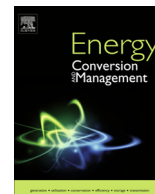




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Utilization of ventilation air methane as an oxidizing agent in chemical looping combustion

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ABSTRACT

Release of fugitive methane (CH₄) emissions from ventilation air in coal mines is a major source of greenhouse gas (GHG) emissions. Approximately 64% of methane emissions in coal mine operations are the result of VAM (i.e. ventilation air methane) which is difficult for use as a source of energy. A novel ancillary utilization of VAM was thereby proposed. In this proposal, the VAM was utilized instead of air as a feedstock to a chemical looping combustion (CLC) process of coal. In this case, Fe₂O₃/Fe₃O₄ particles were shuttled between two interconnected reactors for combustion of syngas produced by an imbedded coal gasifier.

The effect of VAM flow rate and methane concentration on the performance of CLC was analyzed thermodynamically using Aspen Plus software. Results indicated that the variations of air reactor temperature with VAM flow rate and methane concentration can be minimized as expected. The effect of temperature and inlet methane concentration on the conversion of CH₄ was examined experimentally in a fixed bed reactor with the presence of particles of Fe₂O₃/Al₂O₃. Not surprisingly, the reaction temperature put a significant influence on the conversion of CH₄. The conversion started at the temperature about 300 °C and the temperature to achieve full conversion was around 500 °C while the temperature in empty reactor between 665 °C and 840 °C. This is due to the catalytic effect of oxygen carriers (i.e. Fe₂O₃/Al₂O₃) on the conversion of methane. Moreover, it was observed that the methane conversion rate decreased with the increase in inlet methane concentration while increasing with Fe₂O₃ loading content.

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1. Introduction

CH₄ is the third most important greenhouse gas after H₂O vapor and carbon dioxide (CO₂) and has a Global Warming Potential (GWP) 25 times that of CO₂ on a 100-year timescale. The release of fugitive methane (CH₄) from coal mines is the major source of GHG emissions, accounting for 8% of the total anthropogenic methane emissions. Typically it is emitted from a coal mine in the form of ventilation air methane (VAM) (0.1–1 vol% CH₄) or coal seam gas (30–95 vol% CH₄) [1]. Approximately 64% of methane emissions in coal mines are the result of VAM which is found to be difficult to mitigate primarily because (1) the methane concentration in the mixture is dilute and (2) the concentration and flow rate of methane is variable.

Several methods have been developed to mitigate the emissions of VAM, which can be categorized into principle use and ancillary use. The principle utilization technology is referred to the use of methane in VAM as the energy source and no need for additional

fuel in assistance to the ignition of methane. In terms of the reaction kinetic mechanism, the existing principle utilization technologies for mitigation of VAM in mining operations can be classified into thermal oxidation and catalytic combustion [1]. The only difference between these two processes is with respect to the use of catalyst. Based on the thermal oxidation principle, a thermal flow-reversal reactor (TFRR) was offered by MEGTEC. But it suffers from an extremely high auto-ignition temperature (above 1000 °C) which causes plenty of energy loss to start the operation [2]. It is also a great challenge to maintain such a high temperature level for a long term run as far as the huge flow rate is concerned. To lower the start temperature, a catalytic flow-reversal reactor (CFRR) was developed by CANMET. As a result of this new technology, the auto-ignition temperature is decreased by several hundred degrees Celsius [2]. Although the CFRR delivers some advantages over the TFRR, it comes up with some challenges on the design and operation. For instance, it needs to consume some expensive metals or metal oxides as catalyst like Pd, Pt, Rh or PdO [3–6]. Also its reactor design is more complicated in order to get enough contact surfaces. The performance of CFRR can be improved using the design of catalytic monolith reactor (CMR) developed by CSIRO,

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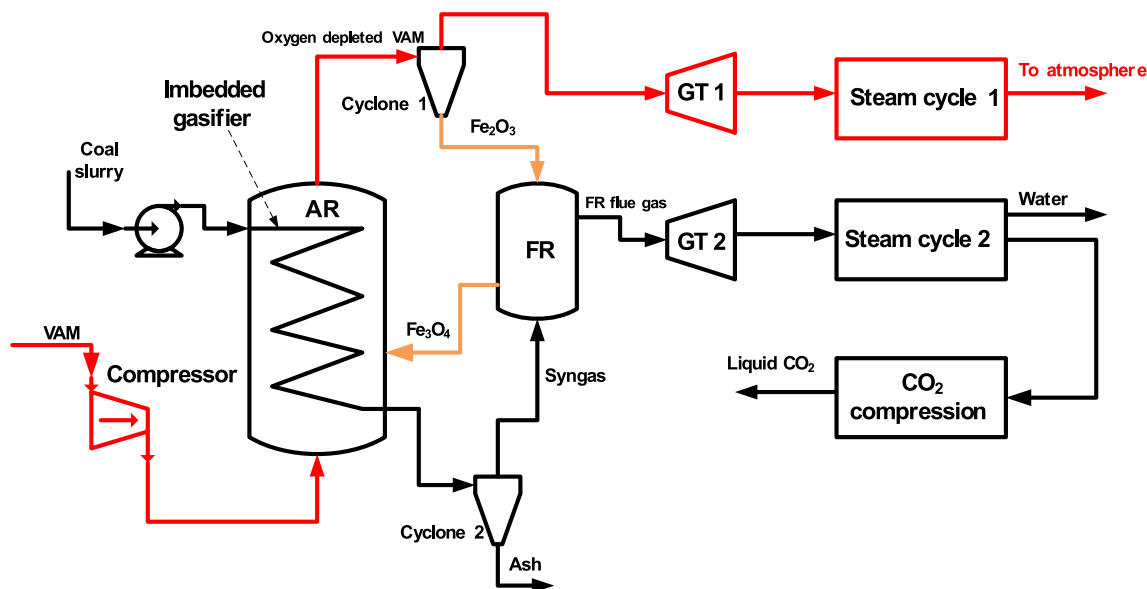


Fig. 1. Flowsheet of the proposed combustion method of VAM.

Australia [4]. This advanced technology is in common use due to its low pressure drop at high mass flow rates and high geometric surface area. However, it can only treat VAM stream with a minimum methane concentration of 0.4 vol% [1].

Relevant studies on catalytic combustion of methane have been carried out for many decades. It was observed that noble metals or metal oxides showed the best activity for methane oxidation at low temperatures. Particularly supported Pd, Pt or PdO has been extensively employed as catalyst on methane combustion [7]. It was believed that the higher reactivity is resulting from the larger surface area and therefore much more active sites. However, they are suffering from some demerits like poor availability and economics. To overcome the above challenges, some non-noble materials are adapted to replace the role of these noble metals, such as Co_3O_4 , CuO, and alumina industry waste or perovskite-type oxides [8–11]. Though they are not as active as noble metals, they possess better thermal stability and resistance to sintering. Iron oxide, as an industry abundant and cheap material, has the potential to be an alternative for complete oxidation of methane as it has been proven to be a good catalyst in a wide field of application like hydrogenation and hydroliquefaction. Barbosa [12] suggested that unsupported iron oxide presented remarkable initial activity but the stability was undermined at the operation temperatures. Fe_2O_3 supported by MgO or $\text{Ce}_{0.67}\text{Zr}_{0.33}\text{O}_2\text{-Al}_2\text{O}_3$ showed better performance in activity and the optimized loading content is found at 6–8% [13,14]. Three iron-based waste materials from aluminium industry were used for catalytic combustion of methane [15]. It was shown that the activity is increasing with the increase in hematite content.

There could be no doubt on the technical and economic feasibility of applying TFRR, CFRR and CMR technologies to mitigate ventilation air methane when the CH_4 concentration in air exceeds the minimum requirement. However, for some cases the continuous operation of these units may need additional fuel due to fluctuation in methane concentration which may bring the CH_4 concentration to levels below the minimum requirement and blowout in a short period. In addition, heat recovery from these units for power generation cannot be accomplished unless additional fuel is applied to increase methane concentration to a constant level, perhaps, at least 0.9 vol% according to the current design practices [1].

Ancillary use of VAM could be another noteworthy option applicable to many industrial settings. Normally speaking, it involves

using VAM to totally or partly replace air in a combustion process. Some applications can be found in internal combustion engines and mine-mouth coal-fired power plants [2]. As a result of the combustion air substitution, not only the ventilation air methane is mitigated but also the fuel purchase cost is reduced. It was shown in You's experimental results that VAM oxidation does not obviously affect the boiler operation when the methane concentration is less than 0.6% [16]. The process under development in our research group for the use of combustion air substitution with VAM in chemical looping combustion (CLC) provides an advanced technology platform for treatment of VAM and can be employed as part of any use and/or destruction mitigation strategies [17]. The motivation under this research is that the process possesses the advantages for both TFRR and CFRR systems. When the system is operating with temperatures higher than 1000 °C, the ventilation air methane will be oxidized mainly by the reaction mechanism of thermal oxidation. For the operating temperatures under 800 °C, the catalytic combustion will activate and take the initiative due to the presence of metal oxide (i.e. oxygen carriers). Under such circumstances, the ventilation air methane is able to be oxidized completely with a flexible operation regime. It is also no doubt that the produced heat can be recovered without any compromise.

The aim of this study is to examine the effect of the variations in VAM flow rate and methane concentration on the system performance. This was accomplished thermodynamically by simulating the process in a commercially available simulation software – ASPEN PLUS. Moreover, sets of experiment were carried out in a fixed bed setup to determine the conversion of VAM in air reactor with $\text{Fe}_2\text{O}_3/\text{Al}_2\text{O}_3$ as oxygen carrier over temperatures lower than 800 °C under which the thermal oxidation is not dominant. The experimental component of this study was carried out because the oxidation characteristics of VAM is an important factor in the overall performance of the proposed ancillary system. Although many of past studies have focused on the catalytic combustion of VAM in air, the prevailing used catalysts were supported noble metals or metal oxides like Pt, Pd or PdO due to their high reactivity even with a very low loading content (less than 1 wt%). Very few works was found on the use of Fe_2O_3 supported by Al_2O_3 as catalyst in the catalytic oxidation of VAM [12,18]. Five metal oxides with different Fe_2O_3 loading content are used to investigate the effect of Fe_2O_3 content on the methane conversion, made it possible

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