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Carbon footprint evaluation of coal-to-methanol chain with the hierarchical attribution management and life cycle assessment

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ABSTRACT

Coal is considered as an abundant energy source in China and coal-to-methanol chain is an essential routing on account of methanol's irreplaceable status in chemical industries. However, coal-based methanol production aroused controversy due to its intensive energy consumption and high greenhouse gas emission, compared with other processes by oil or natural gas. Carbon footprint is an improved indicator that evaluates both direct and indirect greenhouse gas emissions in the life cycle perspective and guides policymakers for better industry-chain planning. In this study we proposed the idea of hierarchical attribution management (HAM) to provide a classified method for evaluating carbon footprint of coal-to-methanol chain, combined with life cycle assessment (LCA) and the tool of ASPEN Plus. The results show that the life cycle carbon footprint was 2.971 t $CO_{2,e}$/t methanol. By the HAM, it's concluded that methanol production process was the largest emission contributor in the defined life cycle system with a share of 92.86%, followed by coal mining process with 4.34%. Gasification unit and water-gas shift unit were two major greenhouse gas generators, accounting for 21.26% and 52.80% of life cycle emission, respectively, while methanol synthesis unit showed the potential for CO_2 utilization and emission reduction. Additionally, the results of sensitivity analysis showed that electricity emission factor with a sensitivity factor of 189.11 was the most extensive influence factor on life cycle emission due to its widest application. The discuss on effects of CCS on life cycle emission showed that carbon footprint approximately decreased by 64.9% when the methanol plant was retrofitted with CO_2 capture and compression, indicating that CCS is an effective way to alleviate global warming.

1. Introduction

The utilization of fossil fuels has triggered global climate change, and the Intergovernmental Panel on Climate Change (IPCC) declared that anthropogenic greenhouse gas (GHG) emission contributes the most to global warming phenomenon [1]. To achieve sustainable development, diverse kinds of new energy resources are regarded as alternative energy with great promise in the future, such as nuclear energy [2], solar energy [3], bio-energy [4], hydrogen energy [5]. Furthermore, other measures have been taken to alleviate global warming, including CO_2 capture and storage (CCS) project to inject captured CO_2 into geological formations, process integration techniques to improve system efficiency and reduce the demand of fossil fuels [6], of which CCS could contribute 19% of total GHG emission reduction to achieve the 2 degree target in 2050 [7].

CCS technology is generally recognized as a feasible solution to address the global warming in a brief period and it covers four processes: capture, compression, transport and storage [8,9]. Significant efforts have been made to overcome technical and economic obstacles, especially for CO_2 capture and CO_2 storage. To date, the most commercial mature application of CO_2 capture in power plant was based on chemical absorption, but it still caused about 10% efficiency penalty independent of power plant types and coal types [7]. Most of the researches focus on the effects of CO_2 capture and compression on different types of power plant in the perspectives of CO_2 capture technologies, economic assessment, feasibility, and so on. In the step of CO_2 storage, the main concern is focused on the risk of CO_2 leakage and slow rate of CO_2 dissolution in geological formations [10,11].

Most chemical processes generate unavoidable GHG emission as by-product due to the unbalanced H/C ratio for utilization,

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Nomenclature

CF_{lc}	life cycle carbon footprint, t $CO_{2,e}$/t methanol
C_j	amount of emission sources consumed, t/y
$CO_{2,e}$	CO_2 equivalent emission, t/y
EM_i	greenhouse gas emission in i process, t/y
EF	emission factor
ELI	electricity intensity, kWh/t raw coal production
EI_{tr}	energy intensity of different transport pattern, MJ/(t·km)
P_{coal}	annual coal production, t/y
Q_{coal}	demanded raw coal of the life cycle coal-to-methanol chain, t/y
R_{HC}	molar ratio of $(H_2\text{-}CO_2)/(CO + CO_2)$
R_S	ratio of syngas into WGS
$Y_{methanol}$	annual yield of methanol product, t/y
ε	carbon footprint reduction efficiency
η	carbon footprint reduction penalty

Subscripts

i	four defined processes in coal-to-methanol chain
cm	coal mining process
ws	coal washing and selection process
transp	coal transport process
mp	methanol production process
pe	process energy for transport vehicles
cc	CO_2 capture and compression
ref	basic coal-to-methanol chain
retr	retrofitted coal-to-methanol chain

Abbreviations

ASU	air separation unit
CCS	CO_2 capture and storage
CRE	carbon footprint reduction efficiency
CRP	carbon footprint reduction penalty
CWS	coal-water slurry
CBM	coal bed methane
GHG	greenhouse gas
GWP	global warming potential
HAM	hierarchical attribution management
LCA	life cycle assessment
WGS	water-gas shift

especially in coal-based industry. Plants equipped with different techniques for flue gas treatment resulted in different efficiency penalties and costs [12]. Fortunately, modern coal chemical industry based on gasification technology has the basis for the application of CCS in this field because generally there already exists the function of CO_2 capture in the unit of H/C ratio adjustment and syngas cleaning, such as water-gas shift and Rectisol wash process, leading to the potential reduction of capital investment for CO_2 capture.

In China, coal has been playing a crucial role as an abundant energy source, contributing to approximately 70% of primary energy from 2000 to 2012, as shown in Fig. 1. Nowadays, coal chemical industry is in a state of rapid development for chemical products instead of oil and natural gas, which whereas emits vast amounts of GHG and other pollutants.

Methanol production is considered as a hub of prosperous chemical industry network and its product is always served as the raw material for producing other chemicals, such as dimethyl ether (DME), acetic acid, acetic oxide, methyl formate, formic acid and oxalic acid, as shown in Fig. 2. In the short and long term, methanol is manufactured by the technologies mainly based on coal, natural gas and coke-oven gas [14], even though biomass-based approach has been deemed as a promising way in some researches [15,16]. What's more, the methanol economy could contribute to a sustainable future where carbon-neutral methanol is produced from biomass and recycled carbon dioxide, appearing to fit with the Nobel Laureate George Olah's vision of Methanol Economy [17]. However, coal-based methanol production aggravated water shortage and increased greenhouse gas emission, which run counter to the theme of cleaner production. Therefore, the evaluation for GHG impact of coal-to-methanol chain would assist the identification of emission distributions.

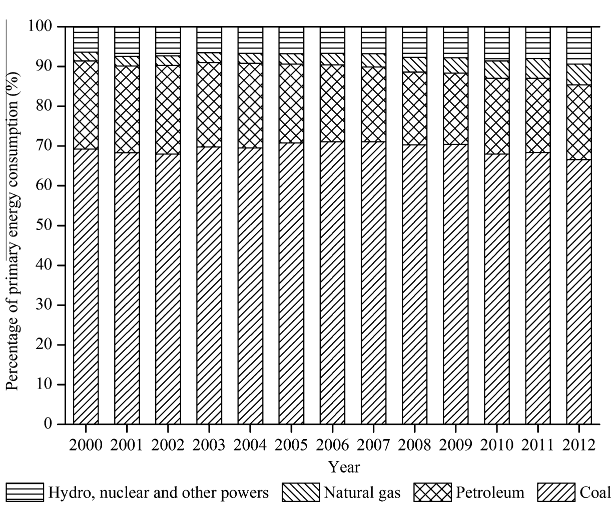

Fig. 1. Structure of primary energy consumption in China [13].

Carbon footprint (CF) has been widely accepted as an advanced evaluation indicator to raise public consciousness about the threat of global climate change. It is a measure of the total GHG emission that is directly and indirectly caused by a process or product over the life stages [18]. There have been a number of works associated with the carbon footprint analysis to assess the environmental impact of products [19], persons [20], regions [21] or technologies [22]. These researches were generally conducted by using the life cycle assessment (LCA) software like SimaPro, Gabi [23], Ecoinvent [24] and Umberto Carbon Footprint [25]. In addition, several methodologies for carbon footprint analysis have been developed, for instance, the IPCC method [1], process-based life cycle analysis (PLCA) [26] and input–output analysis (IOA) [27].

Up to now there have been some studies about coal-derived methanol for vehicles in the view of cradle-to-grave to evaluate the environmental impact. Li et al. [28] analysed the possibilities of using methanol as a hydrogen carrier by the 3E (energy, environmental, and economic) analysis and showed that the coal-derived methanol pathway with distributed reforming utilities was well suited for China's specific energy situation due to its kind environmental effect. Ou et al. [29] compared the GHG impact of vehicles driven by coal-to-liquid fuels and coal-based electricity and concluded that electric vehicles achieved better environmental performance than the vehicles fuelled with coal-to-liquid even if CCS technology was employed. Wei et al. [30] analysed the environmental impacts of