2O \rightleftharpoons H_2 + CO_2$$ (1)

The equilibrium of the WGSR favours low temperatures and is unaffected by the operating pressure. In the first stage of the industrial-scale process units, a high temperature (HT-WGS) packed bed reactor is used to exploit fast kinetic rates. The outlet stream of the HT-WGS reactor is cooled and fed to low temperature (LT-WGS) packed bed reactor where the temperature is beneficial towards the equilibrium henceforth leading to a high CO conversion [3] (see figure 1).

The goal of the present study is to find an answer to a question: is it possible to achieve higher CO conversion with only one stage (HT-WGS) packed bed reactor and moreover, to

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design a simple control strategy to control the reactor to maintain its optimum performance. Furthermore, since the WGSR is exothermic in nature, the reaction is often conducted in adiabatic shift reactors to avoid catalyst overheating as well as to improve the reaction conversion [4]. Then again, an excessively high temperature can cause the catalysts activity to degenerate and in turn leads to a poor conversion of CO. For this reason, an effective temperature control of the reactor in which the WGSR takes place is very important to avoid serious catalyst deactivation.

The WGSR has been extensively studied in several works, both experimentally and using numerical simulations. Simulation is a very useful tool for predicting and studying these reactors, but the results obtained are highly dependent on the accuracy of the models used [5]. Most models proposed in the literature, (e.g. [6], [7] and [8]), often disregard the influence of intra-particle mass and heat transfer effects, although they can be important because of fast kinetics of this reaction.

With respect to control strategies in a packed bed tubular reactor (PBTR) where complex reactions such as the WGSR takes place, a single-loop controller is hardly effective for controlling the reactors effluent temperature due to significant transport delay along the reactor. In this case, a feed-forward control strategy could be employed to reject the disturbance before it can greatly upset the performance of the reactor. Even so, feedforward controller only rejects a specific measured disturbance and not all of the disturbances that occur in a process. For this reason, it is a common in industry that the feedforward controller is used together with feedback control system against certain disturbances [9]. Interestingly, both the feedback and feedforward controllers can be effectively designed based on the recently proposed multi-scale control (MSC) scheme in [10].

In this study, we focus on one stage (HT-WGS) packed bed reactor with inter-stage cooling. A rigorous dynamic distributed model is developed based on the first principles that integrate both the intra-particle heat and mass transfers. In addition, three control strategies (i.e., single loop control, conventional feedforward and feedback control, and MSC feedforward and feedback control schemes) will be implemented to regulate the reactor temperature at a selected location of the WGS reactor to achieve: (1) high CO conversion and (2) to suppress the temperature from reaching the threshold value of catalyst deactivation temperature.

The paper is structured as follows: the next section (section 2) includes the WGSR kinetics and HT-WGS packed bed tubular reactor (PBTR) modelling. Section 3 presents the formulation of control strategies. Section 4 provides the simulation results and discussion. Finally, Section 5 highlights some conclusions and future research directions.

2. WGSR modelling

2.1. WGSR kinetics

The detailed explanation on kinetics and reactor modelling of WGSR can be found in [1]. Here, we only use Power Law (PL) model to describe the kinetics as it is the simplest empirical rate expression, which disdain any mechanism such as redox or Langmuir-Hinshelwood (LH). Likewise, several researchers have been reported using this kinetic model for high temperature
WGSR, e.g., [11] and [12]. The PL model is given by:

\[-r_A = k_0 e^{-E_a/RT} N_{CO}^a N_{H_2O}^b N_{CO_2}^c N_{H_2}^d (1 - \beta)\]  

(2)

where, \( r_A \) (mol/g.hr) is the rate of reaction, \( N_i \) (mol/hr) is the corresponding molar flow rate of component \( i \) whereas, \( a, b, c, \) and \( d \) are the apparent reaction orders of component \( CO, H_2O, CO_2, \) and \( H_2 \), respectively; \( k_0 \) is the pre-exponential factor, \( E_a \) is the activation energy (J/mol), \( R \) is the universal gas constant (8.314 J/mol.K), and \( T \) is the reaction temperature (K). is the term for backward reaction and is defined as follows:

\[\beta = \frac{N_{CO_2} N_{H_2}}{K_P N_{CO} N_{H_2O}}\]  

(3)

where, \( K_P \) is an equilibrium constant for WGS reaction and the equation for \( K_P \) calculation is as below:

\[\ln K_P = \left( \frac{4577.8}{T} \right) - 4.33\]  

(4)

The dimension of the PBTR system used in this study is scaled to 1 (one) metre in length with additional 0.1m for inter-stage cooling and 16cm in diameter (\( D_t \)) based on the pilot-scale membrane reactor commercialized by [13]. For the PBTR modelling, \( Fe_3O_4-Cr_2O_3 \) catalyst is used with its property as reported in [5]. For the purpose of dynamic modelling, the length of the reactor is sub-divided into eleven equal parts. One of the parts is the inter-stage cooling as shown in figure 2 where \( L_1 \) and \( L_3 \) are subdivided into 5 equal lengths of 0.1m. \( L_2 \) represents the inter-stage cooling in-between the reactor. Figure 3 shows the cross section of the inter-stage cooling where the tube is arranged in triangular (30°) pattern as this pattern manages to produce high turbulence and consequently results in high heat transfer coefficient [14].

2.2. Packed bed tubular reactor model with inter-stage cooling

In order to estimate the outlet temperature of the reactor, a theoretical numerical model is used for modelling the PBTR. This model consists of mass and energy balance equations with the incorporation of intra-particle mass and heat transfers. The assumptions that govern these equations are: (1) only the axial temperature change is taken into account, (2) the radial temperature change is neglected due to the fact that the diameter of the reactor is small and adiabatic (no heat loss from the surface of the reactor); see [15], (3) momentum conservation equation has been disregarded in this model because the bed height is too small to create a significance amount of pressure drop, i.e., see [16], (4) no reaction in the inter-stage cooling, and (5) inter-stage cooling consists of several small tubes therefore radial temperature change

**Figure 2.** Division of the packed bed tubular reactor into 11 sub-sections.

**Figure 3.** Cross-sectional diagram of the inter-stage cooling \( L_2 \) in PBTR.
Equations (5) - (7) show the mass and energy balance equations for the PBTR system.

\[ \epsilon V \frac{dC_{i,j}}{dt} = F(C_{i,j-1} - C_{i,j}) \pm r_{A,j} \eta_{gl} W \] (5)

\[ (1 - \epsilon)V \rho_s C_{p,s} \frac{dT_{s,j}}{dt} = r_{A,j} \eta_{gl} W \Delta H_r - U A(T_{s,j} - T_{f,j}) \] (6)

\[ \epsilon V \rho_f C_{p,f} \frac{dT_{f,j}}{dt} = FC_{p,f} \rho_f (T_{f,j-1} - T_{f,j}) + U A(T_{f,j} - T_{f,j}) \] (7)

Here, \( \epsilon \) denotes the bed voidage factor, \( V \) (m³) the sub-section volume of the reactor, \( C_{i,j} \) (mol/m³) the molar concentration of component \( i \) at the j-th sub-section, \( F \) (m³/hr) the feed flow rate, \( W \) (g) the weight of catalyst in the reactor sub-section, \( \rho_s \) and \( \rho_f \) (kg/m³) the density of catalyst and fluid respectively, \( C_{p,s} \) and \( C_{p,f} \) (kJ/kg.K) the specific heat capacity of catalyst and fluid, \( T_{s,j} \) (K) the catalyst temperature at j-th sub-section, \( T_{f,j} \) (K) the fluid temperature at j-th sub-section, \( \Delta H_r \) (kJ/mol) the heat of reaction, \( U \) (kJ/hr.m².K) the convective overall heat transfer coefficient, \( A \) (m²) the heat transfer area and \( \eta_{gl} \) is the global effectiveness factor.

In equation (5), the \( \pm \) sign indicates either species consumption or generation; for \( CO \) and \( H_2O \) the sign is minus, while for the products the sign is plus. As for the inter-stage cooling, the temperature profile of the coolant and the feed will be calculated based on energy balance model as follows:

\[ \epsilon V \rho_f C_{p,f} \frac{dT_{f,j}}{dt} = FC_{p,f} \rho_f (T_{f,j-1} - T_{f,j}) - h A_t (T_{f,j} - T_{c,out}) \] (8)

\[ V_{c} \rho_c C_{p,c} \frac{dT_{c,out}}{dt} = F_c C_{p,c} \rho_c (T_{c,in} - T_{c,out}) + h A_t (T_{f,j} - T_{c,out}) - F_c \rho_c L_c \] (9)

where all the subscript \( c \) stands for coolant, \( h \) (kJ/hr.m².K) is the overall heat transfer coefficient, \( A_t \) (m²) is the area of heat transfer in the inter-stage cooling and \( L_c \) (kJ/kg) is the latent heat of vaporization. The \( h \) is calculated based on Gnielinski correlation [17] and Sieder and Tate correlation [18].

The global effectiveness factor takes into account both mass transfer resistances, internal and external to the catalyst particles. In the global effectiveness calculation, the effective diffusion coefficient used is approximated by Knudsen diffusion in accordance with the physical properties of the gas compounds and textural properties of the catalyst. Meanwhile, the mass transfer resistance external to the catalyst particle is modelled using the mass transfer coefficient for transport in packed bed and the mass transfer resistance internal to the catalyst particle is modelled using generalized Thiele modulus. The internal effectiveness factor is used to model the contribution of the mass transfer resistance inside the porous catalyst particles. The expression of the internal effectiveness factor is suitable for a catalyst in the form of pellets and expression of the used in this equation can deal with the reversible power-law type kinetic equation. The equations mentioned above are written as follows.

\[ \eta_{int} = \frac{1}{\Phi} \left( \frac{1}{\tanh(3\Phi)} - \frac{1}{3\Phi} \right) \] (10)

\[ \Phi = \frac{d_p/4 \rho_s r_{A,j}}{\sqrt{2D_{ep}\rho_s C_{eq} r_{A,j} dC_i}} \] (11)

\[ D_{ep} = \frac{d_{pore} \epsilon_{int} 4}{\tau} \frac{RT}{3 \Pi M} \] (12)
Table 1. Fixed numerical parameters.

| Name                                      | Numerical Parameters | Reference |
|-------------------------------------------|----------------------|-----------|
| Length of reactor, \(L\)                 | 1 m                  | [13]      |
| Diameter of reactor, \(D_t\)             | 16 cm                | [13]      |
| Catalyst density, \(\rho_s\)             | 1945 kg/m^3          | [5]       |
| Catalyst diameter, \(d_p\)               | \(2 \times 10^{-3}\) m | [5]       |
| Catalyst pore diameter, \(d_{pore}\)     | \(9 \times 10^{-9}\) m | [5]       |
| Catalyst bed porosity, \(\epsilon\)      | 0.4                  | [5]       |
| Catalyst tortuosity, \(\tau\)            | 4                    | [5]       |
| Heat of reaction, \(\Delta H_r\)         | -41.1 kJ/mol         | [3]       |
| Feed flowrate                             | 16 m^3/hr            | [3]       |
| Steam/Carbon (S/C) ratio                  | 2.4                  | [3]       |
| Inlet temperature, \(T_{in}\)            | 873 K                | [3]       |
| Coolant inlet temperature, \(T_{c,in}\)  | 300 K                | [3]       |

\[a_s K_G = \frac{6 D_{COm}}{d_p^2} (2 + 1.1 Re^{0.60} Sc^{0.33})\]  
\[\eta_{gl} = \frac{1}{\eta_{gl} + \frac{\rho_s}{a_s K_G} \left( \frac{\partial a_s}{\partial CO} \right)}\]  

where \(\Phi\) denotes the generalized Thiele modulus, \(d_p\) (m) is the diameter of the catalyst particle, \(D_{ep}\) is the pore effective diffusion coefficient, \(a_s K_G\) denotes the mass transfer coefficient, \(d_{pore}\) (m) is the diameter of the catalyst pore size, \(\epsilon_{int}\) is the internal porosity, \(\tau\) is the tortuosity, \(M\) (kg/mol) is molar weight, \(D_{COm}\) (m^2/s) is molecular diffusion of CO in the reaction mixture, \(Re\) is Reynolds number and \(Sc\) is Schmidt number.

The PBTR model is solved using MATLAB (v. 2013b) software. Not to mention that the software has been used to model packed bed reactors with satisfactory results in previous works [19]. Notice that in this work we sub-divide the reactor length into 11 sub-sections of equal lengths to capture the effects of temperature and concentration distributions along the reactor (the actual model should be in PDE form). Hence, a set of ordinary differential equations (ODEs) is used instead of partial differential equations (PDEs). All the numerical parameters used in this simulation are listed in Table 1.

3. Control strategies

3.1. Single loop feedback control strategy

Single loop control strategy consists of a primary process \(P_1\) and a controller \(G_{c1}\) in a single closed loop. The controller for this single loop strategy is tuned based on the model obtained by linearizing the nonlinear PBTR described in equations (5) - (7). The transfer function is obtained through linearizing the control variable against the manipulated variable which in this case is the flow rate of the inter-stage coolant. Equation (15) shows the transfer function obtained from linearizing the abovementioned variables.

\[H(s) = \frac{Y_1(s)}{R(s)} = \frac{G_{c1} P_1}{1 + G_{c1} P_1}\]
3.2. Generalized feedback-feedforward control strategy

Figure 4 depicts the block diagram of a conventional feedback plus feedforward control structure commonly used in process industry. Here, \( G_c \) is the feedback controller; \( F \) the feedforward controller.

In figure 4, the feedforward controller \( F \) is derived by inverting the nominal model \( P_1 \) with an assumption that, the output disturbance effect via \( G_d \) is cancelled out by the feedforward controller (ideal design), thus:

\[
[F(s)P_1(s) + G_d(s)]D(s) = 0
\]  

(16)

So, the ideal feedforward controller \( F \) is obtained as:

\[
F(s) = -\frac{G_d(s)}{P_1(s)}
\]  

(17)

3.3. MSC feedforward-feedback control strategy

Figure 5 illustrate the block diagram of the proposed feedback-feedforward control structure based on the multi-scale control (MSC) scheme [9]. The feature that distinguishes this MSC-based feedback-feedforward (MSC FB-FF) control structure from the conventional one (figure 4) is that the MSC-FB/FF treats the measured disturbance \( D \) as though it is an input disturbance introduced directly into the fast inner-loop of the MSC control system. In this way, the effect of disturbance \( D \) on \( Y \) is mainly removed by the fast inner-loop sub-controller \( K_1 \).

The details of the MSC-FB/FF control strategy can be obtained from [9]. In figure 4, the transfer function from \( U_{ff} \) to \( U \) is given by

\[
H_{fu}(s) = \frac{1}{1 + K_1W_1(s)}
\]  

(18)

where \( K_1 \) is the inner-layer sub-controller which is often chosen as a P-only controller because this leads to simple controller tuning and \( W_1 \) is a multi-scale predictor. Meanwhile, a PI controller is often chosen for the outermost sub-controller \( K_0 \); an integral action is added to remove steady-state offset in the output.

The MSC FB-FF structure (figure 5) can be reduced to the same structure as that of the conventional single-loop feedback-feedforward control. One can derive an equivalent MSC-based feedforward controller as follows:

\[
F_{msc}(s) = H_{fu}(s)F_{ss}^*(s)
\]  

(19)
Equation (19), $F_{ss}$ denotes the optimised static feedforward controller while $F_{msc}$ denotes the MSC-based feedforward controller, i.e., the subscript msc is used to differentiate it from the conventional feedforward controller in figure 4. The formula for obtaining static the feedforward is obtained via the proposed method in [9].

4. Results and discussion

4.1. Numerical parameters and model verification

The fixed numerical parameters used in this study are listed in Table 1. In Table 1, catalyst bed properties and reaction kinetic parameters are taken from the study of [5]. The operating conditions are based on the study of [3]. As shown in [20] which is the extended study from [5], the numerical model was verified with the experimental data in [20]. Using the data listed in Table 1, the numerical model was verified again to ensure its validation.

Meanwhile, figure 6 shows the comparison between the numerically predicted CO conversions for the packed-bed reactor with experimental data reported in [1]. As shown in figure 6, the agreement between numerical model and experimental data is good.

4.2. Effects of inter-stage cooling

Based on the verification discussed above, the numerical model is adequately extended to study the PBTR under inter-stage cooling shown in figure 2. Water coolant is used in this work. Using the fixed parameters and base operating conditions listed in Table 1, figure 7 show the reactor conversion comparison between traditional PBTR and PBTR with inter-stage cooling. For traditional PBTR, a maximum conversion is reached at the 5th reactor section and an increase in the length of the reactor does not improve the conversion as WGS is a reversible reaction equilibrium limited.

On the contrary, if the inter-stage cooling is placed right after the maximum conversion is achieved, it can be seen from figure 7 a new maximum conversion can be attained. This inter-stage cooling will not only bring out a better conversion, it will also reduce the risk of catalyst deactivation. The temperature decreases to a lower temperature just before the reactor temperature reaches the catalyst deactivation temperature, which is $T_{deac} = 1200$K for the catalysts used in this work.

![Figure 6](image1.png)

**Figure 6.** Numerical model verification by comparing computed CO conversion with experimental data reported by [21].

![Figure 7](image2.png)

**Figure 7.** Comparison of CO conversion between traditional PBTR and PBTR with inter-stage cooling.
4.3. Effects of inter-stage cooling placement

Further investigation on the effect of placement of the inter-stage cooling on the PBTR performance is also examined. As shown in figure 8, these different sections referred to the placement of $L_2$.

To illustrate, with the inter-stage cooling at section 3, $L_1 = 0.3$m followed by $L_2 = 0.1$m and $L_3 = 0.7$m. Note that, the combination of $L_1$, $L_2$ and $L_3$ should only give a value of 1.1m in total reactor length.

Based on the results shown in figure 8, the placement of inter-stage cooling influences the CO conversion. The maximum CO conversion is obtained when the inter-stage cooling is placed around sections 5 and 6. Implementation of inter-stage cooling at other than 5th and 6th sections were observed to accomplish lower CO conversion but still higher compared to the traditional PBTR. This is because inter-stage cooling placed at section 5 and 6 has already achieved a maximum conversion at the first part of the reactor, $L_1$ with ample of length in $L_3$ reactor for the reaction to occur again.

4.4. Inter-stage cooling outlet temperature effect

The outlet temperature mentioned in this section would be the temperature exiting $L_2$ just before entering reactor $L_3$. Figure 9 shows the impact of outlet temperature of $L_2$ on reactor conversion. The outlet temperature of $L_2$ is set to be manipulated around these three temperatures (i.e. the initial temperature, 1st section, and 2nd section reactor temperature). Figure 9 show that by dropping the outlet temperature of $L_2$ equivalent to the 1st section reactor temperature, it will result in the highest CO conversion. The fastest reaction kinetics can be achieved at this particular temperature.

This shows that the outlet temperature of $L_2$ which also corresponds to the entrance temperature of reactor zone $L_3$ is very important in terms of thermal management. This behaviour of the reactor motivates the need for a control strategy that is able to control and maintain the outlet temperature of $L_2$ in order to achieve maximum conversion. Not to mention, the control strategy must be able to suppress temperature below the maximum catalyst temperature in the presence of disturbances.

**Figure 8.** Comparison of CO conversion between different placements of inter-stage cooling at length $L_3 = 0.4$m to 1m.

**Figure 9.** Comparison of CO conversion between different outlet $L_2$ temperatures for inter-stage cooling at section 3.
### 4.5. Placement of feedback controller

The effect of the placement of feedback controller is explored under the influences of the inlet feed temperature of 10°C (input disturbance source) while the controlling variable is the flow rate of the inter-stage coolant.

It is shown in figure 11 that the placement of the feedback control at different sections of the reactor plays a significant role in disturbance rejection. In this case, the best and the worst placement of the controller is compared and the conclusion drawn is that the controller at sub-section 9 shows 5% of overshoot, while controller at sub-section 6 shows 15% of overshoot.

### 4.6. Feedback control vs. generalized FF-FB control vs. MSC FF-FB control strategy

The performance of a single-loop feedback control scheme is compared with both the conventional FF-FB and MSC FF-FB control strategy scheme under the same type of disturbance with additional disturbance of ratio concentration of $CO/H_2O$.

The feedback controller (primary measurement) is located at sub-section 9 because the placement here shows the best control performance in single-loop control which has been discussed in Section-D. However, the feedforward controller in this work only targeted inlet feed temperature disturbance. In this case study, it is found that conventional feedforward-feedback control scheme show little improvement from feedback control strategy. Therefore, to further improve the control scheme in order to maintain the optimum temperature, multi-scale control (MSC) strategy is implemented into the conventional FF-FB control scheme.

Figure 13 shows the nominal closed-loop response of the outlet fluid temperature at section 9 when the system is subjected to sequential step changes: (1) magnitude of 10 in feed temperature at $t = 2$ and (2) magnitude of 3 in $CO$ concentration at $t = 3$. It can be seen that MSC FF/FB has produced significant improvement over the conventional FF/FB control strategy. The overshoot has been drastically reduced by 350%.

### 5. Conclusion

By introducing an inter-stage cooling in a high-temperature PBTR for WGSR, it can significantly improve the $CO$ conversion. In this paper, it is shown that there is an optimum placement of the inter-stage cooling which is around the middle section of the PBTR. An interesting finding is that the effect of the temperature exiting the inter-stage on the $CO$ conversion is significant.
It is shown that the exit temperature has to be roughly the same as the 1st section of the reactor for an increased maximum conversion to be achieved. Thus, care has to be taken to ensure that the inter-stage outlet temperature does not fluctuate much in the presence of disturbance. Hence, this has suggested that there is a need to find a good control strategy for this process system. Further, we have found out that there is an optimum placement of the conventional feedback controller (primary measurement) located roughly at the upper middle-end of the reactor, at sub-section 9. By using the optimum placement of the feedback controller, a feedforward controller has been combined together with the feedback to improve the reactor temperature control performance. However, the conventional FF-FB control strategy is still lacking in terms of controlling the overshoot. Interestingly, this has been improved via the implementation of the MSC-based FF-FB control strategy. In future works, we will extend the current study to study and develop more complex control strategies for different configurations of the WGSR systems.

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