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Emergy evaluation of aromatics production from methanol and naphtha

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ABSTRACT

Aromatics are traditionally produced by the catalytic reforming of naphtha. However, with the demand of aromatics increasing and the reserves of petroleum resources declining, measures should be made to reduce the dependence of aromatics production on petroleum resources. Methanol-to-aromatics is proved to be an effective way to replace traditional naphtha-to-aromatics path. In order to compare the economic and environmental performance of aromatics production from naphtha and methanol, this paper carries out an emergy evaluation for each system by sorting out the simulation and literature data. Based on the emergy data collected, the emergy indices of each system are calculated. The results show that the sustainabilities of methanol-to-aromatics systems are higher than that of the naphtha-to-aromatics system, indicating the advantages of aromatics production from methanol. Among the methanol-to-aromatics systems, the aromatics from biomass-methanol system has the highest sustainability, indicating that the biomass based methanol-to-aromatics system is worth promoting. The sustainability indexes of methanol-to-aromatics systems based on coal and coke oven gas are less than 1, which means unsustainable. Meanwhile, the sustainability of natural gas based system is slightly higher than 1. The economic and environmental benefits of these systems can be optimized by improving resource utilization and reducing investment costs. Furthermore, the combination of different raw materials for methanol production should be considered.

1. Introduction

Benzene, toluene and xylene (BTX) are traditionally produced by continuous catalytic reforming of naphtha [1]. However, as the global demand for aromatic products increases and the reserves of petroleum resources decrease, alternative technology for BTX production needs to be carried out to reduce dependence on petroleum resources [2].

At present, the production capacity of methanol in China is surplus [3]. Therefore, the development of methanol-to-aromatics technology to replace petroleum is feasible [4]. In recent years, there have been some academic research [5] and industrialization attempts [6] on methanol-to-aromatics technology.

The raw materials for methanol production mainly include coal [7], natural gas [8], coke oven gas [9] and biomass [10]. At present, the economic and environmental analysis of methanol production by different routes has been performed. Li *et al.* [9] conducted a life cycle assessment of methanol production based on coke oven gas (COG) and compared it with coal and natural gas production routes. The results show that the environmental impact of the COG route is lower than that of the coal route. In addition, the methanol production cost of the COG route is 25.1% and 19.8% lower than that of the coal and natural gas routes, respectively. Chen *et al.* [7] also conducted a life cycle assessments on coal to methanol (CTM), coke oven gas to methanol (CGTM) and natural gas to methanol (NTM) systems. The results show that the CTM system has the greatest environmental impact. There is also a certain economic research on the overall route of methanol-to-aromatics basing on different raw materials. Niziolek *et al.* [8] proposed a comprehensive process framework based on system optimization to determine the most favorable process for the production of aromatics from natural gas. The results show that the net present value of aromatics production from natural gas is as high as 3800 MM$, and the investment cost is 65% less than that of a coal-to-aromatics system of the same scale. Niziolek *et al.* [11] quantified the effect that biomass type has on the overall profit of a refinery by investigating forest residues, agricultural residues and perennial crops as feedstocks. The economic analysis was carried out on the production of different ratios of *p*-xylene, *o*-xylene and *m*-xylene. The results show that the most profitable aromatics refineries produce p-xylene, while o-xylene refineries have the

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lowest required investment costs. Jiang *et al.* [12] presents a comparative techno-economic and environmental analysis of producing aromatics from methanol and naphtha. The results indicate that the net present value of producing aromatics from methanol is significantly higher than producing aromatics from naphtha. Moreover, methanol derived from natural gas leads to the least freshwater aquatic ecotoxicity potential, while that derived from biomass shows the lowest abiotic depletion potential, global warming potential, ozone layer depletion potential, and photochemical ozone creation potential.

The previous researches conducted economic and environmental analysis of methanol-to-aromatics systems. However, the sustainability of methanol-to-aromatics based on different raw materials has not been studied considering both economic and ecological performance.

Emergy theory was proposed by Odum [13], which was defined as the amount of available energy of one kind directly or indirectly used to make a service or product. Because the input and output could be considered systematically and converted into the same baseline [14], the emergy analysis can evaluate the economic and environmental performance of a system comprehensively [15]. Therefore, it is widely used in an industrial system [16] to evaluate the sustainability of the system by calculating emergy indices [17]. Cao and Feng [18] proposed a systematic procedure of emergy analysis in multi-product systems. They believe that the emergy for each product equals that for the entire system in an inseparable multiproduct system while in a semi-independent multi-product system, all the necessary input should be included and all the unnecessary inputs should be excluded.

In this paper, the emergy evaluation of aromatics production from methanol and naphtha is carried out. The methanol-to-aromatics systems include coal-methanol, natural gas-methanol, coke oven gas-methanol and biomass-methanol to aromatics systems. Firstly, the input and output data of each system are organized based on simulation data as well as relevant literature data. Secondly, the emergy transformities of each input factor are collected and transformed into the baseline in this paper. Meanwhile, the emergy indices of each system are calculated. Thirdly, the economic and environmental performances of each aromatics production system are analyzed based on the emergy indices. In addition, the sustainability indexes of different systems are compared and discussed.

2. Method and Data Collection

The steps of emergy evaluation mainly include the definition of research scale, determination of the emergy baseline, collection of the input and output data and calculation of the emergy indices.

2.1. Aromatics production systems

The aromatics production systems analyzed in this paper are shown in Fig. 1. The products of the systems are mainly benzene, toluene and xylene.

Fig. 2 is the emergy flow diagram of an aromatics production system, which is composed by the reforming reaction and aromatics separation parts. According to Fig. 2, the input of a system could be divided into three types, which are renewable emergy (R), nonrenewable emergy (N) and purchased emergy (F). The renewable emergy mainly includes the air, water and biomass while the nonrenewable emergy includes coal and nature gas. Other emergy inputs such as the capital costs, operation costs and labor costs belong to the purchased emergy.

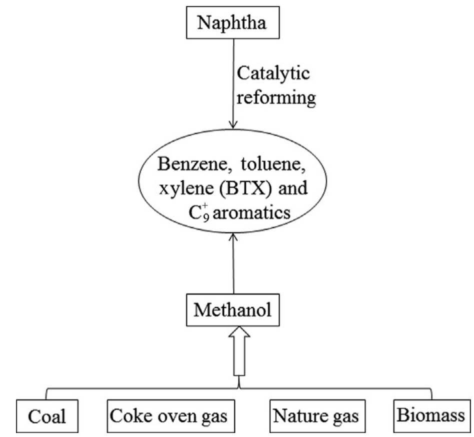

Fig. 1. Different pathways of aromatics production.

2.2. Data collection

The input and output data of each aromatics production system are collected and shown in Table 1. The data of aromatics production comes from process simulation [12], and the input data of the methanol production refers to the life cycle assessment of different methanol production systems by Chen *et al.* [7] and Khoo *et al.* [19]. The investment data of methanol production refers to the emergy evaluation of methanol production from different raw materials by Cao and Feng [18] and techno-economic analysis on methanol production from biomass by Yang *et al.* [20]. The scale factor is considered to be 0.7 [21].

The ecological emergy baseline used in the current emergy theory is composed of solar, geothermal and tidal momentum exergy received by the Earth [22]. There have been some different results of emergy baselines with different factors and methods into consideration. Odum firstly calculated the emery baseline as 9.44×10^{24} solar emjoules per year (sej·a^{-1}) in 1996 [13]. Brown and Ulgiati used the approximate solar equivalence ratio to represent the other two resources as the solar equivalent exergy and revised the emergy baseline as 12.1×10^{24} sej·a^{-1} [23]. Vilbiss *et al.* [24] used the gravitational potential energy dissipated in the generation of Earth's main renewable energy sources to calculate Earth's geo-biosphere emergy baseline. By this way, the emergy baseline is obtained, which is in the range of 11.1×10^{24} sej·a^{-1} and 13.8×10^{24} sej·a^{-1}. According to the studies of emergy baselines and the recent emergy evaluation work [17,25], the emergy baseline of 12.1×10^{24} sej·a^{-1} is adopted in this paper.

Table 2 shows the emergy transformities of different input elements. The values in the second and third columns of Table 2 are the emergy transformities and the emergy baselines in the references. All the emergy transformities from references are converted into the emergy baseline in this paper, which are shown in column 4 [23]. The emergy values of the input items are obtained by multiplying the input data in Table 1 and the corresponding emergy transformities in Table 2, which are completely shown in Table 3.

2.3. Method

The expressions and meanings of emergy indices used in this paper are shown as follows [13].

The emergy yield ratio (EYR) is the ratio of the total emergy output of a system to the purchased emergy input. A high EYR indicates that the system has little dependence on the purchased emergy and will be more competitive.

$$\mathrm{EYR} = \frac{Y}{F} = \frac{R+N+F}{F} \tag{1}$$

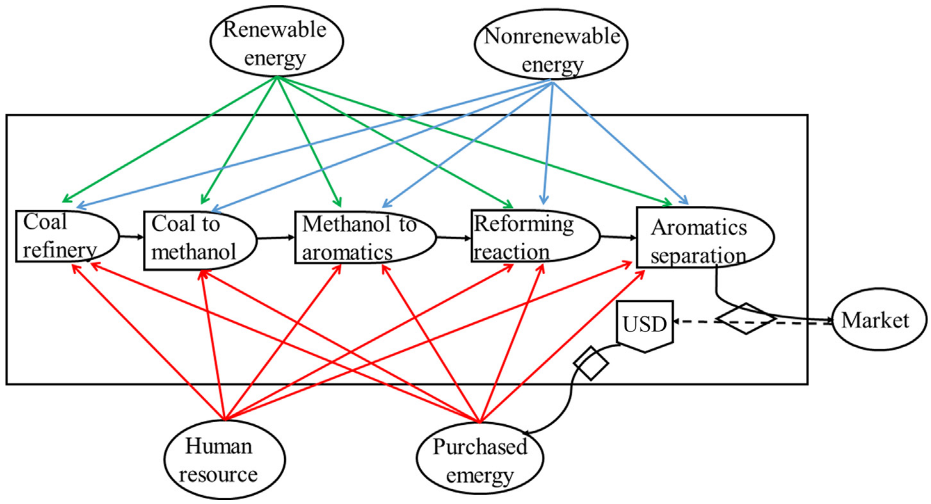

Fig. 2. Emergy flow diagram of an aromatics production system.

Table 1
Input and output data for different aromatics production systems

Items	Coal-methanol to aromatics	COG-methanol to aromatics	Nature gas-methanol to aromatics	Biomass-methanol to aromatics	Naphtha to aromatics
Renewable input					
Biomass/J·a^{-1}	–	–	–	1.30×10^{16}	–
Air/m^3·a^{-1}	5.26×10^9	3.28×10^9	–	1.53×10^9	–
Water/t·a^{-1}	2.99×10^8	8.76×10^7	8.87×10^7	6.59×10^7	3.96×10^7
Oxygen/m^3·a^{-1}	–	–	2.22×10^8	–	–
Nonrenewable input					
Naphtha/kg·a^{-1}	–	–	–	–	4.56×10^8
Electricity/J·a^{-1}	1.13×10^{16}	8.85×10^{15}	3.32×10^{15}	7.86×10^{13}	1.02×10^{14}
Coal/J·a^{-1}	7.03×10^{16}	6.13×10^{14}	2.17×10^{15}	–	–
Nature gas/J·a^{-1}	–	–	3.59×10^{16}	–	–
Purchased input					
Capital cost/USD·a^{-1}	6.11×10^7	3.6×10^7	2.96×10^7	1.99×10^8	5.12×10^6
Operation and maintenance/USD·a^{-1}	9.36×10^7	9.36×10^7	9.36×10^7	2.27×10^8	7.22×10^7
Utility/USD·a^{-1}	5.35×10^8	2.07×10^8	4.03×10^7	5.34×10^7	2.05×10^7
Product output					
H_2/kg·a^{-1}	–	–	–	–	4.65×10^7
Dry gas/kg·a^{-1}	7.76×10^7	7.76×10^7	7.76×10^7	7.76×10^7	1.86×10^6
LPG/kg·a^{-1}	3.07×10^8	3.07×10^8	3.07×10^8	3.07×10^8	2.05×10^7
Pentane/kg·a^{-1}	6.06×10^7	6.06×10^7	6.06×10^7	6.06×10^7	1.75×10^7
C_6^+ alkane/kg·a^{-1}	–	–	–	–	6.66×10^7
Benzene/kg·a^{-1}	2.41×10^7	2.41×10^7	2.41×10^7	2.41×10^7	2.37×10^7
Toluene/kg·a^{-1}	7.69×10^7	7.69×10^7	7.69×10^7	7.69×10^7	7.25×10^7
Xylene/kg·a^{-1}	1.58×10^8	1.58×10^8	1.58×10^8	1.58×10^8	1.33×10^8
C_9^+/kg·a^{-1}	4.45×10^7	4.45×10^7	4.45×10^7	4.45×10^7	7.38×10^7
H_2O/kg·a^{-1}	8.16×10^8	8.16×10^8	8.16×10^8	8.16×10^8	–

where Y is the total emergy input/output of a system. The emergy input of a system is equal to the emergy output of a system due to the total inheritance rule of emergy theory. R represents renewable emergy, N is nonrenewable emergy, and F represents purchased emergy.

The environmental loading rate (ELR) indicates the environmental pressure brought by the system during its production process. Therefore, if the system has a higher ELR, it will put greater pressure on the environment.

$$\mathrm{ELR} = \frac{N+F}{R} \tag{2}$$

The sustainability index (ESI) is the ratio of EYR to ELR. Generally, a system with an ESI higher than 1 is sustainable, but higher than 10 is a sign of insufficient resource utilization.

$$\mathrm{ESI} = \frac{\mathrm{EYR}}{\mathrm{ELR}} \tag{3}$$

The emergy transformity (Tr) is the ratio of total emergy input to energy/mass value of the product obtained. The Tr could be understood as the emergy consumed to obtain a unit product. Generally, a high Tr value indicates that the product has a high emergy level in the system. However, for a certain product, the low Tr value represents a less emergy consumption of the system. The aromatics production systems studied in this paper are inseparable multi-product systems since each product cannot be independently produced. As a result, each product inherits the total emergy input of the system [28]. Therefore, the Tr of each product is calculated as:

$$Tr_i = \frac{Y}{E_{\mathrm{i,\ output}}} \tag{4}$$

Table 2
Emergy transformities of input elements of the systems

Items	Reference transformities/sej·unit^{-1}	Reference baseline/sej·a^{-1}	Transformities in this paper/sej·unit^{-1}
Biomass/sej·J^{-1}	4.40×10^{4}	9.44×10^{24} [16]	5.59×10^{4}
Air/sej·m^{-3}	6.68×10^{10}	9.44×10^{24} [18]	8.49×10^{10}
Water/sej·t^{-1}	6.64×10^{11}	9.44×10^{24} [18]	8.44×10^{11}
Oxygen/sej·m^{-3}	6.35×10^{11}	9.44×10^{24} [18]	8.06×10^{11}
Naphtha/sej·kg^{-1}	2.99×10^{12}	9.44×10^{24} [26]	3.80×10^{12}
Electricity/sej·J^{-1}	1.59×10^{5}	9.44×10^{24} [27]	2.17×10^{5}
Coal/sej·J^{-1}	4.00×10^{4}	9.44×10^{24} [13]	5.08×10^{4}
Nature gas/sej·J^{-1}	4.80×10^{4}	9.44×10^{24} [13]	6.10×10^{4}
Cost/sej·USD^{-1}	1.42×10^{12}	12.1×10^{24} [17]	1.42×10^{12}

Table 3
Input and output emergy values of the systems

	Coal-methanol to aromatics/sej	COG-methanol to aromatics/sej	Nature gas-methanol to aromatics/sej	Biomass-methanol to aromatics/sej	Naphtha to aromatics/sej
R					
Biomass	–	–	–	7.26×10^{20}	–
Air	4.47×10^{20}	2.78×10^{20}	–	1.30×10^{20}	–
water	2.52×10^{20}	7.40×10^{19}	7.49×10^{19}	5.56×10^{19}	3.34×10^{19}
Oxygen	–	–	1.79×10^{20}	–	–
N					
Naphtha	–	–	–	–	1.73×10^{21}
electricity	2.44×10^{21}	1.92×10^{21}	7.21×10^{20}	1.46×10^{20}	2.21×10^{19}
Coal	3.57×10^{21}	3.11×10^{19}	1.10×10^{20}	–	–
Nature gas	–	–	2.19×10^{21}	–	–
F					
Capital cost	8.67×10^{19}	5.12×10^{19}	4.20×10^{19}	2.82×10^{20}	7.27×10^{18}
Operation and maintenance	1.33×10^{20}	1.33×10^{20}	1.33×10^{20}	3.22×10^{20}	1.03×10^{20}
Utility	7.59×10^{20}	2.94×10^{20}	5.72×10^{19}	7.58×10^{19}	2.90×10^{19}
Y					
Total emergy input/output	7.69×10^{21}	2.78×10^{21}	3.51×10^{21}	1.73×10^{21}	1.93×10^{21}

in which $E_{i,\text{output}}$ is the output of the ith product.

According to the emergy transformities, the emergy value could be obtained by Eq. (5):

$$Em_i = E_{i,\text{output}} \times Tr_i \tag{5}$$

in which Em_i is the emergy value of the ith item.

3. Results and Discussion

Fig. 3 shows different types of emergy inputs for each aromatics production system. For comparison, aromatics production of each system is consistently set to be 300 kt·a^{-1}. According to Fig. 3, the nonrenewable emergy input of the biomass based methanol-to-aromatics system is the lowest, for the reason that the raw materials used by the system are renewable resources. As for other systems, the coal based methanol to aromatics system has the largest demand for nonrenewable resources. The reason is that coal mining, washing and gasification processes in the synthesis of coal-based aromatics production requires a large amount of electricity, which causes the highest nonrenewable emergy input.

3.1. Comparison of emergy indices

Fig. 4 is a comparison of the emergy transformities for benzene, toluene and xylene products in each system. The products of biomass based methanol-to-aromatics system have the lowest emergy transformity, indicating that this system is superior to others in terms of emergy input consumption. The reason is that in the methanol production stage, the addition of steam and CO_2 into gasification improved the methanol yield. In addition, the water consumption of biomass to methanol process is much lower than the conventional coal to methanol process, indicating a better resource utilization efficiency. Therefore, the emergy consumption of the biomass based aromatics is lower than other resource based aromatics to perform a unit of product [20]. Otherwise, the emergy transformities of the naphtha-to-aromatics system are relatively low, which is close to those of the biomass based methanol-to-aromatics system. The reason is that the technology of naphtha-to-aromatics system is relatively mature, which leads to less investment cost to produce the same amount of aromatics.

Fig. 5 shows the emergy yield ratio (EYR) of different systems. The EYR value is mainly affected by the proportion of purchased emergy. Consequently, it could be optimized by decreasing the investment cost. According to Fig. 5, the natural gas based methanol-to-aromatics system has the highest EYR, mainly because the process for nature gas-to-methanol is relatively simple and the product quality is high. Therefore, it has better economic benefits than other systems. The naphtha-to-aromatics system is superior to other methanol-to-aromatics systems due to its mature technology and lower investment cost. The biomass based methanol-to-aromatics system has the lowest EYR, for the reason that the system has a high investment cost in the methanol production stage. Therefore, the economic benefits of this system can be improved by reducing the investment cost of the biomass-to-methanol process.

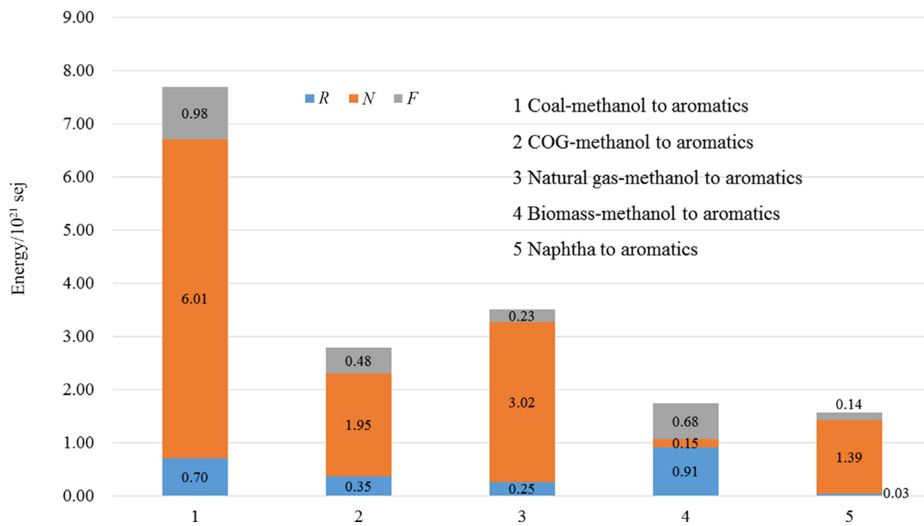

Fig. 3. Emergy input of different aromatics production systems.

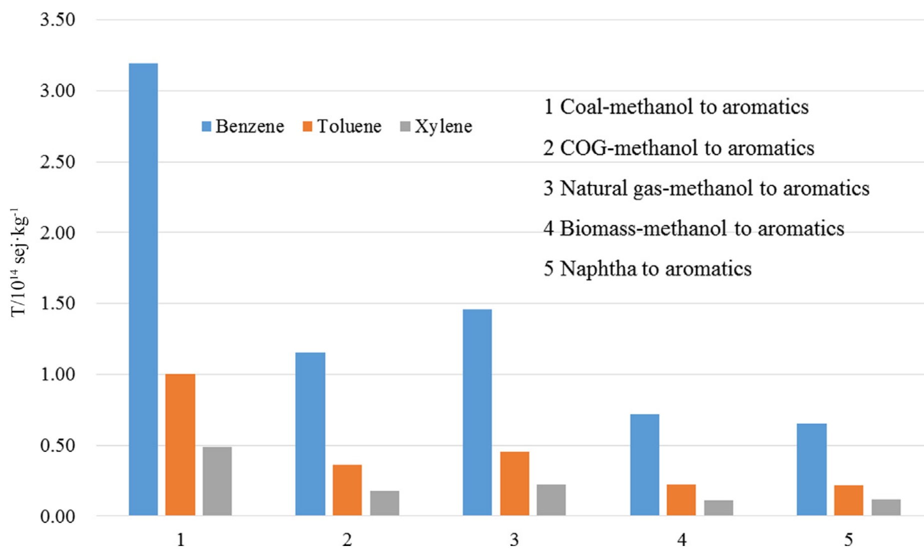

Fig. 4. Emergy transformities of different aromatics production systems.

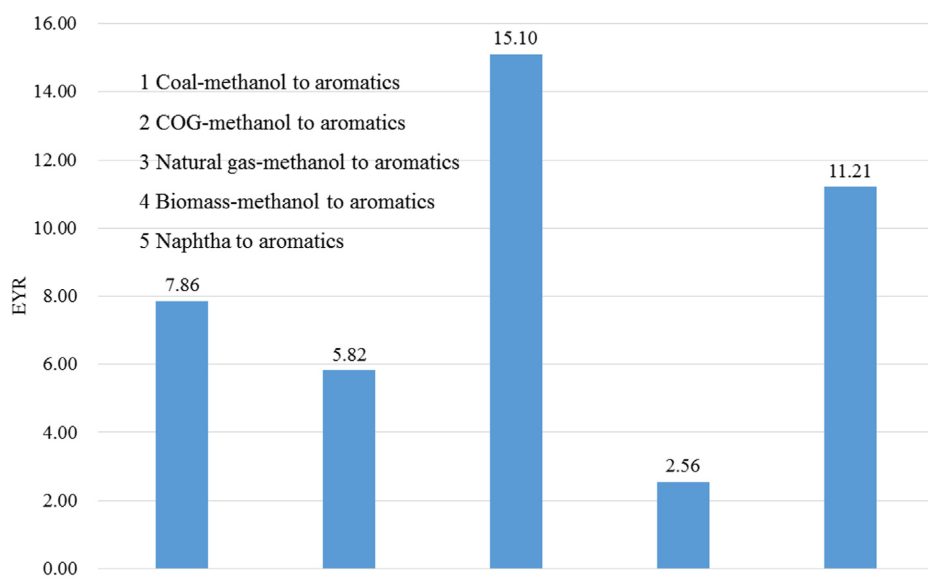

Fig. 5. Emergy yield ratio of different aromatics production systems.

Fig. 6 is the environmental loading rate (ELR) of different systems. The ELR is the ratio of the non-renewable and purchased emergy to the renewable emergy. Therefore, a high renewable emergy proportion will decrease the ELR value while a high non-renewable and purchased emergy proportion will increase the ELR value. As a result, the environmental loading could be reduced by decreasing the consumption of non-renewable and purchased emergy. According to Fig. 6, the ELR of the naphtha-to-aromatics system is higher than that of all methanol-to-aromatics systems, mainly due to its higher proportion of non-renewable resources. Therefore, in terms of environmental benefits, methanol-to-aromatics system is worthy promoting. Among the methanol-to-aromatics systems, the value of ELR has a great relationship with the source of methanol. The order of ELR from high to low is natural gas, coal, coke oven gas and biomass based methanol-to-aromatics systems. The coke oven gas based methanol-to-aromatics system has a benefit of waste utilization, so its ELR is lower comparing to coal and natural gas based systems. The biomass based methanol-to-aromatics system has a lowest ELR due to the high proportion of renewable resource input.

Fig. 7 is a comparison of the sustainability index (ESI) of each system. The ESI is the ratio of the EYR to the ELR, which represents the economic benefit obtained by unit environmental press. The sustainability of the system could be improved by improving the utilization efficiency and reduce the environmental impact. The ESI of the naphtha-to-aromatics system is lower than that of all the methanol-to-aromatics systems, which further demonstrates the advantages of the methanol-to-aromatics systems. For the

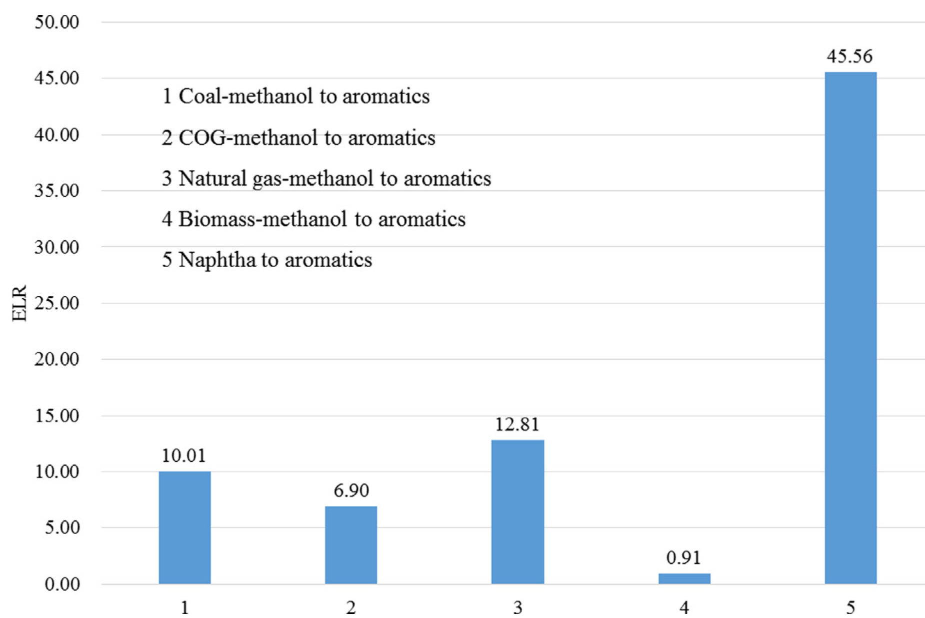

Fig. 6. Environmental loading rate of different aromatics production systems.

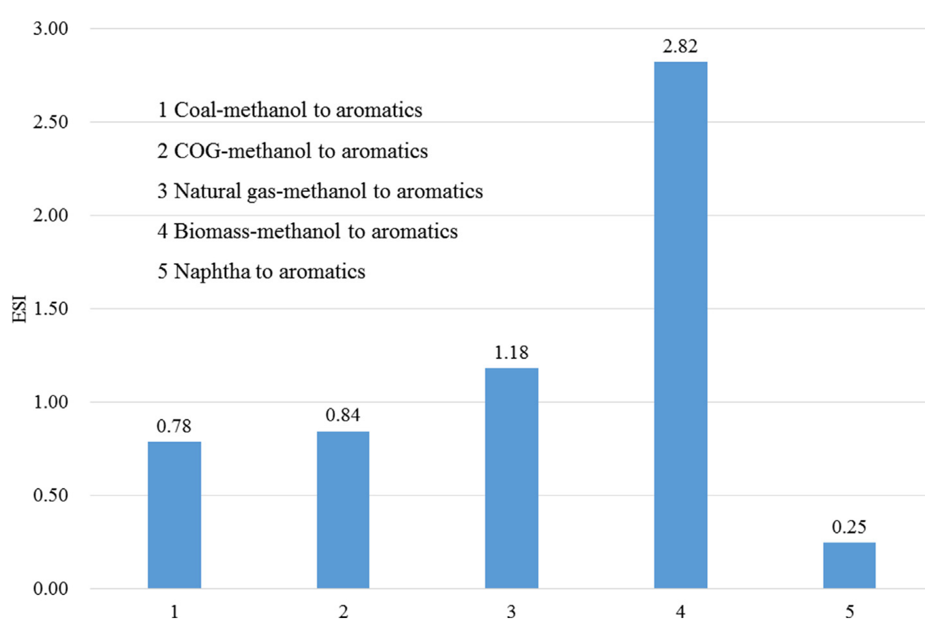

Fig. 7. Sustainability indexes of different aromatics production systems.

Table 4
Emergy indices comparison of different methanol to aromatics systems

Systems	EYR	ELR	ESI
Coal based methanol to aromatics	7.86	10.01	0.78
Coke oven gas based methanol to aromatics	5.82	6.90	0.84
Nature gas based methanol to aromatics	15.10	12.81	1.18
Biomass based methanol to aromatics	2.56	0.91	2.82
Combination of different raw materials	6.63	6.09	1.09

inner comparison of methanol-to-aromatics systems, the biomass based methanol-to-aromatics system has the highest ESI, which indicates a good development prospect. In the future, the technology could be improved and investment cost should be reduced to further improve the sustainability of the system. The ESIs of the coal and coke oven gas based methanol-to-aromatics systems are similar and less than 1, which means unsustainable. The ESI of natural gas based system is slightly higher than 1 and the sustainability still need to be improved. The economic and environmental benefits of these systems can be optimized by improving resource utilization efficiency and thereby increasing their sustainability.

3.2

Based on the above analysis, different raw materials based methanol production systems have advantages and disadvantages. In order to overcome the shortcoming of each system, the combination of different raw materials is studied. Different kinds of raw materials are considered to be evenly distributed in the combined system. Therefore, the emergy indices of the combined system could be obtained by superposing the emergy data of each methanol to aromatics system. The comparison of emergy indices are shown in Table 4. The EYR is improved comparing to the biomass based methanol to aromatics system and the ELR is reduced comparing to the coal, coke oven gas and natural gas based systems. The ESI of the combined system is higher than 1, which means sustainable. The results indicate that it is potential to combine different raw materials for aromatics production. In the further study, the proportion of different raw materials can be optimized to improve the sustainability of the system.

4. Conclusions

Based on the simulation data of the aromatics production system and relevant literature data, this paper analyzes and compares different aromatics production systems from the perspective of emergy. The following conclusions could be got.

1、甲醇制芳烃经济效益不如石脑油制芳烃，但环境效益优于石脑油制芳烃

(1) Overall, the methanol-to-aromatics system is inferior to the naphtha-to-aromatics system in terms of economic benefits. However, its environmental benefits is better than naphtha-to-aromatics system.

2、基于生物质甲醇制芳烃拥有最佳环境效益，比其他芳烃生产系统更具有可持续性

(2) The performance of the methanol-to-aromatics systems has a clear relationship with the source of methanol. The biomass based methanol-to-aromatics system has the best environmental benefits and is more sustainable than other aromatics production systems.

(3) The natural gas based methanol-to-aromatics system has the highest EYR and its ESI is higher than other systems except biomass based system. However, its ELR is also higher than that of other methanol-to-aromatics systems. Therefore, reducing the consumption of nonrenewable resources by improving the utilization efficiency of natural gas could thereby improve its environmental performance. The EYR and ELR of the coal and coke oven gas based methanol-to-aromatics systems are moderate, and the ESIs of the systems are higher than that of the traditional naphtha-to-aromatics system.

(4) In order to further optimize the emergy proportion and emergy indices of aromatics production from methanol, the combination of raw materials could be considered. The proportion of different raw materials for aromatics production could be optimized in the further study.

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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Nomenclatures

$E_{i,\text{output}}$	energy/Mass value of the *i*th product output
Em_i	emergy value of the *i*th product
ELR	environmental loading rate
ESI	sustainability index
EYR	emergy yield ratio
F	purchased emery
N	non-renewable emergy
R	renewable emergy
Tr	emergy transformity
Y	total emergy input/output

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