



# Transformation of low-rank coal to clean syngas and power via thermochemical route



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## ABSTRACT

An emerging gasification was proposed for the transformation of low-rank coal to clean syngas and electricity. The excess heat from char combustion was used to evaporate moisture in coal and to generate power via steam turbine generator. The simulation was developed in the Aspen Plus based on the proper thermodynamic model. The validation exhibited a good concurrence between the present model and the experiments under the same condition with the relative error <10% in the pyrolysis stage. The moisture content reduced the energy efficiency by ~10% but increased the H<sub>2</sub> production by ~50% which results in the increase of H<sub>2</sub>/CO up to 10. The highest energy and exergy efficiencies (91% and 79%, respectively) was observed on the O<sub>2</sub> equivalence ratio and steam to ratio of 0.21 and 0.06, respectively. O<sub>2</sub> is an appropriate gasifying agent to produce syngas as the energy source, while steam gives high-quality syngas as the feedstock for chemical industries. The CO<sub>2</sub> emission of the proposed configuration is below 50 kg CO<sub>2</sub>/GJ (half of the conventional coal combustion, ~100 kg CO<sub>2</sub>/GJ) when the fraction of char to combustor is less than 0.4.

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## 1. Introduction

Coal is a promising energy resource due to its abundant deposits in many countries. Coal is intensively utilized via direct combustion to supply the energy demand. Consequently, coal combustion is the majority responsible for the increase of CO<sub>2</sub> emission, as compared to natural gas and oil [1]. Coal is also targeted to substitute natural gas [2] and oil [3], and as the feedstock of petrochemical industries [4] and synthetic fuel (i.e. dimethyl ether) [5]. An appropriate conversion technology of coal into energy is crucial to meet the worldwide demand for energy and petrochemical products. Gasification is a suitable process for coal utilization due to its ability to convert coal into various products at high efficiency (up to 81%) [6].

Gasification is a thermal conversion process which converts coal into syngas, with a major constituent of H<sub>2</sub> and CO. The syngas composition is strongly influenced by the coal properties, which widely diverse due to the natural source [7]. In addition, the composition of gasifying agents [8] and gasifier operating conditions [9] strongly affected the syngas quality. It is worth noting that the composition of syngas is crucial when syngas is targeted as an intermediate chemical to produce petrochemical products. For instance, the methanol synthesis reactor required syngas with a H<sub>2</sub>/CO ratio of 2.0 [10]. Meanwhile, for the power generation purpose, the syngas composition delivers a minor influence on the process given it only focuses on the conversion of thermal energy [11].

One should note that the H<sub>2</sub>/CO ratio is strongly affected by the natural properties of coal (i.e., coal composition) [12]. For instance, Wang et al. [13] reported that different H<sub>2</sub>/CO molar ratios were observed on the gasification (fixed bed down-draft gasifier) of three types of coal from different mining sites in China. Considering this nature challenge, various efforts (i.e., addition of steam, incorporation of catalysts, and gasifier design) have been conducted to adjust the syngas composition in order to satisfy the required petrochemical feed specs, particularly the H<sub>2</sub>/CO ratio [14].

The selection of steam (H<sub>2</sub>O) gasifying agents has been reported

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as a promising strategy to adjust the H<sub>2</sub>/CO ratio. The introduction of steam into the gasifier increases the H<sub>2</sub>/CO ratio as steam provides H atom for H<sub>2</sub> formation. Kumar et al. [15] conducted gasification experiments using Indian coal with the mixture of air and steam as the gasifying agent. They reported that the increase of steam flow into the gasifier delivers a positive effect on the H<sub>2</sub>/CO ratio. A similar conclusion also has been reported by Crnomarkovic et al. [16] using lignite Kolubara coal in an entrained flow gasifier. One should note that increasing the H<sub>2</sub>/CO ratio by the addition of steam technically promotes the water-gas shift and endothermic steam reforming reactions [17]. Therefore, in an auto-thermal gasification system, the high steam flow reduces gasifier temperature, which inhibits the rate of both reactions. Therefore, though the addition of steam successfully enhances the H<sub>2</sub>/CO ratio, the obtained results still require improvement to meet the required syngas quality due to the trade-off between the steam injection and gasifier temperature.

Another way to adjust H<sub>2</sub>/CO ratio is by introducing catalysts to the gasification process. In the gasification process, catalysts have a substantial role to facilitate tar reforming reaction (i.e., converting tar into H<sub>2</sub> and CO) [18]. To cite a few, the Ni/CeZrO<sub>2</sub> catalysts have been reported to facilitate the reforming of tar in a modeled gasification of coal at the gasification temperature above 700 °C, which results in the increase of the H<sub>2</sub>/CO ratio up to 1.92 [19]. The addition of K<sub>2</sub>CO<sub>3</sub> catalysts also showed a positive effect on the H<sub>2</sub> concentration in gasification of sub-bituminous coal under high-pressure condition (3.5 MPa), with the highest H<sub>2</sub>/CO ratio of ~5.5 [20]. The incorporation of Ni on the gasification of coal also boosted the H<sub>2</sub> production, which results in a positive effect on the H<sub>2</sub>/CO ratio [21]. Zhang et al. [21] reported that the gasification of coal using Ni catalysts produced syngas with the highest H<sub>2</sub>/CO ratio of 6. Though the catalytic gasification exhibited good performance, the operating cost will significantly increase due to the catalyst's expenses.

The modification of gasifier design can be an alternative strategy to improve the H<sub>2</sub>/CO ratio by avoiding additional costs for catalysts procurement. A number of researchers reported the configuration of the coal gasification process both numerically and experimentally. A numerical investigation is an attractive approach due to its effectiveness in terms of time and cost. Chen et al. [22] studied numerically integrated supercritical gasification of coal to enhance energy efficiency. In their configuration, the boiler feed water was generated by using the hot gasifier products prior to entering the boiler, which results in increased energy efficiency. The identical configuration was proposed by the same group [23] with an addition of a combined cycle to improve electricity production. Yilmaz et al. [24] proposed integrated coal gasification to produce high-quality syngas and electricity by the combination of a multi-generation plant including gasification, power generation and electrolyzer. Based on the mentioned literature earlier, hydrogen production is solely enhanced by maintaining the gasifier temperature. Indeed, higher gasification temperatures promote hydrogen production. However, higher temperature results in higher operating cost. An interesting gasification design was reported by Xiao et al. [25], which proposed dual-loop gasification to minimize tar content and enhance H<sub>2</sub> concentration. Olivine was circulated between the reactors and played a role as a catalyst for tar minimization. With this configuration, the high-quality syngas (H<sub>2</sub>/CO ratio varies between 4.8 and 6.8) were successfully produced from the gasification of Shenmu bituminous coal by varying the operating temperature from 700 to 850 °C. However, the implementation of circulating solid is a strenuous process and requires high operating costs [26].

Aiming to avoid the dependency on the catalysts or requiring intensive energy which leads to high operating cost, the present

study proposes the development of a highly efficient gasification process by proposing an emerging configuration of low-rank coal gasification to produce high-quality syngas and electricity. The low-rank coal was selected as the feedstock given it is evenly distributed worldwide with an attractive price [27]. In contrast to the earlier reported strategies (i.e., utilization of catalyst, modification of the gasification with looping heat carriers, and addition of steam), the proposed novel configuration allows the adjustment on the wide-range H<sub>2</sub>/CO ratio by controlling the amount of char into the gasification and combustion chambers, as illustrated in Fig. 1 (detail process description is presented in Section S1, Supplementary Information). A novel model of pyrolysis is used to accommodate tar formation during coal thermal decomposition. The proposed configuration was developed in Aspen Plus simulation software which allows the main chambers including reforming, gasification and combustion operate at the thermodynamic equilibrium. The gasification performance is assessed based on the syngas quality and the energy conversion with varying the flow rate of gasifying agents (i.e., O<sub>2</sub> and steam). The energy conversion is evaluated based on overall energy efficiency, overall exergy efficiency and cold gas efficiency. The quality of syngas is determined by the composition of major components (H<sub>2</sub>, CO and CO<sub>2</sub>) and the H<sub>2</sub>/CO ratio. Regarding low-rank coal, the operation and economic problem of low-rank coal majorly depend on the moisture content [28]. Thus, the effect of coal moisture on the process performance was also evaluated in the present study.

## 2. Performance evaluation

As mentioned above, the performance evaluation was conducted in both terms: energy conversion and syngas quality. The quality of syngas was measured by the flow rate, H<sub>2</sub>/CO ratio and distribution of major constituents (H<sub>2</sub>, CO, and CO<sub>2</sub>). The cold gas efficiency (CGE) is the energy ratio of the syngas to the feed, which is defined as:

$$CGE(-) = \frac{m_{sg} LHV_{sg}}{m_{cl} LHV_{cl} + H_s m_s} \quad (1)$$

where *LHV*, *m*, and *H* are lower heating value, mass flow rate, and enthalpy, respectively. The subscript *sg*, *cl* and *s* denote syngas, coal and steam, respectively. The overall energy efficiency ( $\eta_{en}$ ) is the ratio of the input energy to the output energy, which is illustrated in the following equation:

$$\eta_{en} = \frac{E_{out}}{E_{in}} \quad (2)$$

where  $E_{out}$  and  $E_{in}$  are the total energy that enters and leaves the system, respectively.  $E_{out}$  is a summation of various types of energy including the energy contains in the syngas, the heat taken by the coolers and the generated electricity.  $E_{in}$  is a total energy input including the energy contains in the feed, the heat required by boiler, the energy to drive CO<sub>2</sub> compressor, the energy to produce O<sub>2</sub> (1.098 MJ/kg pure O<sub>2</sub> [29]) and the energy required by CO<sub>2</sub> absorber (3 GJ/ton of CO<sub>2</sub> captured) [30].

The exergy represents the maximum amount of work that can be obtained from a system which goes to the equilibrium condition of its environment [31]. The exergy analysis is referred to the earlier literature [32]. The overall exergy efficiency is defined as the ratio of the exergy in the outlet streams ( $\xi_{sy,out}$ ) to its counterpart in the inlet streams ( $\xi_{sy,in}$ ), as shown in Eq. (17) [33]:

$$\eta_{ex} = \frac{\xi_{sy,out}}{\xi_{sy,in}} \quad (3)$$

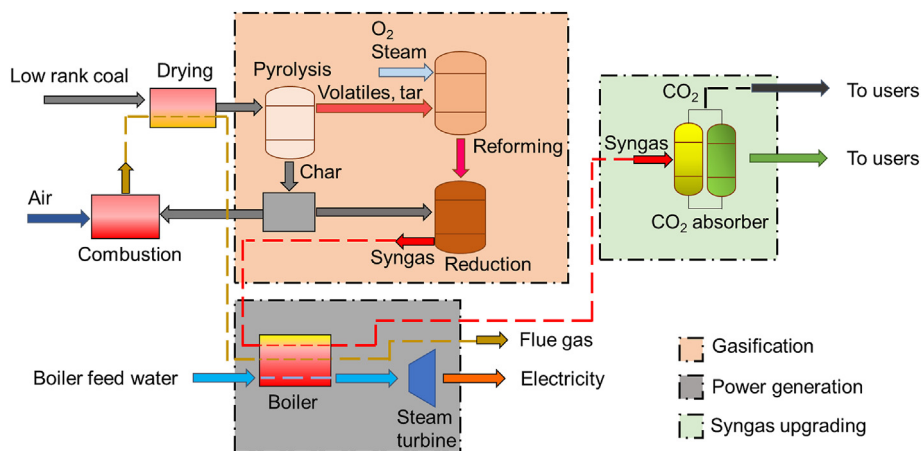


Fig. 1. Scheme of the integrated configuration for gasification of low-rank coal with char combustion for drying and power generation.

The calculation of exergy system is discussed in Supplementary Information (Section S.3).

### 3. Model validation

The validation was carried out by comparing the results of this study with the literature. One should note that in addition to designing the gasification process, we also proposed the pyrolysis model to accommodate tar. Firstly, we compare the pyrolysis product from the simulation with the literature. The experimental results of the pyrolysis of low-rank coal have been reported in the literature [34]. One can clearly see in Table 1 that the pyrolysis product was dominated by tar. This expected distribution is due to low pyrolysis temperature (723.15 K) as a number of researchers [35] reported that lower pyrolysis temperature promotes the formation of tar. Table 1 shows that the distribution of major pyrolysis products obtained from the model was comparable with the one obtained from the experimental literature. The lowest relative error was found on tar compounds, while the highest relative error was observed on char products. However, the results were acceptable given the relative error remains below 10%. Secondly, we compare the product of the entire gasification system under similar operating conditions. It can be clearly seen in Fig. 2 that the syngas flow rate and gasification temperature of the present study are in line with its counterpart from the literature [36,37]. This is in line with the previous literature [36–39] that reported good accuracy of the thermodynamic approach to simulate the gasification process. The slight difference between this study and the literature can be attributed to the presence of tar formation in our model, while the tar formation phenomenon is neglected in the literature [37]. Consequently, the estimated gasification temperature in this study is slightly lower than the one in the literature due to the occurrence of endothermic tar reforming reaction (Eq. S8, Supplementary Information).

Table 1  
The composition of the pyrolysis product.

Comp.	Literature [34]	This study	Error
Volatile matter	13.3%	14.2%	7%
Tar	52.0%	54.0%	4%
Char	34.7%	31.7%	9%

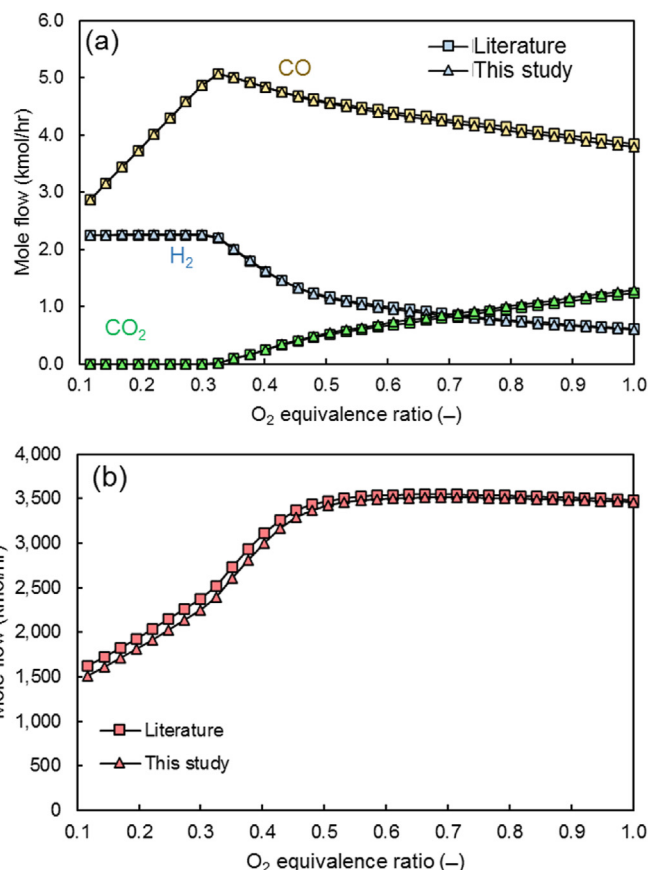


Fig. 2. The effect of O<sub>2</sub> equivalence ratio on (a) the syngas flow rate and (b) gasification temperature. The triangle marks indicate the results from our study, while the square marks indicate its counterpart from literature [36,37].

### 4. Results and discussion

The gasification performance was assessed by varying the flow rate of the gasifying agent at different coal moisture contents. In addition, the effect of char distribution into the combustion stage (CR) and the reduction (RD) was also evaluated in this study. The term “char to CR” represents the fraction of char from the pyrolysis (PR) to CR. For instance, char to CR of 0.1 indicates that 10 wt% of char from PR is directed to the CR, while 90 wt% of char goes to RD.

Those parameters were selected due to their strong influence on gasification performance [40]. The discussion on the gasification performance begins with the syngas quality in terms of its composition including the  $H_2/CO$  ratio. The discussion continues with energy production in the form of syngas and electricity. Finally, the overall gasification performance is presented in terms of energy and exergy efficiencies.

#### 4.1. Oxygen equivalence ratio

The study was conducted by flowing different flow rates of high-purity  $O_2$  into the reformer (RE) with a constant coal flow rate, which leads to the variation of  $O_2$  ER between 0.0 and 1.0. Meanwhile, no boiler feed water was directed to the boiler (BR) to maintain the S/C ratio of 0.0.

The major component of syngas on a dry basis significantly changes with the change of  $O_2$  ER, as illustrated in Fig. 3a. One can notice that a higher amount of  $O_2$  promotes the formation of CO and  $CO_2$ . Meanwhile, the mole fraction of  $H_2$  was significantly diminished. For instance, the mole fraction of CO and  $CO_2$  increased from 0.52 to 0.75 and 0.00 to 0.04, respectively, while the mole fraction of  $H_2$  reduced from 0.46 to 0.15, when the  $O_2$  ER was added from 0.12 to 1.00 on the char fraction to the CR of 0.1 with the coal moisture content of 15%. This finding can be related to the oxidation reactions (Eq. (S1) – (S3)) due to the interaction between char,  $H_2$  and CO with the sufficient amount of  $O_2$  to produce  $CO_2$  and  $H_2O$ . An identical trend is also found in the gasification of low-rank coal with the moisture content of 30% and 40%. In addition, the gasification at various char fractions (0.4 and 0.7) showed a similar trend, as illustrated in Fig. 3b and c, respectively, with different magnitudes for each configuration. This is in line with the experiments by Xiao et al. [25], using Shenmu bituminous coal. They reported that

increasing the  $O_2$  flow rate delivers high CO productions but suppresses the formation of  $H_2$ . The same group [41] also reported a similar conclusion using lignite coal.

One can easily notice from Fig. 3a that moisture has a positive effect on  $H_2$  production, while the negative influence of moisture was found on CO. For instance, the  $H_2$  mole fraction increased from 0.46 to 0.61, while the mole fraction of CO reduced from 0.52 to 0.22 when the moisture content elevates from 15% to 45% at the  $O_2$  ER of 1.1 and char fraction to CR of 0.1. This is because the presence of water promotes steam reforming reaction (Eq. (S6)), as reported experimentally by Jia-liang et al. [42], who reported higher char conversion with elevating the steam concentration. In addition, water also promotes the water-gas shift reaction (Eq. (S5)), which further converts CO and  $H_2O$  into  $CO_2$  and  $H_2$ . This is confirmed by the elevation of the  $CO_2$  mole fraction as the moisture content increased. Again, the comparable fashion is also found on the char fraction to CR of 0.4 and 0.7, as illustrated in Fig. 3b and c, respectively. A similar results are also reported by Spiegl et al. [43], who carried out gasification experiments using German lignite coal in a bench-scale fluidized bed reactor.

The effect of char distribution to CR and RD on the syngas composition can be noticed by comparing Fig. 3a, b and c. From Fig. 3 one can see that a higher fraction of char to CR delivers a negative influence on the  $H_2$  concentration. Interestingly, the CO concentration increased with increasing char flow rate to CR, while the concentration of  $CO_2$  remained constant. For instance, the mole fraction of  $H_2$  depleted from 0.50 to 0.43, while the mole fraction of CO elevated from 0.44 to 0.50 when the char fraction to CR was raised from 0.1 to 0.7, using coal with the moisture content of 45% at the  $O_2$  ER of about 0.30. This result indicates the occurrence of exothermic partial oxidation (Eq. (1)), to produce CO. Meanwhile, the minimum amount of char promotes  $H_2$  oxidation into  $H_2O$  (Eq.

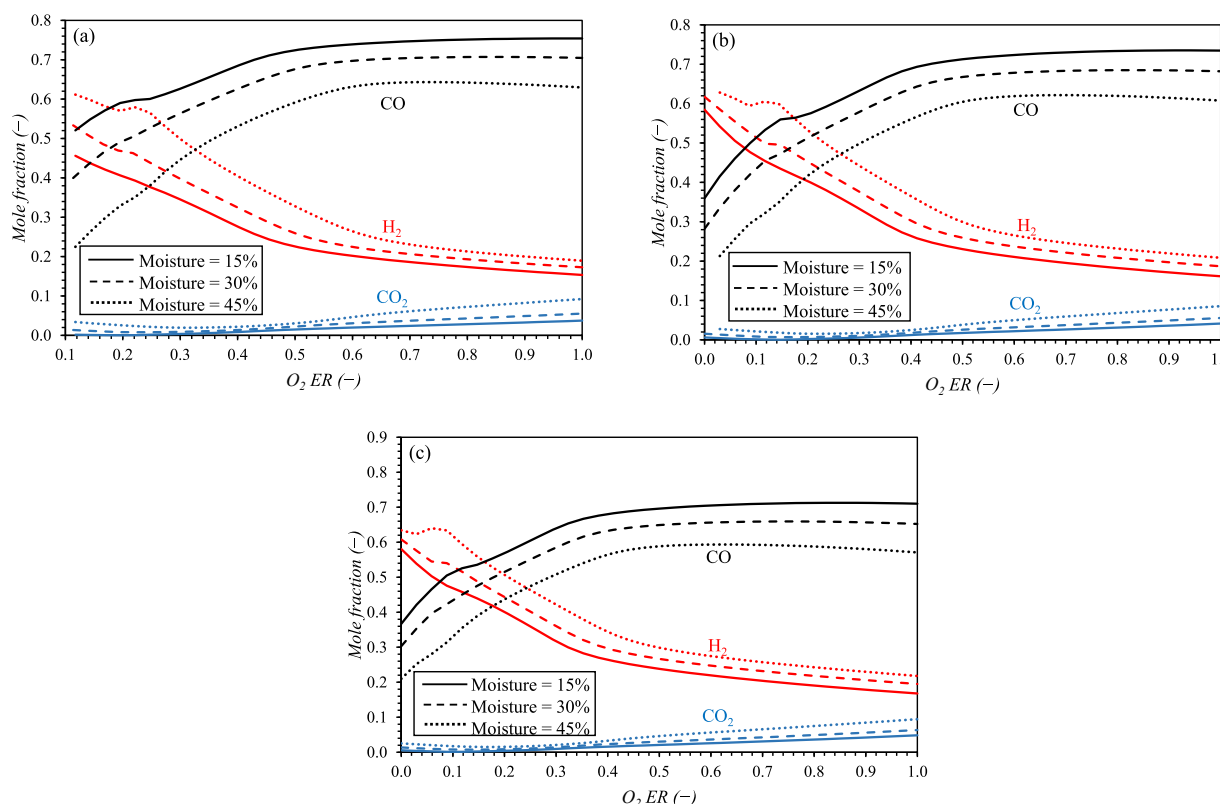


Fig. 3. The effect of  $O_2$  ER ratio on the major constituents in the syngas at different char distributions: (a) char to CR = 0.1, (b) char to CR = 0.4 and (c) char to CR = 0.7.

(2)) given  $H_2$  is highly reactive under  $O_2$  atmosphere [44]. An identical trend is also observed on other moisture contents.

The variation of syngas composition indeed influences the  $H_2/CO$  ratio. One can notice from Fig. 4 that the present configuration produced a wide variation of the  $H_2/CO$  ratio by varying  $O_2$  from 0.00 to 1.00. In addition, the higher moisture content gave a good effect on the  $H_2/CO$  ratio due to high  $H_2$  production. An interesting result was found on the char distribution. The higher fraction of char to CR has an adverse effect on the  $H_2/CO$  ratio. This indicates that minimum carbon supply by reducing char into RD suppresses both CO and  $H_2$  production. Furthermore, the decrease of  $H_2$  production due to increasing char to CR is higher than the decrease of CO production. The reason for this can be explained by the excess amount of  $O_2$  at a high fraction of char into CR, which promotes  $H_2$  oxidation reaction (Eq. (S2)) instead of partial oxidation of carbon (Eq. (1)) due to the absence of carbon source.

The syngas composition also strongly influences the cold gas efficiency as the representation of syngas heating value. One can clearly see in Fig. 5 that each gasification configuration exhibited the highest cold gas efficiency at different  $O_2$  ER. For instance, the highest CGE of 0.92 was observed on the gasification of coal with 15% moisture content at  $O_2$  ER 0.22. The addition of  $O_2$  supply above  $O_2$  ER 0.22 results in lower CGE. This result strongly relates to syngas production, where at low  $O_2$  ER, the syngas flow rate is minimal due to incomplete conversion of coal. Consequently, the unconverted coal was disposed of from the system instead of being converted into syngas. This is in line with the earlier literature [45], showing the strong influence of syngas flow rate on the CGE on biomass gasification. It is worth noting that high  $O_2$  ER results in a low  $H_2$  concentration, while the CO concentration is high. This also can be the reason for the decrease of CGE with increasing  $O_2$  ER as the heating value of  $H_2$  is much higher than its counterpart of CO. A

similar finding was also reported in the earlier literature [46] using coal and biomass as the feedstock. One can notice that the present configuration suppresses the amount of unconverted coal by directing char into the CR. In this configuration, the CGE reduces with increasing the char fraction into the CR, but it is counter-balanced by the increase of power generation, as shown in Fig. 5. For instance, the CGE diminished from 75% to 49%, while the electricity production increased from 28 to 72 kW, when the char fraction to CR was augmented from 0.1 to 0.7 on the gasification of coal with 15% moisture content at the  $O_2$  ER of about 0.37.

The energy and exergy balances gave a comprehensive insight into the heat and energy flow within the gasification system. The  $O_2$  flow rate strongly influenced the energy conversion performance, as illustrated in Fig. 6. The addition of  $O_2$  ER has a positive influence on the overall efficiency until the optimum  $O_2$  ER is reached. For instance, the gasification of coal with 15% moisture content and char fraction to CR of 0.1 exhibited the optimum  $O_2$  ER of 0.22 with the overall energy efficiency and the overall exergy efficiency of 89% and 76%, respectively at the  $O_2$  ER of 0.22, as depicted in Fig. 6a. The increase of  $O_2$  ER above 0.22 slightly reduced the energy efficiency, but significantly suppressed the exergy efficiency. This finding can be explained that energy efficiency only considers the energy balance, regardless of the syngas composition. Thus, the decrease in energy efficiency only comes from the losses in the equipment as listed in Table S1. On the other hand, in the exergy evaluation, syngas composition is considered as a part of energy quality. With this end, the decrease of exergy efficiency is also associated with the increase of  $CO_2$  concentration and the decline of  $H_2$  concentration as the  $O_2$  ER ratio elevated. The comparable trend is also found on the char fraction into CR of 0.4 and 0.7, as illustrated in Fig. 6b and c, respectively. The specific  $CO_2$  emission is also lower at the optimum  $O_2$  ER (Figure S2, Supplementary

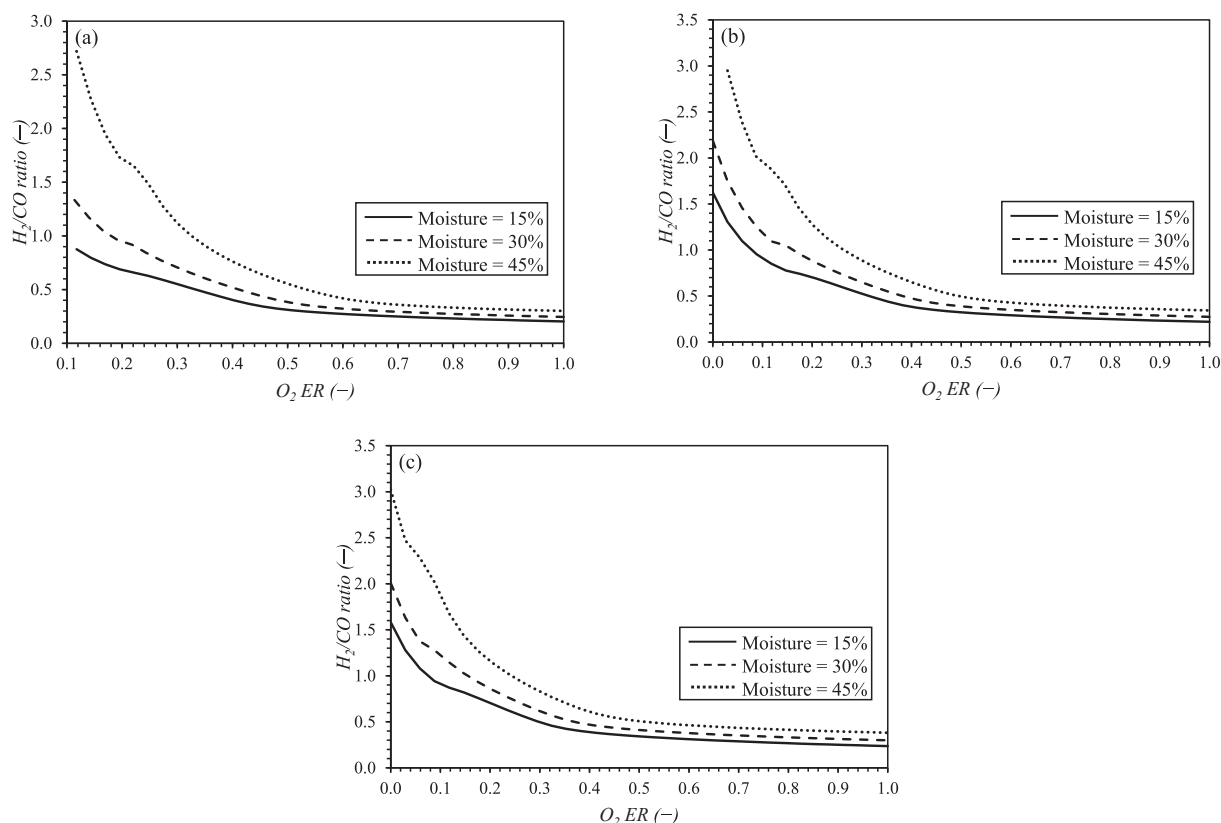


Fig. 4. The effect of  $O_2$  ER ratio on the  $H_2/CO$  ratio in the syngas at various char distributions: (a) char to CR = 0.1, (b) char to CR = 0.4 and (c) char to CR = 0.7.



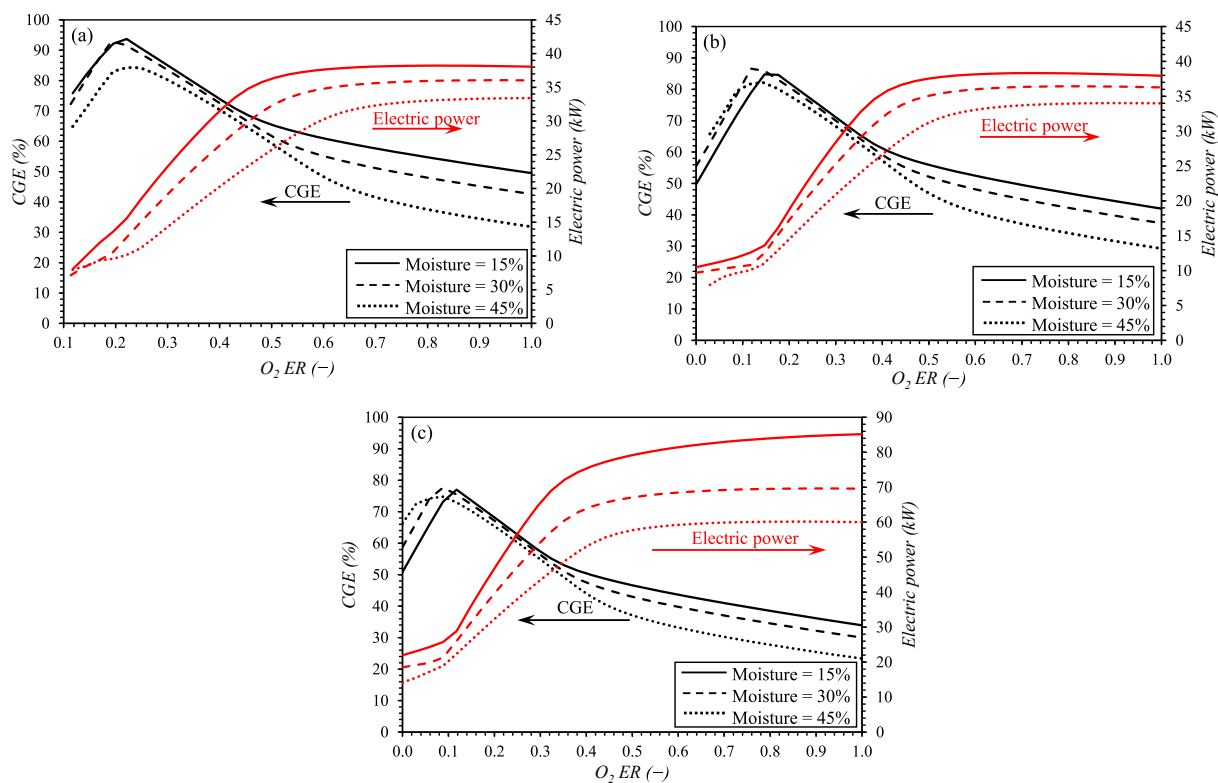


Fig. 5. The effect of  $O_2 ER$  ratio on the cold gas efficiency and electricity production at various char distribution: (a) char to CR = 0.1, (b) char to CR = 0.4 and (c) char to CR = 0.7.

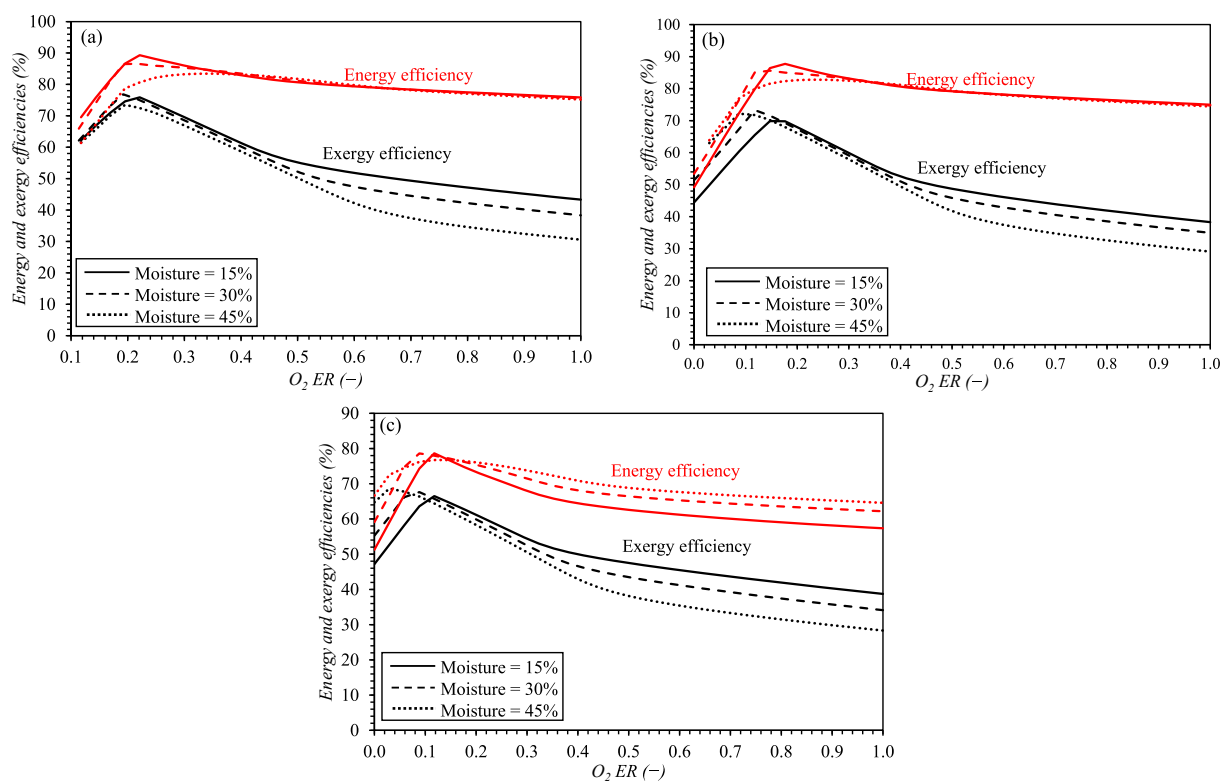


Fig. 6. The effect of  $O_2 ER$  ratio on the overall energy efficiency and overall exergy efficiency at various char distribution: (a) char to CR = 0.1, (b) char to CR = 0.4 and (c) char to CR = 0.7.

Information).

Indeed, the char distribution also has a strong influence on the energy and exergy efficiencies. For instance, the increase of energy and exergy efficiencies from 75% to 86% and 67% and 70% were found on low  $O_2$  ER (0.14) when the char fraction to CR was elevated from 0.1 to 0.4, as depicted in Fig. 6a and b, respectively. This positive finding can be related to the minimum amount of unconverted char at a higher char fraction into CR due to a minor concentration of  $O_2$  in the GR. An interesting finding was found when the char fraction into CR was augmented from 0.4 to 0.7 as the energy and exergy efficiencies went down from 86% to 77 and 70%–65%, respectively. This observation indicates that at low  $O_2$  ER, the excessive char into CR delivers a negative effect on the gasification performance due to the low flow rate of syngas, though, the electricity production significantly increased with increasing char fraction into CR. In addition, there is energy lost during the conversion of thermal energy into electricity which contributes to the decrease of gasification performance.

#### 4.2. Steam to carbon ratio

The investigation was implemented by flowing different flow rates of boiler feed water (BFW) into BR at a constant flow rate of coal feed, to obtain the S/C ratio between 0.0 and 1.0 with a constant  $O_2$  ER of 0.21. One can easily notice that the steam addition has a good effect on the formation of  $H_2$  and  $CO_2$ , as illustrated in Fig. 7. For instance, the mole fraction of  $H_2$  and  $CO_2$  elevated from 0.40 to 0.62 and 0.00 to 0.04, respectively, when the steam is added from the S/C ratio of 0.0–1.0 at the char fraction to CR of 0.1 and the moisture content of 15%, as depicted in Fig. 7a. This expected observation can be associated with the existence of steam reforming of carbon reaction (Eq. (S6)) under  $H_2O$  atmosphere,

which converts steam and carbon into  $H_2$  and  $CO$ . Furthermore, the  $CO$  reacts with the excess  $H_2O$  to form  $CO_2$  and  $H_2$  by facilitating the water gas shift reaction, which also explains the decrease of  $CO$  mole fraction at a high S/C ratio. This explanation also valid for the identical phenomena when the moisture content was increased from 15% to 45%. The earlier experiments by Vijay et al. [15] using Indian coal also showed a similar conclusion that the injection of steam promotes  $H_2$  production. The identical trend was also observed on the various char fraction into CR, as shown in Fig. 7b and c, respectively. It is worth noting that the gasification of coal with the moisture content of 45% exhibited a slight decrease of  $H_2$  production at a high S/C ratio. This finding can be attributed to lower RD temperature, which inhibits the steam reforming reactions due to the excess amount of steam. The identical fashion is reported by Yang et al. [47] in their experiments using Mengdong coal in the absence of iron ore.

One can see that the effect of char fraction into CR on the syngas composition depends on coal moisture content, as shown in Fig. 7a, b and c. For the coal with a moisture content of 15%, the change of char fraction into CR has a minimum effect on the syngas composition. However, the char fraction into CR gives a strong influence on the distribution of major constituents in the syngas at the coal moisture of 45%. The reason for this lays in the amount of char in the RD which tends to react with steam. At low moisture content,  $H_2O$  plays a role as a limiting reactant given it enables the steam reforming of carbon reaction (Eq. (S6)), to convert char into syngas. The minor effect of char fraction into CR on the syngas composition indicates that the char fraction into CR up to 0.7 still resulted in the unconverted char due to lack of  $H_2O$ . Meanwhile, the high moisture content of coal gives sufficient  $H_2O$  for the complete conversion of char into syngas, which results in a significant change of syngas composition when the char distribution was varied. One can clearly

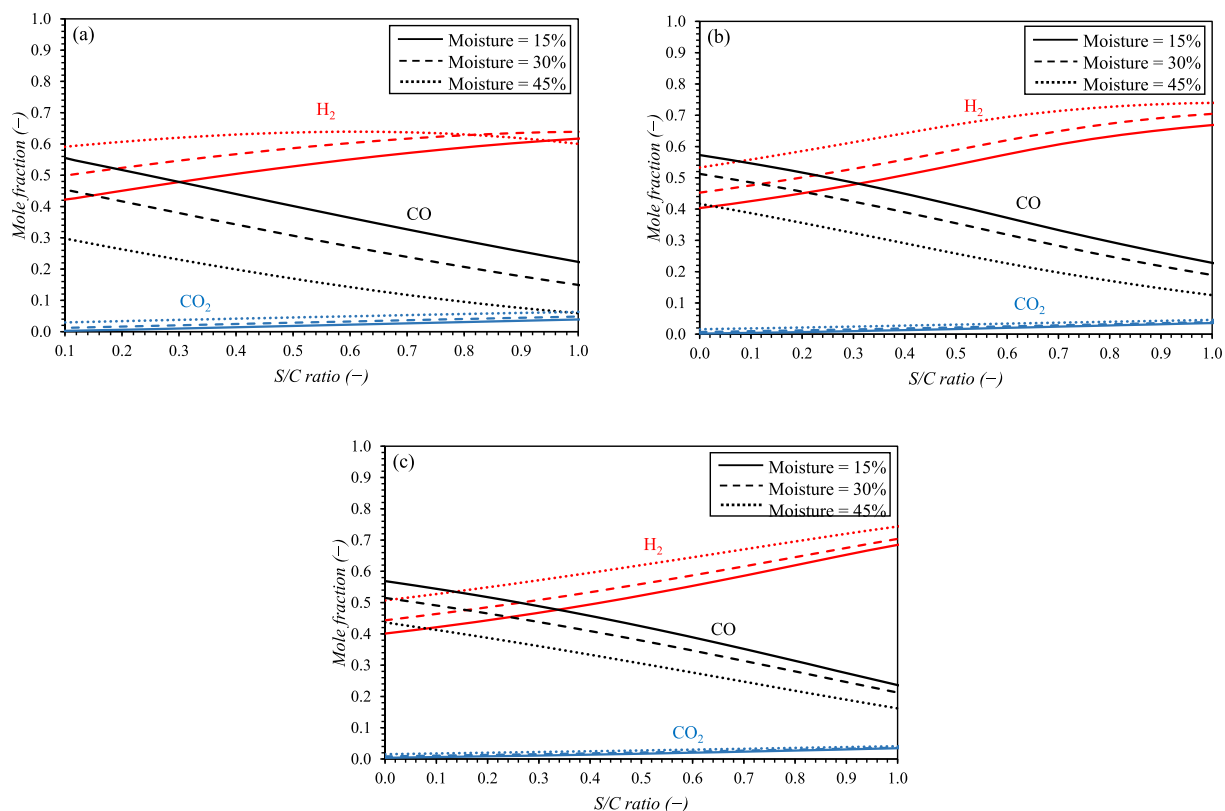


Fig. 7. The effect of S/C ratio on the major constituents in the syngas at different char distributions: (a) char to CR = 0.1, (b) char to CR = 0.4 and (c) char to CR = 0.7.

notice that the increase of char fraction to CR diminished the  $H_2$  mole fraction, while a positive effect was found on the CO mole fraction.

This result can be explained by higher RD temperature at higher fractions of char to CR due to the minimum amount of char in the GR, which promotes the reverse water-gas shift reaction (Eq. (S5)). This reason is supported by the fact that the  $CO_2$  mole fraction declines at higher char fractions to CR. Murakami et al. [48] also reported low  $H_2$  production at lower steam gasification temperature using Indonesian sub-bituminous coal in their experiments. The comparable results also reported by Wang et al. [49].

The change of the  $H_2/CO$  ratio with respect to the S/C ratio is presented in Fig. 8. One can see in Fig. 8 that the S/C ratio has a positive influence on the  $H_2/CO$  ratio. This is expected given high  $H_2O$  supply to the RD promotes  $H_2$  production via water-gas shift reaction (Eq. (S5)). The comparable phenomena were also observed on higher moisture contents. This finding is in line with the experimental work by Xiao et al. [50], showing the increase of the S/C ratio elevates the  $H_2$  production due to water-gas shift reaction. In addition, high moisture content allows the gasification system to provide a wide range of  $H_2/CO$  ratios. For instance, the  $H_2/CO$  ratio varies between 2.0 and 10.2 when the S/C ratio was changed between 0.1 and 1.0 at the char fraction to CR of 0.1, as shown in Fig. 8a. The effect of the S/C ratio on the  $H_2/CO$  ratio diminished when the char fraction to CR was increased to 0.4 and 0.7, as illustrated in Fig. 8b and c, respectively. The explanation of this result lays in the fact that a lower amount of char in the RD elevates the operating temperature which promotes the reverse water-gas shift reaction (Eq. (S5)). Thus, the steam injection is a suitable approach when one targets the specific value of the  $H_2/CO$  ratio in the syngas.

In regard to energy production, the gasification performance based on the CGE and power generation is depicted in Fig. 9. One

can see that both the CGE and power generation slightly decreased with increasing the S/C ratio. The decrease of CGE indicates that the energy required for steam generation is higher than the energy produced in the syngas. In addition, the high S/C ratio also diminished the power generation given the excess steam suppressed the RD temperatures. An interesting result was found when the char fraction to CR was varied from 0.1 to 0.7. At this range, the CGE decreased, while the power generation increased with increasing char fraction to CR. This result can be explained by the lower amount of syngas flow rate due to the minimum amount of char in the GR. This also causes a higher temperature of RD as the endothermic steam reforming of carbon (Eq. (6)) is minimal. Consequently, the CR received more char which leads to high temperature flue gas. The high-temperature of RD and CR products provide high energy in the power generation system, which results in an increase in electricity production.

The evaluation of the entire process showed that the addition of steam slightly reduced the gasification performance in term of energy conversion, as shown in Fig. 10. The explanation of this finding can be related to the decrease of syngas flow rate and heating value. This is confirmed by the identical trend of the CGE with increasing S/C ratio, as indicated in Fig. 9. One can clearly see that the overall energy efficiency is higher than the overall exergy efficiency. Again, this is because the exergy evaluation also considers the energy quality. This also explains the decrease of overall exergy efficiency with increasing the char fraction to CR, while the overall energy efficiency was relatively constant. One should note that at high char fraction to CR, most of the char is converted into  $CO_2$  by complete oxidation, which contains lower energy quality (i.e., lower chemical exergy) than its counterpart of CO. In addition, the energy loss in the power generation system also contributes to reduce the overall exergy efficiency.

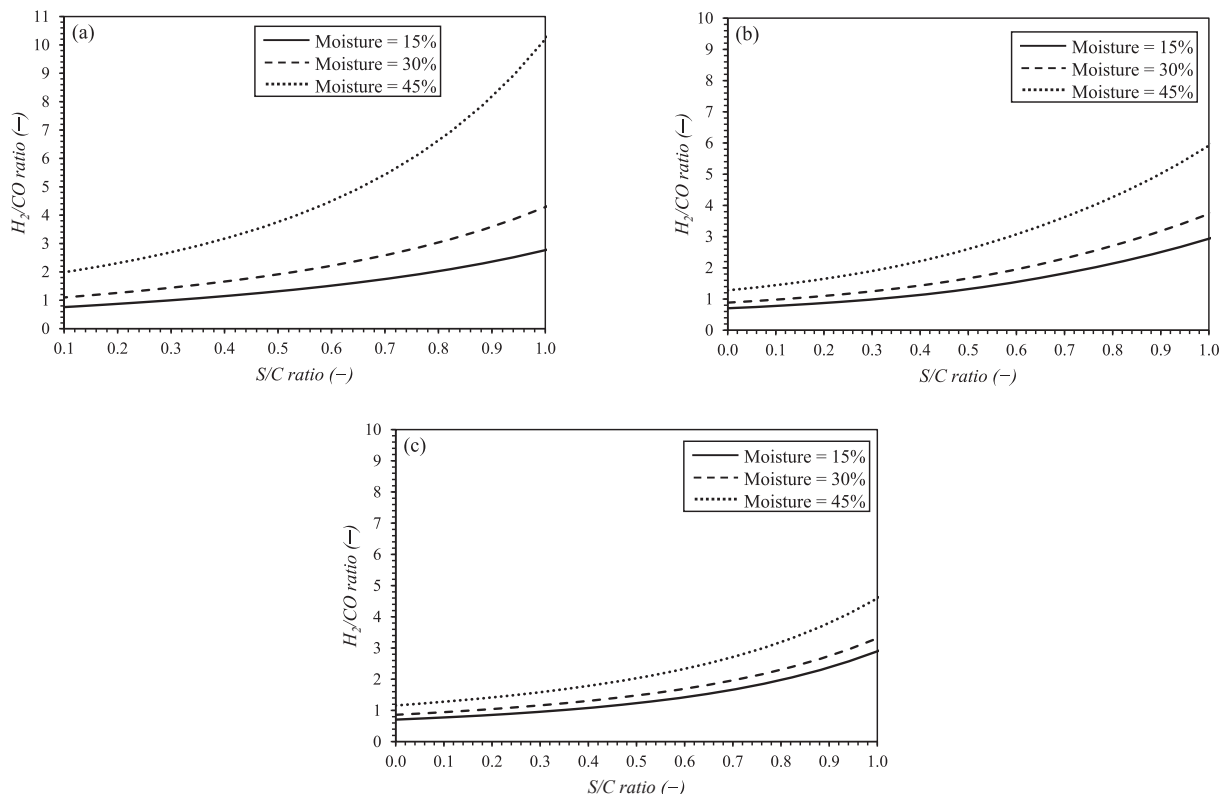


Fig. 8. The effect of S/C ratio on the  $H_2/CO$  ratio of the syngas at various char distribution: (a) char to CR = 0.1, (b) char to CR = 0.4 and (c) char to CR = 0.7.



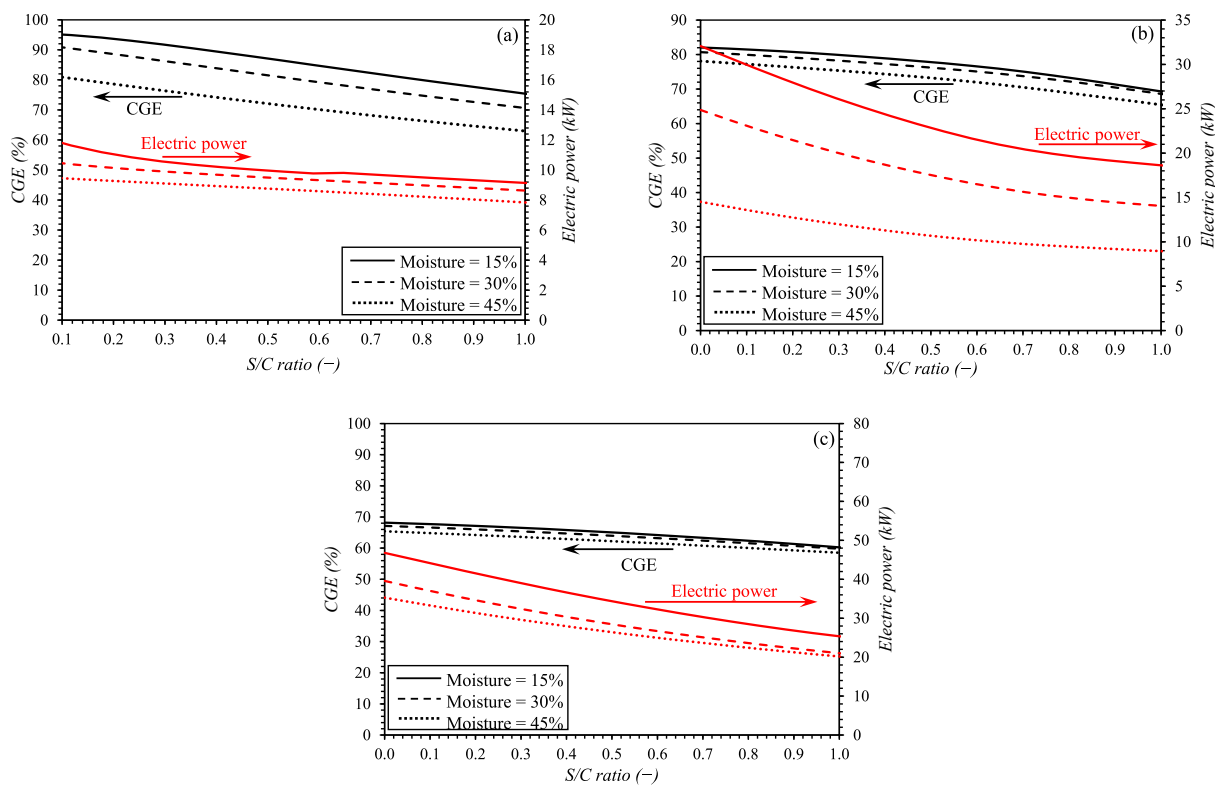


Fig. 9. The effect of S/C ratio on the CGE and electricity production at various char distribution: (a) char to CR = 0.1, (b) char to CR = 0.4 and (c) char to CR = 0.7.

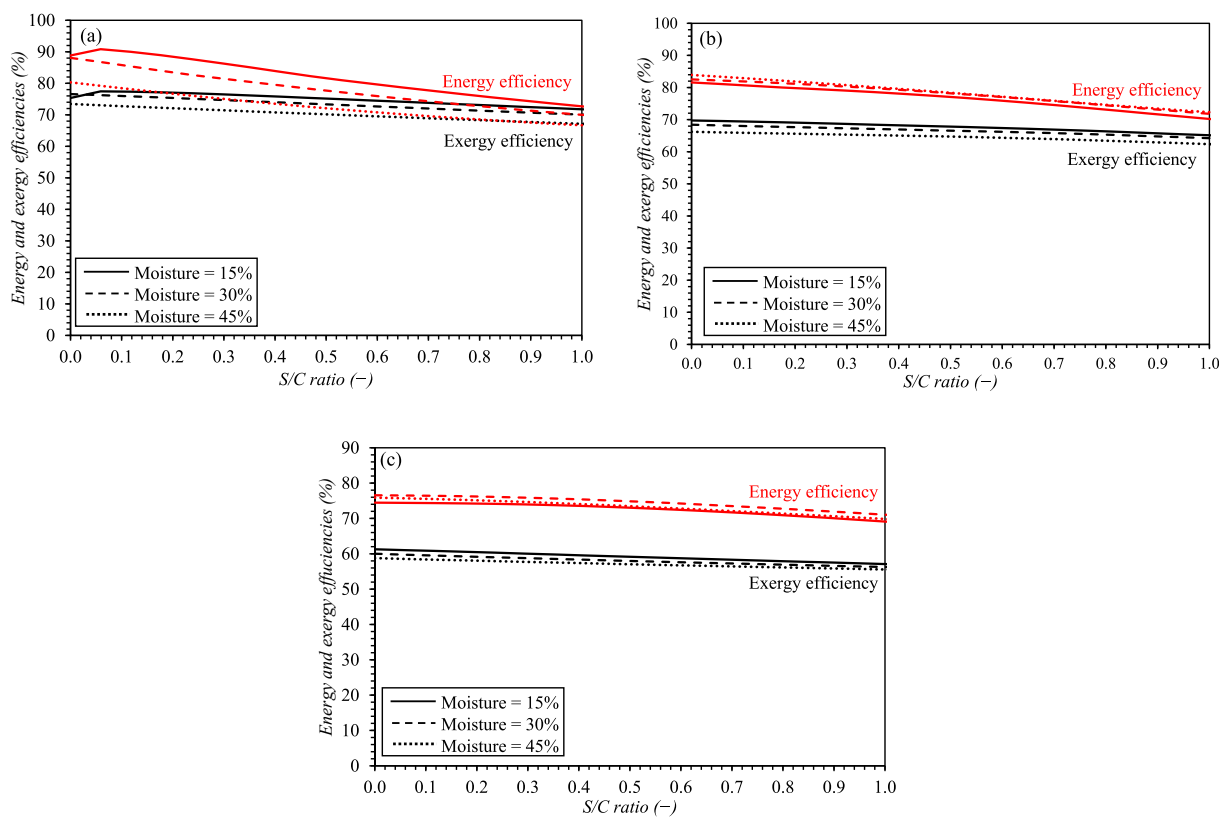


Fig. 10. The effect of S/C ratio on the overall energy and exergy efficiencies at various char distribution: (a) char to CR = 0.1, (b) char to CR = 0.4 and (c) char to CR = 0.7.

## 5. Conclusions

A novel gasification configuration was proposed to convert the low-rank coal into syngas and electricity. The gasification performance was evaluated based on the thermodynamic model in order to obtain the benchmark for the pilot-scale experiment. The present model exhibited a good agreement with the experimental parts as indicated by low relative error (below 10%) on the pyrolysis stage.

The use of O<sub>2</sub> as the gasifying agent provided high-quality syngas, which was indicated by high cold gas efficiency (up to 92%). The distribution of char into CR diminished the H<sub>2</sub>/CO ratio, but increased the electricity production. The present model enabled the adjustment of syngas composition, particularly the H<sub>2</sub>/CO ratio, by varying the steam flow rate and the char distribution to CR. The H<sub>2</sub>/CO ratio varied between 0.7 and 10.2. The moisture content of low-rank coal suppressed the gasification performance in terms of energy conversion (i.e., CGE, the overall energy efficiency and the overall exergy efficiency), but increased of H<sub>2</sub>/CO ratio of the produced syngas. The highest performance in terms of the overall energy efficiency (91%) and the overall exergy efficiency (79%) was found on the O<sub>2</sub> ER and S/C ratio of 0.21 and 0.06, respectively, with the char fraction to CR of 0.1.

The adjustable H<sub>2</sub>/CO ratio of syngas enables syngas utilization in the petrochemical industry. This can minimize the CO<sub>2</sub> emission given some portion of carbon from the low-rank coal is stored as petrochemical products. Recently, low-rank coal is only consumed by limited number of power generation station which leads to weaken its bargaining value from the economic perspective. The proposed gasification process potentially elevates the economic value of low-rank coal given it facilitates converting low-rank coal into flexible intermediate products with various application.

## Statement of author

M.A.A: Conceptualization, simulation and writing first draft. AH: Editing and revision. OM: Conceptualization, editing and revision. MMH: Simulation and revision.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.energy.2021.121505>.

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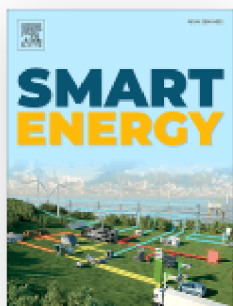
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