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Review article

Flue gas desulfurization effluents: An unexploited selenium resource

Patricia Cordoba[a], Lucian C. Staicu[b,*]

[a] Institute of Environmental Assessment and Water Research, Spanish National Research Council (IDÆA-CSIC), C/ Jordi Girona 18-26, 08034 Barcelona, Spain
[b] Faculty of Applied Chemistry and Materials Science, University Politehnica of Bucharest, Polizu Street 1-7, 011061 Bucharest, Romania

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ABSTRACT

With the advent of the Industrial Revolution, the natural cycle of selenium (Se) Se has been altered, thus leading to cases of severe environmental degradation. Nowadays, an important share of the global energy is generated by coal combustion. During this process, Se is concentrated in Flue Gas Desulfurization (FGD) wastewaters. FGD effluents can be treated by bioremediation, using bacterial metabolism to convert soluble and toxic forms of Se into elemental Se (nano)particles. Se and bacteria are intimately linked in a complex interplay since the element serves nutritional and metabolic energy generation functions, in addition to behaving as a powerful toxicant. In this article we explore the coupling of FGD treatment and resource recovery in the framework of circular economy. This approach reduces the burden of Se pollution for aquatic ecosystems, while also providing a way to recover a valuable resource, generating profit and offsetting the treatment costs.

1. Introduction

Industrial Revolution has interfered with the natural cycles of numerous chemical elements, including selenium (Se) [1]. A number of environmental incidents caused by Se pollution were documented in North America, the most severe ones leading to the virtual elimination of various ecological groups such as fish and aquatic birds [2]. The main toxicological mechanisms of selenium poisoning are related to sulfur substitution leading to dysfunctional biomolecules and to the production of reactive oxygen species such as hydroxyl radical ($^{\cdot}OH$), hydrogen peroxide (H_2O_2), and superoxide radical ($^{\cdot}O_2^{-}$) [3]. Selenium pollution is associated with industrial activities such as energy production based on fossil fuel combustion, metal and oil refining, and agricultural practices. Among them, coal-fired power generation is one of the major Se contributors to the environment.

Currently, coal is an important fuel used to generate energy worldwide [4]. The European Union (EU) generates a quarter of its power by burning coal (http://www.coalmap.eu/#/coal-fleet/30plus). In the EU, coal-fired power generation is unevenly distributed with countries using low amounts of coal (e.g. France), while others heavily relying on this fossil resource (e.g. Germany, ~40%, Poland, ~70%) (http://www.coalmap.eu/#/coal-fleet/30plus). Other big coal consumers for energy purposes are China (~62%), India (~57%), and South Africa (~70%) [4].

Coal-fired power plants are regarded as big polluters; pulverized coal combustion leads to the generation of coal combustion by-products, mostly ashes and gaseous pollutants. However, increased pressure exerted by the public, various environmental protection groups and politicians has led to a gradual reduction of pollution incidence associated with coal combustion. Modern coal-fired stations contain a number of pollution control devices aiming at reducing the pollution load. Once coal is burned in the boiler to produce energy used to generate superheated steam, an important component of the power station is devoted to cleaning up the flue gas [5]. The coarser fraction of ashes, bottom ash or slag, is removed from the bottom of the boiler, whereas the finest particles, fly ashes, are separated using filter bags or electrostatic precipitators (ESP). Gaseous pollutants such as NO_x and SO_2 are removed using Selective Catalytic Reduction (SCR) and Flue Gas Desulfurization (FGD) systems, respectively (Fig. 1). The most popular FGD system is wet FGD using a slurry of lime/limestone that is sprayed in the flue gas, resulting in the production of solid and aqueous by-products [6]: gypsum slurry (aqueous phase + gypsum sludge), FGD-gypsum, and filtered water. Gypsum slurry is the total fraction of the slurries (solid + water), while the term aqueous phase of gypsum is employed for the water fraction of the total slurry. Gypsum sludge is referred to the solid fraction of gypsum slurry, and FGD-gypsum is the solid end-product after filtration process. Filtered water is the resulting water from gypsum slurry filtration [6–8].

Depending on the characteristics and properties of the by-products and the availability and resources of the power plant, the fate of FGD-gypsum and filtered water can be the disposal in landfills and water treatment or recycling, respectively [6,9]. FGD effluents have a complex matrix containing anions (e.g. sulfate, selenate, carbonate, chloride), major cations (e.g. Ca, Mg), and a wide spectrum of metals

* Corresponding author.
E-mail address: lucian.staicu@upb.ro (L.C. Staicu).

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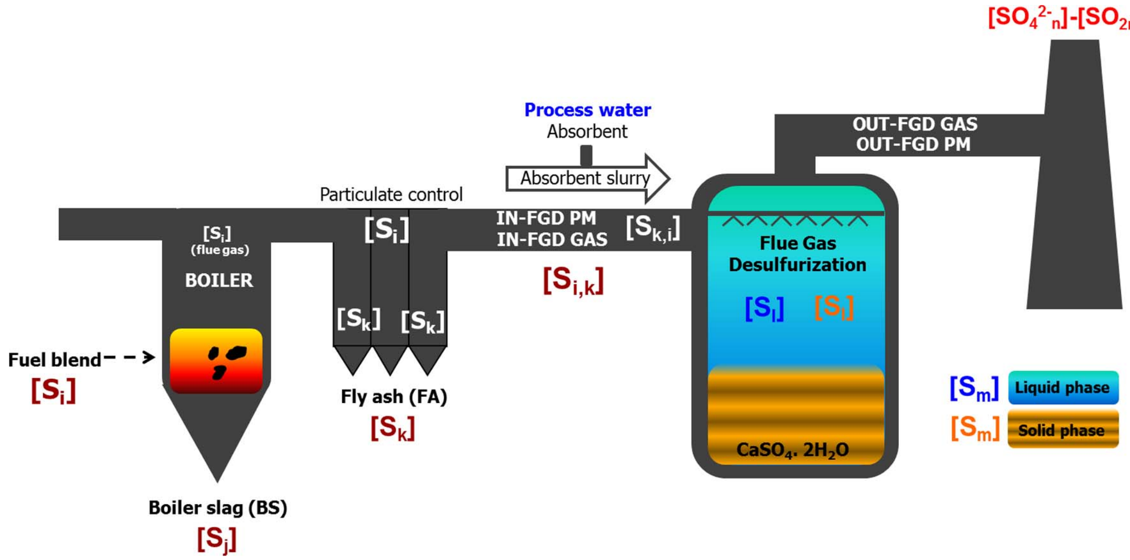

Fig. 1. Diagram of a coal-fired power plant equipped with a flue gas desulfurization (FGD) system. Legend: S_i to S_m refers to the partitioning of a given element in the *in* and *outgoing* streams during PCC-FGD process.

(e.g. Cd, Cr, Hg, Ni, V, Zn) [10–12]. It is worth mentioning that Se displays several valence states (+VI, +IV, 0, −II) with contrasting water solubility and toxicology profiles. FGD effluents contain Se mainly (> 90%) in its most oxidized state (selenate, +VI, SeO_4^{2-}), which has high water solubility and is bioavailable [12,13]. FGD effluents are currently a big challenge for power industry because of the following characteristics: i) are generated in large volumes, ii) have a complex matrix, and iii) the discharge limits for Se are low and difficult to achieve using most treatment systems. For a detailed presentation of the main Se treatment strategies, the reader is referred to Tan et al. [10] and Staicu et al. [10–14].

An interesting treatment approach using microbial inocula (bioremediation) is gaining in popularity worldwide. A recent development of this approach seeks to couple wastewater treatment with resource recovery [15]. Bioremediation of Se-laden effluents is founded on the microbial conversion of Se oxyanions, selenate and selenite (Se, +IV, SeO_3^{2-}), into solid elemental Se, Se^0, which is less toxic owing to its limited bioavailability. Currently, Se^0 resulted from bioremediation is not considered as a potential resource, although it has high recovery potential and Se is a critical element with multiple industrial and domestic applications.

In this review article, we explore the production of FGD effluents containing high levels of Se and link the generation of these effluents with bioremediation in an attempt to achieve both waste treatment and resource recovery. Firstly, the current situation of coal combustion for energy production and the generation of FGD effluents are presented and discussed. Secondly, the fundamentals of the microbial conversion of Se and the biotechnological applications including bioremediation are reviewed. Finally, the last part unifies the FGD generation with the biological treatment, coupled with Se^0 recovery in the framework of the circular economy strategy.

2. Power generation from coal

2.1. Status of coal power generation

Coal plays an essential role in our global energy scheme for power generation. However, coal is currently a target to comply with the Paris climate agreement for both countries and companies. In 2016, world coal production fell by 6.2% (231 million tonnes of oil equivalent (mtoe)), the largest decline on record. US production fell by 19% or 85 mtoe, while China's production fell by 7.9% or 140 mtoe [4].

In 2016, coal's share of global primary energy consumption also fell to 28%, the lowest share since 2004 [4]. The largest declines in coal consumption were seen in the US (−8.8% fall). Chinese coal consumption also declined (−1.6%) for the third consecutive year, which led China to resuming its position as the world's largest importer of coal [4]. In the UK, global coal prices were amplified by the increase in the UK's Carbon Price Floor in 2015, which resulted in the UK's last three underground coal mines closing, consumption falling back to where it was roughly 200 years ago around the time of the Industrial Revolution [4].

However, coal is the world's most abundant energy resource. There are 1,139,331 million tonnes of proven coal reserves worldwide, sufficient to meet 153 years of global production. Total proven coal reserves are shown for anthracite and bituminous (including brown coal) and sub-bituminous and lignite. In comparison, proven oil and natural gas reserves are equivalent to around 50 and 53 years, respectively, at 2016 production levels [16]. Therefore, despite the decline in coal production and consumption, coal is and will be a reliable source for power generation.

2.2. Coal combustion

Pulverized coal combustion (PCC) is the most widely used technology for coal power generation. In this process, coal is milled, pulverized, and injected in the boiler with air to allow combustion. Combustion takes place at temperatures from 1300 to 1700 °C, depending largely on coal rank [17]. The heat generated is used to produce a steam of high pressure (25–30 bars) and temperature to power high and medium turbines that are connected to a generator which produces electric energy (Fig. 1).

Pulverized coal combustion leads to the generation of coal combustion by-products, mostly ashes. The coarser fraction of ashes, bottom ash or slag, is removed from the bottom of the boiler, whereas the finest particles, or fly ashes, are retained from the flue gas stream by the particulate controls, usually Electrostatic Precipitators (ESP) or Fabric Filters (FF). Despite the high efficiency of the ESPs and FFs (> 99%) [18,19], a small fraction of fly ashes escapes from the controls and reaches the FGD system (Fig. 1).

The conventional process of coal combustion can be modified to improve the energetic efficiency of the process and reduce the environmental impacts. The *Fluidized Bed Combustion* (FBC) allows a better use of the fuel and a better transference of the heat for power

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