



A feasibility study on a novel stone dust looping process for abatement of ventilation air methane



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ABSTRACT

This paper describes the development of a novel stone dust looping process that relates to the removal of ventilation air methane using stone dust. The working principle behind the stone dust looping process is incredibly simple which involves the catalytic oxidation of methane followed by carbonation and calcination reactions. In the current work, laboratory scale fluidized bed experiments and process simulations were conducted to evaluate the feasibility of the stone dust looping process. The experimental work concluded that oxidation of ventilation air methane in the stone dust looping process can be successfully achieved at temperatures between 500 and 650 °C. The experimental results indicated that oxidation of methane was found to increase at higher temperatures while carbon dioxide capture efficiency showed a declining trend with increasing temperature. Furthermore, higher methane conversion and optimum (thermodynamic) carbon dioxide capture efficiency were observed for lower ventilation air methane flow rates and higher bed inventory. The concentration of methane in ventilation air methane and stone dust particle size did not have a significant effect on methane conversion or carbon dioxide capture. Also, comparison with synthetically prepared CuO and Fe₂O₃ catalysts has been made with CaO for VAM oxidation. CaO was found to be comparable to Fe₂O₃ and superior to CuO. From the process simulations, it was concluded that thermal energy generation in the carbonator was increased with higher methane and carbon dioxide concentrations. However, at the same time for higher methane and carbon dioxide concentrations, a greater CaO flux was required in the carbonator and hence a larger amount of goaf gas was required for the calcination reaction. The higher thermal energy generation in the carbonator was expected to improve the autothermicity of the stone dust looping process at concentrations of methane in the ventilation stream <0.2 vol.% (thermodynamic limit).

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1. Introduction

Methane formed during the coalification and biogenic process is retained within the coal seam and adjacent rock strata [1]. The methane removed from working mines via ventilation techniques is known as Ventilation Air Methane (VAM) [2]. VAM contains very dilute levels of methane; typical concentrations range between 0.2 and 1 vol.% methane, well below the Lower Explosive Limit (LEL) [3]. With few exceptions, VAM is released to the atmosphere without attempting capture, destruction or utilization [2]. Methane is considered as the second most abundant greenhouse gas emitted by human activities after carbon dioxide and has a much greater short-term global warming potential [1]. This suggests that increasing methane emissions has a significant impact on rising temperatures, particularly over the short-term, while reducing methane emissions brings large short-term benefits in slowing down the rate of climate change. Out of the total fugitive

methane emissions from coal mines, approximately 64% is accountable from the VAM stream [3].

With ongoing debate on the fate of future carbon taxes worldwide and the current version of the direct action plan in Australia, the carbon footprint of coal and its associated greenhouse gas emissions during mining and utilization (in the steel and power generation industries) has become an economic, as well as an environmental concern [1]. This is especially true for emissions of methane as VAM, due to its 25 times higher global warming potential, which has become a pending liability for coal mine operators [2]. The need to abate VAM and thus lower the greenhouse footprint is seen as a necessary mining activity. Hence, efforts have been made in recent times to explore potential systems that can either destruct VAM through a thermal oxidation process or utilize it for heat or power generation.

There exist numerous challenges with the destruction/utilization of VAM [4]. This is due to the fact that ventilation airflows are very large, and the methane concentration is so dilute that conventional combustion processes cannot oxidize it without additional auxiliary fuel [4]. Moreover, fluctuations in the VAM flow rate and methane concentrations, together with the changing location of the working face in relation

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Nomenclature

| | |
|------------------|----------------------------------|
| SDL | Stone dust looping |
| VAM | Ventilation air methane |
| TFRR | Thermal flow reversal reactor |
| CLAS | Chemical looping air separation |
| CLCR | Chemical looping carbon arrestor |
| GCL | Greenhouse calcium looping |
| CLOC | Chemical looping oxy-combustor |
| kW _{th} | Kilowatt thermal |

to ventilation shafts increase processing difficulty, and thus constitute technical barriers to the recovery and use of VAM [4]. VAM mitigation generally requires it to be treated either in its dilute state or increase its concentrations up to levels that can be used in conventional/lean gas engines/turbines [3]. In this regard several technological options have been explored worldwide [5]. The processes that destruct/utilize VAM in its dilute form primarily work on the principle of thermal oxidation [6]/catalytic oxidation [7]. Concentrators that use carbon composites [8] have also been considered to increase the concentrations of methane in the VAM stream and can be integrated with any conventional gas engine/turbine technology. VAM utilization technologies are generally divided into two categories: ancillary uses and principal uses [9]. For the ancillary uses, VAM is used as a substitute to ambient air in combustion processes, including fossil fuel based thermal power plants [10], gas turbines [9] and internal combustion engines [9]. While for principal use, the methane in VAM is considered as a primary fuel. The technologies, as listed in Table 1, fall under the principle use category. The ancillary use of VAM is expected to be very limited due to the fact that at coal mine sites the availability/requirement of a thermal power plant is very rare [10]. Also, the installation of gas turbines or internal combustion engines will be limited due to the high requirement of auxiliary fuel inventory at site [9]. Under this scenario, principle use applications and concentrators may be found to be more attractive. However, principle use technologies also suffer from numerous challenges. For example, the high temperature and high pressure operation of Thermal Reversal Flow Reactors (TFRR) and concentrators respectively present potential safety hazards [11]. Also, high moisture and dust concentrations in VAM may deactivate the catalysts or block the pores of thermal mass media used in reversal reactor configurations [6,10–11]. The specific challenges for principle use technologies are highlighted in Table 2.

Full scale installations of such VAM abatement technologies can't be realized in Australia or worldwide until the associated challenges, as described earlier, with current and emerging technologies are satisfactorily resolved. Given the aforementioned challenges, an entirely different process/approach is proposed in this work. This approach involves a novel process called "Stone dust looping (SDL)" being developed at the University of Newcastle, Australia. The SDL process is a form of chemical/calcium looping process being developed for cleaner power generation. The University of Newcastle is known for applying chemical/calcium looping principles to areas other than power

Table 1
List of Principle Use Technologies.

| Technology | Technology Provider |
|--|-----------------------------|
| Thermal flow reversal reactor (TFRR) | VOCSIDIZER, MEGTEC |
| Catalytic flow reversal reactor (CFRR) | CANMET |
| Catalytic monolith reactor (CMR) | CSIRO |
| Catalytic lean-burn gas turbine (CLBGT), Recuperative lean-burn gas turbine (RLBGT) | CSIRO, Ingersol-Rand EDL |
| Ventilated air methane regenerative after burner (VAM-RAB) | Corkys Pty Ltd |
| Ventilation air methane chemical oxidizer (VAMCO) | University of Newcastle |

Table 2
Specific Challenges with Principle Use VAM Abatement Technologies.

| Parameters | Challenges |
|---|--|
| Large volumetric flow rate, dilute concentrations of methane and fluctuations in both | Scale up, heat recovery, constant power generation |
| High moisture and dust present in the VAM stream | Blockage of heat exchanger tubes/pores, deactivation of expensive catalysts, hot spot generation, abrasion, corrosion Safety hazard |
| High temperature/High pressure operation of VAM abatement unit Requirement of large amount of auxiliary fuel | Reactor not autothermal at lower concentration of methane in VAM and Safety, cost and storage issues associated with auxiliary fuel |
| Integration of VAM and Goaf gas streams | Mixing of VAM And Goaf gas, higher footprint due to requirement separate destruction/utilization technologies for VAM and Goaf gas streams |

generation. A few examples are the Chemical Looping Air Separation (CLAS) process for oxygen production [12], Chemical Looping Carbon Arrestor (CLCR) process for methanol synthesis [13], Greenhouse Calcium Looping (GCL) process for providing heat and CO₂ to the greenhouses [14], Calcium Looping Gasification Process for hydrogen production [15] and Chemical Looping Oxy-Combustor (CLOC) process coal combustion [16]. The SDL process described in this work is expected to eliminate several shortcomings associated with the current and emerging VAM mitigation processes. If successful, it would provide a step change solution to mine sites in abating VAM emissions. The details of the SDL process are presented in the following section.

2. Stone dust looping process concept

The SDL process uses a stone dust rich in Calcium Carbonate (CaCO₃) as a CO₂ scavenger, which is routinely used at underground coal mine sites as a primary inert agent, in the prevention of coal dust explosions. The process is a form of carbonate looping which is expected to operate at lower cost due to its low/moderate operating temperature and low raw material cost. The process can be explained by looping reactions 1 and 2 in separate calciner and carbonator reactors (See Fig. 1). Reaction 1 is called the "calcination" reaction in which stone dust is decomposed using either natural gas or goaf gas by maintaining appropriate CO₂ partial pressure in "calciner" reactor. The Calcium Oxide (CaO) formed after stone dust decomposition then undergoes the "carbonation" reaction in the presence of VAM and converts back to calcium carbonate (i.e. stone dust) as shown in reaction 2 in "carbonator" reactor. Also, CO₂ present in the VAM stream, as an inert gas, will undergo the carbonation reaction. The stone dust formed by carbonation in the carbonator is then recycled to the calciner for CaO regeneration. The CO₂ captured in carbonator can then be released during calcination, in a concentrated form. And stored or used as a by-product. The stone dust particles rich in calcium are looped between the carbonator and calciner reactors and hence the origin of the process name, stone dust looping. Thermodynamically, the calcination reaction may occur at temperatures between 625 and 800 °C by maintaining appropriate CO₂ partial pressure in the carbonator. While the carbonation reaction is feasible at temperatures between 100 and 625 °C for 1 vol.% methane in VAM. However, due to slow reaction rates at lower temperatures, the carbonator temperature is suggested to be around 450–625 °C, whilst the calciner temperature is recommended to be around 700–800 °C. In the SDL process, the carbonator will be operated in an exothermal mode, whilst the calciner will always be in an autothermal condition.

The SDL process offers several advantages. First of all, the carbonator (where VAM oxidation will occur) can be operated at temperatures as

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