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A review on carbon dioxide capture and storage technology using coal fly ash

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HIGHLIGHTS

- The potential of the CCS technology using coal fly ash (FA) is reviewed.
- Alkali species in FA are dissolved and consumed to sequestrate CO_2 in the wet process.
- FA can be used as a support or as a raw material of dry sorbents to capture CO_2.
- The technology can stabilize the harmful components in FA during the process.
- Therefore, the technology may be another option of CCS to a limited extent.

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ABSTRACT

This work reviews the availability and the potential of the carbon capture and storage (CCS) technology using coal fly ash (FA). Because the technology can be effectively applied on-site to coal fired power plants and as FA contains sufficient alkali components, the technology may be another option of CCS technology to a limited extent.

The technology can be divided into wet and dry processes. In the former, the available components for CCS in FA are leached into solution by the solvent where they are subsequently consumed for carbonation to store CO_2. Particularly, the CO_2 storage capacity of CaO-enriched FA solution mixed with brine under high pressure may be equal to or greater than the true CO_2 emission reduction achieved by applying FA as a cement additive.

In the dry process, FA can be used as a direct support or as the raw material of the sorbent supports for CO_2 capture. The dry process is effectively applied for CO_2 capture rather than storage because the sorbents should be regenerated. Another advantage of the technology is the stabilization of the harmful components present in FA, which are mostly co-precipitated with carbonated FA during the process.

1. Introduction

The amount of solid waste residue (SWR) generated from the large-scale industrial processes such as coal fired power plant (CFPP), cement plant, steel, paper, oil shale industry and solid waste incinerator is increasing every year and some SWRs are substantially harmful to the environment. Therefore, the SWR disposal situation has been intensively aggravated to become an important issue [1–5]. Considering that most of the processes that generate SWR emit a great amount of CO_2, SWR can be directly or indirectly used as a material for on-site CO_2 capture and storage (CCS), which is another option of CCS technology [6–13].

This potential is based on the following reasons. Firstly, because industrial SWRs contain substantial alkali and alkali earth metals [14–17], the mineral carbonation with SWR is readily achieved to store CO_2 and which may be a permanent solution compared to geological and ocean storage. In addition, CO_2 can be partially recovered from the instable carbonated (or bi-carbonated) SWR products. Secondly, they are geochemically instable and reactive because they are generally formed at very high temperature (over 1200 °C) and subsequently produced by rapid cooling [7]. Finally, they are porous materials with relatively high surface area.

The first study on CCS using SWR was conducted by Reddy et al. group with alkaline byproduct of oil shale combustion process in the mid-1980s [18] and they proposed the technology to increase the reaction rate of former process in the early 1990s [19]. Both works had inspired a number of subsequent literatures dealt with mineral carbonation with various SWRs [8,20–22].

Among industrial SWRs, interest has focused on coal fly ash (FA) generated from the pulverized CFPP because the CO_2 emissions from CFPP and the FA portion are the largest [23–28]. Although

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the characteristics of FA differ substantially according to the coal and combustion conditions [29], currently generated FA is mostly used as cement and concrete additives [30,31]. However, its utilization ratio remains less than 30% of the total generated amount [32] in terms of the worldwide production, which is estimated at 700 million tons per year. In US, at least 150 million tons FA is generated annually and 27% is reused, while the remaining is landfilled or surface-impounded [30].

Although CO_2 emission reduction using FA can be achieved via some methods, the primary technology is based on mineral carbonation because FA contains a high content of alkali components, which are essential materials for mineral carbonation [33–35]. For example, the CaO and MgO contents are more than 20% in Class-C FA (FA generated from younger lignite or sub-bituminous coal burning). Furthermore, the CaO concentration is very high in FA collected at flue gas desulfurization (FGD) process [36]. Therefore, several attempts have been made to use FA as the caustic material to increase the pH of the acidic brine solution generated from oil and gas production.

One of the advantages of FA use for CO_2 emission reduction is its on-site application in CFPP. This is very effective to minimize the CCS cost while saving or reducing the current transfer, treatment and disposal costs of FA. In addition, the final carbonated FA products have the potential to be effectively reused as construction materials or additives because their physical structure might be changed, and their mechanical strength and leaching resistance are improved. Therefore, these can substantially enhance the economy of the carbonation process and decrease the environmental impact of FA.

Many papers have reported the FA carbonation effect by leaching process, irrespective of CCS. However, the research focusing on the application of FA to one of the CCS technologies began to be reported since the late 1990s. The present paper reviews the CCS technologies using FA based on previously reported studies. Firstly, the technology using FA is divided into two categories: wet and dry processes. Secondly, the features and performance of each technology are investigated in detail. Finally, the availability and CO_2 emission reduction potential of the technology are discussed.

2. Status of coal fly ash (FA) in Korea

2.1. Amount of FA generated

Total amount of coal ash, including FA, generated per year since 2001 in Korea is listed in Table 1.

The nation's total electricity production in 2010 was 433,604 GW h [37], of which 45%, 195,000 GW h, was generated from CFPP via 5 major electric utilities. For this electricity generation, 78.89 million tons of coal was consumed to generate 8.41 million tons of coal ash [38–40]. The amount of generated FA was estimated to be 5.29 million tons [41], of which more than 90% was utilized as additives in cement, concrete and brick.

2.2. Application as a cement additive

As aforementioned, most FA is currently used as a cement additive. When FA is mixed with the Portland cement, SiO_2 and Al_2O_3 in FA react with slaked lime and water according to the Pozzolan reaction. Therefore, calcium silicate and calcium aluminate hydrates are produced, which substantially increase the chemical resistance and the long-term strength of the cement [42]. Therefore, this Portland FA cement (PFAC) is generally used as the concrete in dam construction.

Another effect of FA application as a cement additive is to reduce CO_2 emission by reducing the cement consumption. In Korea Standards, PFAC is classified into three types according to the FA content: A (~10% of FA), B (10–20%) and C (20–30%). Currently, type B is most widely used. Therefore, the average portion of FA in PFAC is assumed to be 15%. When 1 ton of Portland cement is produced, 0.83 ton of CO_2 is known to be emitted, of which 0.46 ton is generated from sintering of $CaCO_3$ and the balance is due to energy consumption [43]. Therefore, assuming that the energy consumption is unchanged and considering the portion of FA substitution for $CaCO_3$, the CO_2 emission reduction achieved by using FA as a cement additive is roughly estimated to be 0.069 ton per ton of cement production.

However, the true CO_2 emission reduction by FA replacement with cement is less because of the additional CO_2 emissions generated from FA treatment as a cement additive such as fractionation, mixing and transportation processes. Due to these penalties, the CO_2 emission reduction contribution of FA as a cement additive may be substantially less than this value.

Therefore, when FA is used as a direct or indirect material for CCS in CFPP on-site, its detailed effects should be analyzed compared to FA application in cement plant. In this case, various conditions should be considered such as additional energy consumption to treat FA for wet or dry based application. The beneficial effects should be also considered which include reuse of carbonated FA products as construction materials and stabilization of harmful components in FA precipitated during carbonation. Although, this feasibility study based on all these conditions would very meaningful, the present review does not include this study. The paper focuses on the CCS capacity of FA or its potential when it is used as a material for CCS.

Except as a cement additive, FA application to other fields has been extensively investigated, including ceramic balls for water purification, zeolite synthesis, polish materials of steel plate, anti-flaming, building and fill materials [44]. However, these are minor applications in terms of CO_2 emission reduction.

3. Study on CCS technology using fly ash (FA)

Recently, many papers have investigated the development of CCS technology using FA. These studies can be divided into three classifications according to the applied technology. The first category is wet and dry processes. In wet technology [32,45–51] CCS components in FA such as Ca, Na, Mg and K are leached by aqueous (acidic) solution and then consumed on carbonation reaction to store CO_2. In the dry process [52–55], FA and some additives are used as direct CCS materials. In addition, FA is used to support the catalytic sorbent for CO_2 capture [56–58] and as a raw material of zeolite [59–63]. The second classification is based on the level of pressure during the carbonation. The reaction can be carried out in atmospheric or high pressurized condition. Generally, carbonation with high pressure is carried out in an autoclave with the object of CO_2 storage. Final classification depends on the number of processes involved. In a one-step process, FA treatment and carbon-

Table 1
Annual coal ash and coal fly ash (FA) generation in Korea.

Year	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
Coal ash (million tons)	4.92	5.14	5.19	5.38	5.95	5.84	6.02	7.61	8.35	8.41
Coal fly ash (million tons)	3.09	3.23	3.26	3.38	3.74	3.67	3.79	4.79	5.25	5.29