

# Numerical Simulation of Iron Ore Reactor Isobaric and Cooling zone to Investigate Total Carbon Formation in Sponge Iron

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**Abstract**—Isobaric and cooling zone of iron ore reactor have been simulated. In this paper, heat and mass transfer equation are formulated to perform the temperature and concentration of gas and solid phase respectively. Temperature profile for isobaric zone is simulated on the range temperature of 873-1163K while cooling zone is simulated on the range temperature of 733-1139K. The simulation results have a good agreement with the plant data. Total carbon formation in the isobaric zone is only 30% of total carbon contained in the sponge iron product. The formation of  $\text{Fe}_3\text{C}$  in isobaric zone reduces metallization degree up to 0.58% whereas reduction of metallization degree in cooling zone up to 1.139%. The decreasing of sponge iron temperature in the isobaric and cooling zone is around 300 K and 600 K respectively.

**Keywords**—Mathematical Model, Iron Ore Reactor, Cooling Zone, Isobaric zone.

## I. INTRODUCTION

SOME iron ore reactor model had been proposed in few literatures [1]-[4]. Reference [2] proposed a model for moving bed counter current reactor. The model was limited on reduction zone on iron ore reactor. Another related model is formulated on direct reduction shaft furnace for producing sponge iron from iron ore as in [4]. Reference [3] shows a model for reduction iron oxides by carbon. This model was formulated on a circulating fluidized bed reactor.

In cooling zone, the effect of cooling gas is closely related with  $\text{Fe}_3\text{C}$  (iron carbide) and C (free carbon) formation. Reference [11] shows the effect of gas flow on simultaneous carburization and reduction of iron ore. It was found that carbon deposition did not occur in the absence of iron or a catalyst, irrespective of the gas flow or heating rate of the gas mixture. However, in the presence of metallic iron, the rates of reduction and carbon deposition were directly related to the gas flow rate.

Reference [6] concerned the formation of iron carbide in the process of methane decomposition on the iron powder. According to this paper, in the temperature range 450–500°C only the formation of  $\text{Fe}_3\text{C}$  occurred, whereas at temperatures above 500°C a carbon deposit was also found. Reference [7] and [8] show the kinetics of carbon deposit formation under the flow of the pure methane, whereas Reference [9] concerned about the kinetics of iron carbide formation.

Modeling of iron ore reactor isobaric and cooling zone is studied in this paper. This model is useful for investigating total carbon (iron carbide and free carbon) formation on those zones since the metallization degree which represents quality of sponge iron product will decrease if it contains total carbon in high amount. The model includes kinetics equations of  $\text{Fe}_3\text{C}$  formation, methane cracking,  $\text{Fe}_3\text{C}$  decomposition, and water gas shift reaction. The model can be used to perform temperature and concentration profiles along reactor cooling zone.

## II. REACTOR MODELING

Iron ore reactor is a kind of counter current reactor to produce sponge iron product from iron ore. Iron ore reactor is divided by 3 zone, those are reduction, isobaric, and cooling zone. The simple scheme of iron ore reactor is shown on Fig. 1. In the reduction zone, there are some reaction occurred between reduction gas and iron ore pellets. These reactions will extract iron metal from the ore to create sponge iron product. The sponge iron product will pass isobaric zone then it is cooled in the cooling zone.

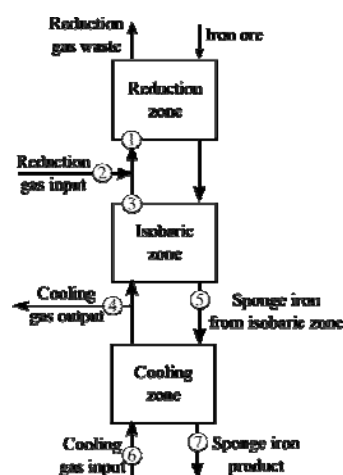


Fig. 1 Scheme of iron ore reactor

Isobaric zone is built to anticipate a mixing between leaking gas from reduction zone and cooling zone. In fact, this leakage is difficult to avoid. Small part of cooling gas usually slips, and reacts with reduction pellets from reduction zone. In this zone, reaction will occur between cooling gas and sponge iron to

form iron carbide through reaction  $\text{CH}_4 + 3\text{Fe} \rightarrow \text{Fe}_3\text{C} + 2\text{H}_2$ . Reference [5] shows that free carbon will be also formed as a result of iron carbide decomposition ( $\text{Fe}_3\text{C} \rightarrow 3\text{Fe} + \text{C}$ ) and methane cracking ( $\text{CH}_4 \rightleftharpoons \text{C} + 2\text{H}_2$ ). Water gas shift reaction also occurs by considering the composition of cooling gas through reaction  $\text{CO}_2 + \text{H}_2 \rightleftharpoons \text{CO} + \text{H}_2\text{O}$ .

The cooling process of sponge iron product in the cooling zone will also produce iron carbide ( $\text{Fe}_3\text{C}$ ) and free carbon (C) in the product. There is also a possibility of water gas shift reaction to occurs in the cooling zone as there is in isobaric zone.

Isobaric zone is built to anticipate a mixing between leaking gas from reduction zone and cooling zone. In fact, this leakage is difficult to avoid. Around 10% of cooling gas usually slips, and reacts with reduction pellets from reduction zone. In this zone, reaction will occur between cooling gas and sponge iron to form iron carbide through reaction  $\text{CH}_4 + 3\text{Fe} \rightarrow \text{Fe}_3\text{C} + 2\text{H}_2$ . Reference [5] shows that free carbon will be also formed as a result of iron carbide decomposition ( $\text{Fe}_3\text{C} \rightarrow 3\text{Fe} + \text{C}$ ) and methane cracking ( $\text{CH}_4 \rightleftharpoons \text{C} + 2\text{H}_2$ ). Water gas shift reaction also occurs by considering the composition of cooling gas through reaction  $\text{CO}_2 + \text{H}_2 \rightleftharpoons \text{CO} + \text{H}_2\text{O}$ .

Cooling process of sponge iron product in the cooling zone will also produce iron carbide ( $\text{Fe}_3\text{C}$ ) and free carbon (C) in the product. Water gas shift reaction also occurs in the cooling zone as there is in isobaric zone.

Mathematical model of isobaric and cooling zone in iron ore reactor are formulated on the gas and solid phase. This model consists of heat and mass transfer equation. Kinetics equation are taken account based on the reactions occurred in the each zone. The equations are formulated by assuming: (i) operating conditions have been steady-state, (ii) heat loss trough the wall of the reactor is considered only in the cooling zone, (iii) ethane and propane decomposition reactions are not be considered as they have very small fraction, and (iv) plug flow is used to approach a model for gas and solid phase. By considering the above assumptions, heat and mass transfer equations can be stated as:

$$\bar{u}_{\text{gas}} \frac{dC_{\text{CH}_4}}{dz} = r_1(\text{CH}_4, \text{H}_2, T_{\text{gas}}) + r_2(\text{CH}_4, \text{H}_2, T_{\text{gas}}) \quad (1)$$

$$\bar{u}_{\text{gas}} \frac{dC_{\text{CO}}}{dz} = r_4(\text{CO}, \text{CO}_2, \text{H}_2\text{O}, T_{\text{gas}}) \quad (2)$$

$$\bar{u}_{\text{solid}} \frac{dC_{\text{Fe}_3\text{C}}}{dz} = r_1(\text{CH}_4, \text{H}_2, T_{\text{solid}}) \quad (3)$$

$$\bar{u}_{\text{solid}} \frac{dC_{\text{C}}}{dz} = r_2(\text{CH}_4, \text{H}_2, T_{\text{solid}}) + r_3(\text{H}_2, T_{\text{solid}}) \quad (4)$$

$$\frac{dT_{\text{gas}}}{dz} = \frac{A_{\text{sp}} \cdot h(T_{\text{solid}} - T_{\text{gas}})}{M_{\text{gas}} \cdot C_{p_{\text{gas}}}} + \frac{\sum \left[ \left( H_{R(298K)} + \int_{298K}^{T_{\text{solid}}} C_{p_R}(T) dT \right) r_i \right]}{M_{\text{gas}} \cdot C_{p_{\text{gas}}}} \quad (5)$$

$$\frac{dT_{\text{solid}}}{dz} = \frac{A_{\text{sp}} \cdot h \cdot Rf(T_{\text{solid}} - T_{\text{gas}})}{M_{\text{solid}} \cdot C_{p_{\text{solid}}}} - \frac{Rf \sum \left[ \left( H_{R(298K)} + \int_{298K}^{T_{\text{solid}}} C_{p_R}(T) dT \right) r_i \right]}{M_{\text{solid}} \cdot C_{p_{\text{solid}}}} \quad (6)$$

$$\frac{dT_{\text{gas}}}{dz} = \frac{A_{\text{sp}} \cdot h(T_{\text{solid}} - T_{\text{gas}})}{M_{\text{gas}} \cdot C_{p_{\text{gas}}}} - \frac{U(T_{\text{gas}} - T_{\text{out}})}{M_{\text{gas}} \cdot C_{p_{\text{gas}}} \cdot \Delta z}$$

$$+ \frac{\sum \left[ \left( H_{R(298K)} + \int_{298K}^{T_{\text{solid}}} C_{p_R}(T) dT \right) r_i \right]}{M_{\text{gas}} \cdot C_{p_{\text{gas}}}} \quad (7)$$

$$\frac{dT_{\text{solid}}}{dz} = \frac{A_{\text{sp}} \cdot h \cdot Rf(T_{\text{solid}} - T_{\text{gas}})}{M_{\text{solid}} \cdot C_{p_{\text{solid}}}} - \frac{Rf \sum \left[ \left( H_{R(298K)} + \int_{298K}^{T_{\text{solid}}} C_{p_R}(T) dT \right) r_i \right]}{M_{\text{solid}} \cdot C_{p_{\text{solid}}}} \quad (8)$$

where

$Rf$ : relaxation factor, 0.088

$G_{\text{mg}}$ : cooling gas flow rate, 52000 Nm<sup>3</sup>/hour

$U$ : global heat transfer coefficient, 250 J/m<sup>2</sup>.K.s

$A_p$ : pellets external area, 0.000804 m<sup>2</sup>

Equation (5)-(6) are heat transfer equations for isobaric zone, whereas (7)-(8) are heat transfer equations for cooling zone. Mass transfer equations both of isobaric and cooling zone are same as shown in (1)-(4). Notation  $C$  is concentration in mol/m<sup>3</sup>,  $\bar{u}$  is superficial velocity in m/s,  $C_p$  is heat capacities in J/mol.K, and  $M$  is molar flow rate in mole/sec.m<sup>2</sup>.

Kinetic equations of iron carbide formation ( $r_1$ ), methane cracking ( $r_2$ ), iron carbide decomposition reaction ( $r_3$ ) and water gas shift reaction ( $r_4$ ) in the isobaric and cooling zone are shown as:

$$r_1 = k_1^+ \cdot f_1^+ \exp \left[ \frac{-E_A^+}{RT} \right] p_{\text{CH}_4} - k_1^- \cdot f_1^- \exp \left[ \frac{-E_A^-}{RT} \right] p_{\text{H}_2} \quad (9)$$

$$r_2 = k_2^+ \cdot f_2^+ \exp \left[ \frac{-E_A^+}{RT} \right] \frac{p_{\text{CH}_4}}{p_{\text{H}_2}^{1/2}} - k_2^- \cdot f_2^- \exp \left[ \frac{-E_A^-}{RT} \right] p_{\text{H}_2}^{1/2} \quad (10)$$

$$r_3 = k_3^+ \cdot f_3^+ \exp \left[ \frac{-E_A^+}{RT} \right] p_{\text{H}_2} \quad (11)$$

$$r_4 = k_4^+ \cdot f_4^+ \exp \left[ \frac{-E_A^+}{RT} \right] p_{\text{CO}}^{0.9} p_{\text{H}_2\text{O}}^{0.25} p_{\text{CO}_2}^{-0.6} \quad (12)$$

Those kinetic equations are adopted from literatures and remodeled by inserting correction factor  $f$  for each pre-exponential constant [8]-[10]. Correction factor for kinetics equation are shown on Table I.

TABLE I  
CORRECTION FACTOR

Symbol	Value
$f_1^+$	Correction factor for $k_1^+$
$f_1^-$	Correction factor for $k_1^-$
$f_2^+$	Correction factor for $k_2^+$
$f_2^-$	Correction factor for $k_2^-$
$f_3^+$	Correction factor for $k_3^+$
$f_4^+$	Correction factor for $k_4^+$

Heat capacities of gas and solid are calculated based on the temperature changes. On the gas phase, heat capacity is considered only for CH<sub>4</sub> and H<sub>2</sub> since the other substances have very small fraction. Heat capacity of gas is evaluated using:

$$C_{p_{\text{gas}}} = \left( \frac{C_{\text{CH}_4}}{C_{\text{CH}_4} + C_{\text{H}_2}} \times C_{p_{\text{CH}_4}} \right) + \left( \frac{C_{\text{H}_2}}{C_{\text{CH}_4} + C_{\text{H}_2}} \times C_{p_{\text{H}_2}} \right) \quad (13)$$

$$C_{p_{(\text{CH}_4 \text{ or } \text{H}_2)}} = aT_{\text{gas}} - bT_{\text{gas}}^2 + cT_{\text{gas}}^3 - dT_{\text{gas}}^4 + eT_{\text{gas}}^5 \quad (14)$$

$$C_{p(Fe)} = 0.0029T_{gas} - 5E^{-6}T_{gas}^2 + 3E^{-9}T_{gas}^3 \quad (15)$$

Constant values of a, b, c, and d for CH<sub>4</sub> and H<sub>2</sub> respectively are obtained from literature [12]. As the gas phase, heat capacity of solid phase (sponge iron product) is also considered only for Fe due to the biggest fraction. Heat capacity of Fe as the function of temperature is obtained from literature [13].

Standard reaction enthalpy ( $H_{R(298K)}$ ) and reaction heat capacity ( $C_{pR}$ ) on (5) and (7) are calculated for iron carbide formation, methane cracking, and iron carbide decomposition reaction respectively since they occur on the solid phase. Reference [12] shows the values of  $H_{R(298K)}$ .

### III. RESULTS AND DISCUSSION

The model was calculated using finite element method. The calculations of isobaric and cooling zone were solved simultaneously, and the results were validated with the plant data. Table II resumes the comparison between plant data and simulation. From this table, one observes that calculation results are in good agreement with the reference data. Some data are quite difficult to get in the plant. However they can be calculated from mass and energy balance in the some point.

The data of leaking gas exit from isobaric zone is not available in the plant report. However leaking gas temperature can be calculated since the temperature of reduction gas in the inlet and the bottom of reduction zone is known. The plant report shows that reduction gas inlet temperature is about 930°C (1203 K) and reduction gas temperature in the bottom of reduction zone is 920°C (1193 K). Since reduction gas in the bottom of reduction zone is a mixing between leaking gas from isobaric zone and reduction gas inlet, from energy balance as in (16), we can obtain the temperature of leaking gas exit from isobaric zone is about 1139 K.

$$M_1.C_{p1}(T_1 - T_{ref}) = M_2.C_{p2}(T_2 - T_{ref}) + M_3.C_{p3}(T_3 - T_{ref}) \quad (16)$$

Subscripts in (16) refer to flow number on Fig. 1.

Based on the plant data, 10% of cooling gas usually slip to isobaric zone, and react with reduced iron from reduction zone. This is caused the formation of total carbon is already started from isobaric zone and continued to cooling zone. Even though there is no plant data about total carbon amount formed in the isobaric and cooling zone respectively, it can be calculated by mole balance reaction. Mole difference between CH<sub>4</sub> inlet and outlet in the cooling gas will give the information about total carbon formation in the cooling zone, since total carbon is formed by methane cracking. From mole reaction balance, it is obtained that total carbon formation in the cooling zone is about 407 gram/second (70% of total carbon formation in the iron ore reactor). We can also get total carbon formation in the isobaric zone from this calculation is about 171 gram/second.

The simulation result is performed on the concentration and temperature profile. Temperature profiles on Fig. 2 show that even though there is only 10% of cooling gas slip in the isobaric zone, the cooling of sponge iron product is already started from isobaric zone. Based on the solid temperature profile on Fig. 2, the decreasing of sponge iron temperature in the isobaric and cooling zone is around 300 K and 600 K respectively.

TABLE II

COMPARISON BETWEEN PLANT DATA AND SIMULATION RESULTS

Flow Number	Note	Plant data	Simulation
2	Temperature of reduction gas inlet (K)	1203	
1	Temperature of reduction gas in the bottom of reduction zone (K)	1193	
3	Leaking gas from isobaric zone (K)	*1139	1139.025
6	COOLING GAS INPUT		
	Flow rate (m <sup>3</sup> /sec.)	11800	11800
	Temperature (K)	316	316
	CH <sub>4</sub> (% vol.)	67.8	68
	H <sub>2</sub> (% vol.)	21.36	21.36
	CO (% vol.)	3.4	3.4
	CO <sub>2</sub> (% vol.)	3.1	3.1
	N <sub>2</sub> (% vol.)	1.86	1.86
	H <sub>2</sub> O (% vol.)	1.96	1.96
	C <sub>2</sub> H <sub>6</sub> (% vol.)	0.23	-
	C <sub>3</sub> H <sub>8</sub> (% vol.)	0.143	-
4	COOLING GAS OUTPUT		
	Temperature (K)	733 K	736 K
	CH <sub>4</sub> (% vol.)	65.66	65.68
	H <sub>2</sub> (% vol.)	24.26	25.31
	CO (% vol.)	2.1	2.14
	CO <sub>2</sub> (% vol.)	4.39	4.28
	N <sub>2</sub> (% vol.)	1.85	1.84
	H <sub>2</sub> O (% vol.)	1.96	0.72
7	SPONGE IRON PRODUCT		
	Temperature (K)	331 K	331 K
	Fe <sub>2</sub> + and Fe metal (% mass)	91.7	91.7
	Total carbon (% mass)	2	2
	Total carbon (gr/sec.)	578	578.0
	Total carbon formation on isobaric zone (gr/sec.)	*171	170.4
	Total carbon formation on cooling zone (gr/sec.)	*407.	407.6
	Gangue (% mass)	6.3	6.3
5	SPONGE IRON FROM ISOBARIC ZONE		
	Temperature (K)	331 K	331 K
	Fe <sub>2</sub> + and Fe metal (% mass)	93.11	93.11
	Total carbon (% mass)	0.59	0.59
	Gangue (% mass)	6.3%	6.3%

\* From plant data calculation

Flow numbers are referred to Fig. 1

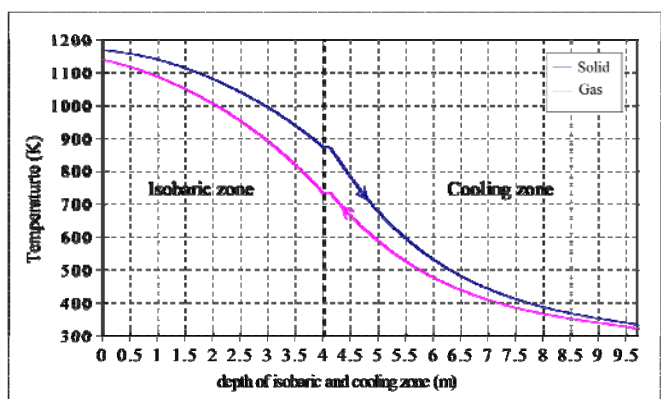


Fig. 2 Temperature profile along isobaric and cooling zone

Fig. 3 performs solid and gas concentration along isobaric and cooling zone. The color dimension represents temperature changes along the depth of isobaric and cooling zone. Gas concentration profiles show that from the beginning of gas inlet point in the cooling zone, methane concentration is constant,

but it starts to decrease at 4.5 m of depth of isobaric and cooling zone. This decreasing is still continued to isobaric zone. The decrease of methane concentration yields because methane starts to decompose to form iron carbide and free carbon. At the same depth, hydrogen profile shows an amplification which is caused by the results of those reactions.

Methane and hydrogen concentration profiles are in agreement with solid concentration profiles in Fig. 3. Iron carbide and free carbon formations start from isobaric zone and continue to cooling zone. At 4.5 m of depth of isobaric and

cooling zone,  $\text{Fe}_3\text{C}$  stops to be formed, whereas C stops at 4.25 m of the depth of isobaric and cooling zone. Calculation of mole balances also depicts a satisfied agreement between  $\text{CH}_4$ ,  $\text{H}_2$ ,  $\text{Fe}_3\text{C}$ , and C moles.

The quality of sponge iron is primarily ascertained by the percentage of metallization (removal of oxygen), which is the ratio of metallic iron to the total iron present in the product. The formation of  $\text{Fe}_3\text{C}$  in isobaric zone reduces metallization degree up to 0.58% whereas reduction of metallization degree in cooling zone up to 1.139%.

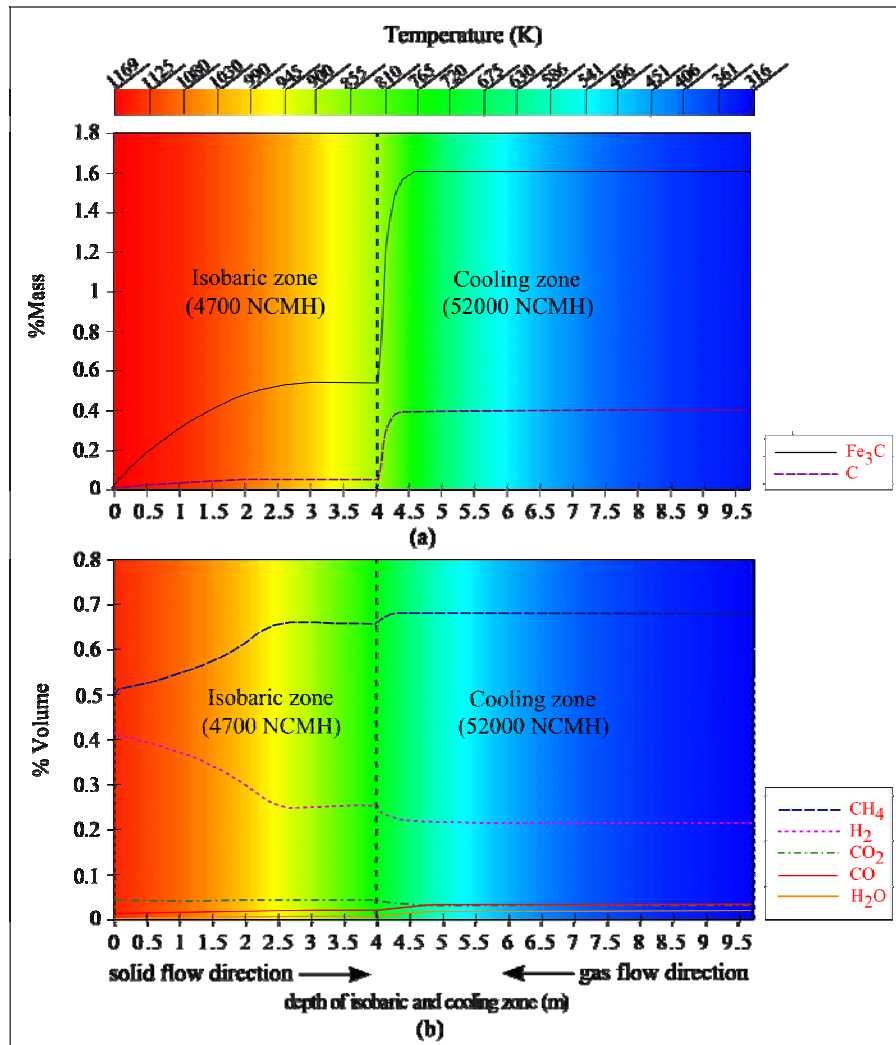


Fig. 3 Concentration profile as a function of the depth of isobaric and cooling zone (a) solid concentration; (b) gas concentration

Solid temperature profile in Fig. 3 implies that  $\text{Fe}_3\text{C}$  and C start to be formed at temperature of 723 K (450°C) and 779 K (506°C) respectively. Reference [6] shows that in the temperature range 450–500°C, the formation of  $\text{Fe}_3\text{C}$  occurs in the process of methane decomposition on the iron powder. The formation of free carbon at the temperature of 500°C also agrees with equilibrium diagram for the system iron–methane–hydrogen [14].

Gas concentration profile on Fig. 3 also shows the slight change of concentration on the some components caused by

water gas shift reaction. Conversion of CO and  $\text{H}_2\text{O}$  turn to  $\text{CO}_2$  and  $\text{H}_2$  make a decreasing in percentage of CO and  $\text{H}_2\text{O}$ , while  $\text{CO}_2$  and  $\text{H}_2$  increase. Calculation of water gas shift reaction mole balances depicts a satisfied agreement between CO,  $\text{H}_2\text{O}$ , and  $\text{CO}_2$  moles.

Based on the gas flow direction on Fig. 2, water gas shift reaction starts to occur around 0.6 m of the depth of reactor cooling zone when gas temperature reaches 631 K. Reference [16] stated that water gas shift reaction is usually performed with oxide iron catalyst at temperature between 370°C (643 K)

and 450°C (723 K), but this reaction can also be carried out using the same catalyst at lower temperature around 327°C (600 K) as in [15].

#### IV. CONCLUSION

Isobaric and cooling zone of iron ore reactor has been simulated. The results have satisfied agreement with the plant data. Even though there is only 10% of cooling gas slip in the isobaric zone, the cooling of sponge iron product is already started from isobaric zone. The decreasing of sponge iron temperature in the isobaric and cooling zone is around 300 K and 600 K respectively.

Although the temperature in the isobaric zone is higher than cooling zone, total carbon formation in the cooling zone is faster than isobaric zone. Total carbon formation in the isobaric zone is only 30% of total carbon contained in the sponge iron product. The formation of  $\text{Fe}_3\text{C}$  in isobaric zone reduces metallization degree up to 0.58% whereas reduction of metallization degree in cooling zone up to 1.139%.

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