INTRODUCTION

Gypsum deposits were formed during the Paleozoic Era, approximately 600 million years ago, when most of the Earth’s surface was covered by salt water. Gypsum is composed of 79.1% calcium sulfate and 20.9% water (by weight). It is a nonmetallic mineral, one of the most abundant in the world, and is found in rock form. Its chemical formula is CaSO4·2H2O, and pure gypsum is white in color. However, gypsum typically contains impurities,
such as clay and other minerals or several soluble salts, that gives it other colors. The impurities contained within the gypsum lead to changes in its physical and chemical properties. Due to these characteristics, the use of naturally mined raw gypsum material in applications typically involves multiple processing procedures and steps. However, synthetic gypsum is highly similar to naturally mined gypsum; hence, specific types of synthetic gypsum have replaced natural gypsum in several industries such as cement plants, building material production, mining industry, and agriculture.

One industrial process that produces synthetic gypsum is flue-gas desulfurization (FGD), which is used to remove gaseous sulfur dioxide from boiler exhaust gas derived from coal-fired power plants, steel plants, and other heating plants. This is important because the reaction of sulfur dioxide with moisture generates sulfuric acid, which causes acid rain and poses a severe threat to the environment. Therefore, an increasing number of coal-fired plants use FGD to remove gaseous sulfur dioxide from their boiler exhaust gas as a method to curb sulfur dioxide emissions.

Depending on the specific chemical reactions that occur and the flow conditions employed, the FGD process can be subdivided into the following three categories: wet, semidry, and dry. Among these, the wet process is the most widely used. Moreover, more than 95% of the sulfur in the combustion gases from coal-fired plants are wetted by limestone slurry in this FGD process, showing that it has an extremely high desulfurization efficiency. The chemical reaction between the limestone slurry and sulfur in the flue gas produces calcium sulfate, while the limestone slurry is converted into a gypsum slurry by the desulfurization process. The latter is then dehydrated to produce FGD gypsum (as shown in the following equations). Thus, the synthetic FGD gypsum can be used to replace natural gypsum in several industries.

**SO₂ in flue gas:**

\[
\text{SO}_2(g) \leftrightarrow \text{SO}_2(aq) \text{ [dissolution of SO₂]} \\
\text{SO}_2(aq) + \text{H}_2\text{O}(aq) \leftrightarrow \text{H}_2\text{SO}_3(aq) \text{ [hydrolysis of SO₂]} \\
\text{H}_2\text{SO}_3(aq) \rightarrow \text{H}^+(aq) + \text{HSO}_3^-(aq) \\
\text{HSO}_3^-(aq) \rightarrow \text{H}^+(aq) + \text{SO}_3^{2-}(aq) \text{ [acid dissociation]} \\
\]

**Dissolution of limestone:**

\[
\text{CaCO}_3(s) + \text{H}^+(aq) \leftrightarrow \text{Ca}^{2+}(aq) + \text{HCO}_3^-(aq) \text{ [dissolution of limestone]} \\
\text{HCO}_3^-(aq) + \text{H}^+(aq) \rightarrow \text{CO}_2(aq) + \text{H}_2\text{O(aq)} \text{ [neutralization]}. \\
\]

**An overall chemical reaction of the above processes, which occurred in the FGD system is given below:**

\[
\text{SO}_2(g) + \text{CaCO}_3(s) + 2\text{H}_2\text{O}(aq) = \text{CaSO}_3 \cdot 2\text{H}_2\text{O(aq)} + \text{CO}_2(g) \\
\]

The produced calcium sulfite (CaSO₃·2H₂O) is then oxidized, resulting in FGD gypsum:

\[
\text{CaSO}_3 \cdot 2\text{H}_2\text{O(aq)} + 1/2\text{O}_2(g) \rightarrow \text{CaSO}_4 \cdot 2\text{H}_2\text{O(s)} \\
\]

The yield and quality of the FGD gypsum depend on parameters such as operating mode, type of fuel/sulfur content, and the type of absorbent desulfurizer technology used. Since the late 1970s, European coal power plants have produced over 100 million tons of FGD gypsum. In 2003, 17 European countries produced 15.2 million tons of FGD gypsum. Production of large volumes of FGD gypsum makes it necessary to fully utilize the large volume of coal combustion products (CCPs) via several different treatment processes. The use of CCPs generated by the construction industry in the European Union has, in general, increased over the past decade; FGD gypsum reached 71% in 2004. More than 100 million tons of FGD plaster has been used in the European gypsum industry for more than 30 years, which has helped increase industrial use. FGD gypsum is currently considered a suitable raw material for several products based on gypsum. In addition, the production of FGD gypsum in China increased abruptly from 2005 onwards, reaching 52 million tons per year in 2012. However, the total recycling rate was only approximately 56%. A large amount of FGD gypsum (70-80 million tons/year) was produced in China recently. The United States produced more than 32 million tons of FGD gypsum in 2017. This indicates that different countries produce large quantities of FGD gypsum every year.

The utilization of CCPs is promoted worldwide by environmental standards and the liberalization of the global electricity market; thus, an increase in the FGD gypsum production is expected. However, there is currently a significant amount of unutilized FGD gypsum; considering that unused or untreated FGD gypsum requires a large area of land, this approach leads to the waste of valuable land resources and the potential threat of secondary environmental pollution to the environment. Therefore, new and innovative strategies are needed to reduce the negative effects of FGD gypsum on the environment.

In addition, FGD gypsum is the most recycled FGD material: 70% of the material is reutilized. Most is used to produce gypsum wallboard products (15.9 million tons), while the development of the FGD gypsum market has also received attention. The latter is due mainly to usage...
in material synthesis, and applications, and water treatment processes.

For material synthesis, the FGD gypsum has mainly been utilized in wallboard production, and concrete/cement and asphalt production, among others. Recent research has focused on producing gypsum boards with higher fire resistance, water resistance, and low density. Existing results show that the thermal insulation capacity of commercial gypsum board is lower than that of 100% FGD gypsum board. Additionally, 100% FGD gypsum board has a higher insulating capacity compared to gypsum board manufactured with different proportions of fly ash and FGD gypsum due to the high free water content in FGD gypsum. In addition, natural gypsum can be used in concrete production to replace the calcium aluminate cement (CAC), and cement produced from natural and synthetic gypsum shows similar compressive strengths. This shows that FGD gypsum can replace natural gypsum in concrete production. However, there is found a 1-hour delay in the initial setting time of concrete containing FGD gypsum compared to that containing natural gypsum. However, several studies tested different FGD gypsum/CAC ratios and concluded that the setting time can be shortened by the addition of FGD gypsum, which is favorable in construction.

It is worth noting that prior to using FGD gypsum in cement, it needs to be dehydrated and crystallized and the cement properties can be directly affected by the heat treatment in this dehydration process. These studies concluded that (a) concrete setting time decreases as the heat treatment temperature of FGD gypsum increases and (b) cement produced with FGD gypsum calcined at 200°C and showed a maximum strength at 3.5% SO3 content.

For land applications, FGD gypsum has mainly been utilized in soil amendment, fertilizer, reduction of soil erosion and eutrophication, mine reclamation, and remediation of heavy metal-contaminated soil. The use of FGD gypsum as a soil amendment is well known and has been investigated thoroughly over the past few decades. Recently, studies have mainly focused on amending sodic soils, specifically in coastal areas. Even though the Na+ content of coastal soil is relatively high compared to inland agricultural soil, several studies have shown that FGD gypsum can be successfully used to reclaim coastal plains. In addition, using FGD gypsum as a fertilizer can provide Ca and S for plant growth. Mg, K, and Se are available in FGD gypsum in large amounts, and excess amounts of Se can be toxic. This shows that reducing the harmful elements in FGD gypsum is an important problem that needs to be solved when using FGD gypsum as fertilizer. The effect of FGD gypsum on crop growth and yield has been studied in peanuts, tomatoes, wheat, cantaloupes, corn, alfalfa, hay, and soybeans. According to the results, as the amount of FGD gypsum applied increased, both corn and hay yields increased (20 Mg/ha). Several studies reported that the FGD gypsum can help to prevent eutrophication. The elevated Ca2+ content in FGD gypsum-amended soil facilitates the flocculation of clay particles, and this increases the porosity of the soil. The higher porosity soil allows for increased water infiltration and reduces surface runoff and sediment transportation, thus preventing eutrophication. In addition, even though FGD gypsum is not as alkaline as fly ash, it has been utilized for mine soil reclamation in several studies.

For water treatment processes, the FGD gypsum has mainly been utilized for the reduction of dissolved organic carbon and metal removal, among others. Some studies have revealed that the application of FGD gypsum can effectively reduce the dissolved organic carbon from surface water, which is used as a drinking water source in Australia. The findings showed that Ca2+ was able to flocculate the natural organic matter present in the water by cross-linking carboxylic and other functional groups within the organic matter and then with clay particles in the sediment. The same study compared the use of FGD gypsum with alum treatment in conventional water treatment processes and reported similar efficacies for both treatments. This led to the use of FGD gypsum in drinking water treatment processes with the potential of being an alternate replacement for alum, especially for reducing the dissolved organic carbon in water. In addition, the use of FGD gypsum to remove metals in the aqueous phase was reported recently. FGD gypsum was used to remove metals from the leachates from coal ash dump sites, resembling a sustainable waste management practice. The generation of $S^{2-}$ from FGD gypsum, with the aid of sulfur-reducing bacteria and organic matter under anaerobic conditions, of which equations is as follows:

$$\text{Organic matter (C, H, O) + SO}_4^{2-} \xrightarrow{\text{sulfur reducing bacteria}} \text{HS}^- + \text{HCO}_3^-$$

(1)

The generated HS$^-$ then reacts with metals in the leachate to form stable sulfidic metal precipitates.

$$\text{Me}^{2+} + \text{HS}^- \rightarrow \text{MeS}_{(s)} + \text{H}^+$$

(2)

Using FGD gypsum to remove Pb and Cd from solutions, Pb removal by FGD gypsum is caused by the formation of PbSO$_4$, whereas Cd removal was attributed to ion exchange: The Cd$^{2+}$ in water is exchanged with Ca$^{2+}$ in the FGD gypsum. In this study, the underlying mechanism of the pH effect (from pH 2 to 7) on Cd and Pb sorption was not described in detail. The Pb removal by FGD gypsum can be correlated to the following arguments, based on the stability of Pb precipitates in Pb-FGD gypsum aqueous systems at different pH:

- pH 2.4—PbSO$_4_{(s)}$
- pH 5.7—PbSO$_4$(CO$_3$)$_2$(OH)$_2_{(s)}$
- pH 6.8—PbCO$_3_{(s)}$
However, both natural gypsum and FGD gypsum have several differences that can either limit or enhance the utilization of the latter.\(^4\)\(^4\)\(^9\)\(^5\)\(^0\) For example, the moisture content of FGD gypsum is higher than that of natural gypsum. FGD gypsum also has a fine grain size, which can affect its processing and handling in existing production facilities meant for natural gypsum. On the other hand, this quality means that it requires less grinding than natural gypsum. Most new plants that produce wallboard products are designed to accommodate FGD gypsum, either solely or in combination with natural gypsum.\(^4\)\(^5\)\(^1\) Several issues, such as the surface crystallization induced in FGD gypsum by chlorides, ash, iron, and manganese compounds, may affect the adherence of and color variation in the final product, rendering it undesirable for several products and applications.\(^5\)\(^2\) Chlorides can also cause flue corrosion and air pollution, increase wastewater treatment costs in power plants, and have a negative impact on the synthetic gypsum it is used to treat.\(^5\)\(^3\)-\(^5\)\(^7\)

Currently, 76.16%-80.19% of chlorides in the flue gas go into wastewater, while 11.35%-12.76% is accounted for in FGD gypsum.\(^5\)\(^8\)\(^5\)\(^9\) The influx of large quantities of chloride ions into wastewater increases the cost of treating the latter. However, as noted above, wetting FGD has a large effect on the removal of total chloride; its efficiency ranged from 91.2% to 96.1%. To solve the problem of excessive chlorine content in FGD gypsum, process water is typically sprayed on it in power plants. This method yields high chloride removal efficiencies; however, it may contribute further to the production of large amounts of wastewater. This increases the cost of subsequent wastewater treatment and increases the possibility of environmental pollution. In addition, although the water of crystallization content in gypsum varies with the conditions of its immediate environment, such as temperature or humidity,\(^6\)\(^0\) the chemical composition of gypsum does not change after heat treatment (Equations 3 and 4).\(^6\)\(^1\) However, FGD gypsum cannot be used to manufacture plaster without heat treatment.\(^5\)

\[
\text{CaSO}_4 \cdot 2\text{H}_2\text{O} \xrightarrow{105 \text{ to } 200^\circ \text{C}} \text{CaSO}_4 \cdot 0.5\text{H}_2\text{O} + 1.5\text{H}_2\text{O}
\] (3)

\[
\text{CaSO}_4 \cdot 0.5\text{H}_2\text{O} + 1.5\text{H}_2\text{O} \rightarrow \text{CaSO}_4 \cdot 2\text{H}_2\text{O} + \text{Heat}
\] (4)

The steam generated after power generation is typically used in steam cycles\(^6\)\(^2\)-\(^6\)\(^5\) or to generate thermal energy.\(^6\)\(^6\)-\(^6\)\(^8\) However, utilizing this steam in this manner is not efficient.\(^6\)\(^9\)-\(^7\)\(^2\) Thus, the use of the steam heat and moisture was considered as part of the treatment of FGD gypsum to reduce its chlorine content.

In this study, saturated steam was used to spray the FGD gypsum in a novel dechlorination method, referred to as the saturated steam spraying method. The FGD gypsum can, after dechlorination, replace natural gypsum in several industrial production processes, decreasing the consumption of natural gypsum minerals and the damage caused to the environment by mining. In addition, it promotes the development of new industries and markets for FGD gypsum products.

However, despite favorable results, research on the dechlorination of FGD gypsum and its wastewater reduction via wastewater treatment, and the utilization of FGD gypsum has been limited.\(^7\)\(^3\)-\(^7\)\(^5\) Therefore, this study combined the results of laboratory experiments conducted in previous studies\(^7\)\(^6\),\(^7\)\(^7\) and developed an emission reduction process for FGD gypsum dechlorination and “zero emissions” wastewater treatment.

The objectives of this study were to (a) develop a technology for FGD gypsum dechlorination and “zero emissions” wastewater treatment, (b) to study the availability of wastewater evaporation products, and (c) to explore feasibility of the craft enlargement.

2 | METHODS

The FGD gypsum samples used in this study were all produced by the Zouxian Power Station (Zoucheng Huadian International Power Co., Ltd.). The samples were not dechlorinated prior to transport from the factory and were wrapped in plastic packaging during transport.

Researchers have proposed several treatment plans for wastewater that results from desulfurization.\(^7\)\(^8\)-\(^8\)\(^1\) Of these, thermal evaporation is a simple process that requires little equipment and a relatively small space. This method involves evaporating spray in the fuel duct between the air preheater and dust catcher by atomizing the wastewater. The evaporated water in the fuel pipeline and the precipitator trap retain any solid particles, achieving zero discharge of waste liquid.\(^8\)\(^2\) Flue duct spray evaporation tests were carried out to reduce the emissions from wastewater generated during the dechlorination process. This was done via laboratory tests on FGD gypsum that included measuring the loss of inorganic chloride via heat treatment as Figure 1 shown.

Tests also considered tap water spray dechlorination, saturated steam spray dechlorination, and flue duct spray evaporation for dechlorination wastewater. As shown in Figure 2, the saturated steam used in the emission reduction process is wet saturated steam (conventional atmospheric pressure, 100°C), which readily condenses into a liquid mist upon cooling.

Of these, the inorganic chloride heat loss test determines the order of the influence of temperature on the chloride
based on the mass loss of chloride at different temperatures. The tap water spray dechlorination test evaluated the dechlorination efficiency and wastewater volume of the conventional dechlorination process by spraying FGD gypsum with tap water (flow: 12 kg/min, spray frequency: 3 s/time, spray times: eight times). These parameters were set to simulate site conditions. The saturated steam spray dechlorination test evaluated the dechlorination efficiency and wastewater volume of the emission reduction process (the wet saturated steam was produced by a Pentium PW518 handheld supercharged steam engine with a total power of 1300 W, steam flow rate of 20 g/min, and spray time of 10 minutes). The flue duct spray evaporation for dechlorination wastewater test evaluated the composition and range of evaporation product particle sizes by evaporating the dechlorination wastewater from the other two processes.

3 | RESULTS AND DISCUSSION

3.1 | Effect of different temperatures on chlorides in FGD gypsum

The heat loss test results of the FGD gypsum inorganic chlorides are shown in Figure 3. For temperatures ranging from 30 to 90°C, none of the chlorides were decomposed in large amounts. According to test results, KCl and CaCl₂ were more influenced by temperature than other chlorides, which resulted in a loss of quality. For temperatures ranging from 95 to 120°C, each chloride exhibited a certain mass loss; the mass losses of MgCl₂ and NaCl were the smallest. This indicates that most of the chlorides in FGD gypsum are difficult to decompose in this temperature range. To increase the degree of chloride decomposition, it is necessary to increase the temperature. Moreover, the quality of each chloride exhibited
a different degree of loss at temperatures ranging from 125
to 180°C.

The results presented above reveal that the temperature has an effect on the mass loss of chloride in FGD gypsum. KCl and CaCl$_2$ were preferentially decomposed under the influence of temperature, and their mass losses were large; MgCl$_2$ and NaCl were the most difficult to decompose. This phenomenon is related to the metallicity of chlorides; as the metallicity increases, the thermal stability increases. Therefore, the effect of the temperature field on the dechlorination efficiency of FGD gypsum should be further investigated.

3.2 Dechlorination process of FGD gypsum

The average chloride ion content in coal ranges from 210
to 274 mg/L. The processes of pyrolysis, combustion, and vaporization in the combustion zone of the boiler cause the chloride ions to be released into the flue gas mainly in the form of HCl. Thereafter, the desulfurization process can remove a portion of the trace elements from the flue gas, chloride enters the FGD gypsum through this process. Furthermore, Chang et al. found that the average chloride ion content in FGD gypsum can be as high as 1041.69 mg/L and that chloride in FGD gypsum exists mainly in inorganic forms such as CaCl$_2$, MgCl$_2$, KCl, and NaCl. However, the enriched chlorine not only has a negative influence on the dechlorination efficiency and wastewater output, but could also affect its availability in and effects on environmental pollution.

According to the ACAA (American Coal Ash Association), chloride content in the desulfurized gypsum should be reduced to below 400 ppm. However, general specifications for FGD gypsum used in a given industry may vary with respect to the manufacturer and product. Until now, most utilization of gypsum waste has involved a pre-heating process. This implies that the use of FGD gypsum can result in high energy consumption, which reduces the environmental benefits of the recycling process. In addition, given that the chemical composition of gypsum does not change after heat treatment, temperature has an influence on the mass that the chlorides in FGD gypsum lose. Waste steam heat remaining after steam power generation, in addition to two experiments (the tap water spray dechlorination test and saturated steam spray dechlorination test), were used to compare the effect of saturated steam and tap water on dechlorination efficiency and wastewater volume.

As shown in Figure 4, there are many differences between the tap water spray dechlorination test and the saturated steam spray dechlorination test (saturated steam: conventional atmospheric pressure, 100°C). In this study, laboratory tests were carried out to simulate the working conditions of two dechlorination processes, and the key factors affecting dechlorination efficiency and wastewater output in the emission reduction process were evaluated.

The results of the two tests are shown in Figures 5 and 6. The average dechlorination rate of the conventional dechlorination process (tap water spray dechlorination) was 82.5%, while the average dechlorination rate of the emission reduction process (saturated steam spray dechlorination) was 80.4%. Although the former was relatively high, it could easily generate a large amount of wastewater effluent (its average wastewater volume was 4.51 L). Furthermore, the average dechlorination rate of the emission reduction process reached 97.5% of the conventional dechlorination process, and the volume of wastewater generated by the entire dechlorination process was small (0.22 L, which is 4.9% of the volume of wastewater generated by the conventional dechlorination process). Therefore, the emission reduction process has a high dechlorination efficiency and a low wastewater output.

To determine the relationship between chlorides, we studied the changes in the chloride elements contained in the FGD gypsum before and after the dechlorination process. The elemental content of FGD gypsum before and after desulfurization by a saturated steam spray is shown in Figure 7. Given that FGD gypsum is largely influenced by high temperatures and atomized droplets in the dechlorination process, the decomposition of chlorides and the migration and specific gravity of chloride ions were reduced.

The decrease in the specific gravity of F, Na, Mg, Al, Si, and Fe, among other elements, indicates that the emission reduction process is suitable for the removal of soluble chlorides such as MgCl$_2$, KCl, and NaCl. In this process, the wet saturated steam provided the temperature at which the chloride decomposed and the moisture was migrated by the chloride ions. These factors have improved the dechlorination efficiency of the emission reduction process and reduced its water consumption.

According to the above results, the chlorides in the FGD gypsum have begun to decompose to different extents, due to the heat of saturated steam, at temperatures near 100°C. The spraying effect of saturated steam not only provides the temperature at which the chlorides decompose but also provides moisture to migrate chlorine, as shown in Figure 8.

The FGD gypsum particles were not larger than 71.31 μm in size, as shown in Figure 10. Thus, they were easily infiltrated and surrounded by the liquid mist. When FGD gypsum is sprayed with saturated steam, a small portion of soluble chlorides are decomposed by the heat and chloride ions migrate into the vapor molecule. The steam forms droplets, which accumulate and continuously flush the gaps of the FGD gypsum particles when it cools. The soluble chloride is then gradually transferred to the accumulated droplets. Finally, these accumulate to form a small amount of dechlorination wastewater, thus decreasing the chloride ion content in.
the FGD gypsum. This residual dechlorination wastewater is then further reduced and removed by subsequent wastewater evaporation processes.

Given that the chlorides in FGD gypsum are mainly derived from flue gas, the use of flue waste heat treatment achieves “zero emissions” for dechlorination wastewater and reduces the amount of chloride ions present in FGD gypsum. This reduction in turn reduces the chlorine content and output of dechlorination wastewater. Therefore, the entire process is continuously cycled to reduce the chloride ion content generated by the process.

Because the tap water used in the conventional dechlorination process contains a small amount of chloride ions, it is necessary to increase the consumption of tap water to achieve the same dechlorination efficiency as the emission reduction process. This leads to a low dechlorination efficiency and large volume of wastewater emitted by the conventional dechlorination process, which results in environmental pollution and a higher processing cost.

The saturated steam spray process meets the dechlorination requirements for FGD gypsum, significantly reduces the wastewater output of the dechlorination process, and provides the heat treatment necessary before its use. Furthermore, it achieves a clean utilization of mineral resources and the “zero emissions” objective of power plants; this facilitates treatment as well as comprehensive development and utilization of FGD gypsum.

### 3.3 Flue duct spray evaporation of the wastewater from dechlorination process

The treatment of FGD wastewater can fully reduce the impact of coal-fired power plants on the water quality, and it provides secondary benefits associated with wastewater reuse. However, current research is focused on the treatment of wastewater generated by the desulfurization process. Furthermore, there is limited research on the dechlorination wastewater produced by the dechlorination process that FGD gypsum is subjected to prior to its removal from the power plant.

Based on the results presented, the thief and conventional FGD gypsum dechlorination processes can affect the output of dechlorination wastewater that increases the cost of the dechlorination process and results in environmental pollution. Moreover, the conventional dechlorination process uses
a series of wastewater treatment methods (chemical precipitation, 78 fluidized bed, 79 membrane separation, and evaporative crystallization 80) to purify the wastewater. However, the wastewater remaining after these treatments that meets emission standards still contains high concentrations of dissolved solids such as chloride. 89 Discharging this wastewater directly into the environment is a waste of water resources and can also easily cause severe secondary pollution. Furthermore, spray evaporation in the fuel duct can reduce the volume of wastewater. This process mixes wastewater and pressurized compressed air, which is then sprayed into the flue gas duct between the air preheater and the electrostatic precipitator or separated evaporation tower to evaporate the wastewater instantaneously using the heat emitted by flue gas. 52 This process can reduce water consumption and energy consumption in flue-gas desulfurization, in addition to the traditional disposal costs associated with wastewater treatment. Hence, this method was used to treat the dechlorination wastewater in the emission reduction process to achieve the “zero emissions” objective.

As shown in Figure 9, compared with the conventional wastewater treatment method, the flue duct spray evaporation test requires the use of the flue waste heat to evaporate the wastewater moisture, and the soluble solids in the wastewater are continuously concentrated and precipitated. If the evaporation products are collected, the precipitated evaporation product is adsorbed to the acidic substance with the atomized wastewater droplets and then discharged into the fly ash through the electrostatic precipitator.

As shown in Figure 10, the particle size of the evaporated product of the dechlorination wastewater exhibited a bimodal distribution due to the evaporation of wastewater droplets in the flue evaporation process, which promoted particle agglomeration. 95,96 Hence, the particle size of the evaporated product was larger than that of the fly ash particles. Moreover, larger sized particles facilitate efficient collection by electrostatic precipitators. 97

Figure 10 demonstrates that the evaporation product has a larger particle size than the FGD gypsum, which is caused by the concentration and precipitation of soluble substances and particle agglomeration in the wastewater evaporation process. Under the influence of a high concentration of fly

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**FIGURE 7** Element content changes before and after emission reduction process of flue-gas desulfurization gypsum

**FIGURE 8** Chloride ion migration process

**FIGURE 9** Dechlorination wastewater flue evaporation process
ash in the flue gas, the droplets of wastewater collide and adsorb fly ash during the evaporation process, which results in agglomerates of wastewater evaporation products and fly ash, as shown in Figures 7 and 9. Under the condition of flue gas with high concentrations of fly ash, the atomized droplets of chlorine wastewater cover the surface of the fly ash particles, thus increasing the effective evaporation areas of the droplets. This promotes the agglomeration of the wastewater droplets with the fly ash particles into a larger sized particle agglomerate, and this affects the wastewater evaporation rate. However, without the influence of fly ash, the particles of dechlorination wastewater are mainly dispersed. This is because, under these conditions, the atomized droplets of dechlorination wastewater exhibit good dispersibility and rapid evaporation. In addition, the agglomeration of evaporation products is not widespread and exhibits high dispersion.

3.4 Availability of dechlorinated FGD gypsum and wastewater evaporation products

As shown in Figure 11, the components of the wastewater evaporation products were found to be CaSO₄·0.5H₂O, CaSO₄·2H₂O, Fe(OH)₃, and NaCl. This further indicates that chloride ions and sulfur ions, among others, migrate into dechlorination wastewater during the dechlorination process. As the wastewater spray into the flue duct evaporates, the chloride element content in the fly ash can gradually increase if the evaporation products are mixed into the fly ash.

For the burning of different types of coal, the increase in the chloride ions content in fly ash ranged from 0.3% to 0.5%. In addition, chloride ions and sulfur ions can act as a stimulant and an early strength agent in fly ash, which indicates that mixing a certain proportion of wastewater products that contain chloride and sulfur ions into fly ash improves the activity and strength of the mineral filling material.

At present, fly ash from power plants is mainly used as a cement admixture and mining fill material. According to “Fly Ash for Cement and Concrete (GB/T 1596-2017)”, there is no recommended content of chlorides that can be mixed into fly ash; however, the chloride content was not greater than 0.06% in “General Purpose Portland Cement (GB175-2007)”. If the evaporated products are mixed into fly ash, the proportion of fly ash in the cement is limited. Thus, a dust collecting device can be used to collect the wastewater evaporation products separately before the electrostatic precipitator stage as shown in Figure 10.

Due to the purity, abundance, and physical and chemical properties of FGD gypsum being similar to those of natural gypsum, FGD gypsum is being used in various applications. In addition, 54% of the total FGD gypsum was used in drywall production and 8% in cement/concrete/asphalt production. Further, 17% of the total FGD gypsum production was used in terrestrial applications such as agriculture, mine reclamation, and structural filling. Despite
these useful treatments, approximately 43% of the total FGD gypsum was still disposed of in landfills. However, since burying FGD gypsum can result in a large amount of land being occupied, it is critical for us to find novel reuse applications of FGD gypsum and its products. Thus, we investigated a new processing method for the FGD gypsum and its products’ availability.

### 3.4.1 Material synthesis

**Wallboard production**

Currently, to incorporate FGD gypsum into wallboard, these basic requirements must be met: (a) CaSO₄·2H₂O content exceeds 92%; (b) moisture content less than 10%; (c) CaSO₄·1/2H₂O content is less than 0.5%; (d) Cl⁻ content less than 200 mg/kg; and (e) the total K⁺, Na⁺, and Mg²⁺ content is less than 0.06%.

The production of gypsum boards with increased fire resistance, water resistance, and low density has been the focus of recent studies. According to Leiva et al.¹⁰ and Li et al.¹⁵, the heat insulating capacity of 100% FGD gypsum panels is higher than that of commercial gypsum panels, attributed to the high free water content in FGD gypsum.

In addition, FGD gypsum is being used as a replacement for natural gypsum to produce gypsum blocks. FGD gypsum-containing blocks have poor water resistance compared to FGD gypsum boards, which reduces the benefit of using FGD gypsum in the blocks. However, recent investigations have shown that the water resistance of gypsum blocks can increase when they are made of FGD gypsum using granulated blast furnace slag, high calcium fly ash,¹⁰⁶ and different water-resistant additives.¹⁰⁷

Due to the fact that dechlorinated FGD gypsum and wastewater evaporation products have been heat-treated, they thus have low Cl⁻ content and high CaSO₄·2H₂O content. These characteristics meet the basic requirements for incorporating FGD gypsum into wallboards. Therefore, dechlorinating FGD gypsum and recycling the dechlorinated FGD gypsum and its products will increase the utilization of FGD gypsum and alleviate a series of problems such as land occupation caused by existing FGD gypsum treatment.

**Concrete/cement and asphalt production**

Similar compressive strengths are exhibited by cement produced from natural and synthetic gypsum, revealing that FGD gypsum can replace natural gypsum in concrete production. In concrete production, there are several benefits of adding FGD gypsum as a replacement for CAC: early strength increments, prevention of strength decline at later ages, shrinkage reduction, cost reduction, and sulfate corrosion resistance improvement.¹⁰⁸ Using FGD gypsum in the preparation of calcium sulfoaluminate cement can result in higher strengths when the pastes are cured at 40°C, but lower strengths for curing temperatures of 0, 10, and 20°C. Prior to its use in cement, FGD gypsum needs to be dehydrated.¹⁰⁹ According to these studies, the results showed that (a) as the heat treatment temperature increased, the concrete setting time decreased and (b) cement produced with FGD gypsum calcined at 200°C and showed a maximum strength at a 3.5% SO₃ content.

During the emission reduction process, the dechlorination step heat-treated the FGD gypsum, and the wastewater evaporation step also heat-treated the wastewater evaporation products. According to abovementioned properties, an FGD gypsum admixture that has undergone high-temperature treatment can improve the strength and setting time of concrete. Due to dechlorinated FGD gypsum and wastewater evaporation products both being heat-treated, we can utilize dechlorinated FGD gypsum and wastewater evaporation products into concrete to improve the utilization of total FGD gypsum and to avoid energy consumption to heat-treat FGD gypsum again.

**Other construction materials**

Other novel building materials can also use FGD gypsum as a component. Currently, it is common to use lightweight FGD gypsum to increase lateral stiffness and shear strength in cold-formed steel framing walls, as the gypsum-filled specimens allow an increase in load of 1.72-2.54 greater than the unfilled specimens.¹¹⁰ Furthermore, to increase the strength and water resistance of FGD gypsum blocks, several studies have added silicate clinker. The block properties can be affected by particle size and amount of silicate clinker added.¹¹¹ These show that FGD gypsum crystals can be produced by a hydration process of calcium silicate, ettringite, and FGD gypsum, which improves the performance of the block as a construction material.

The dechlorinated FGD gypsum has a high CaSO₄ purity and contains fewer impurities than FGD gypsum. Furthermore, the main component of the wastewater evaporation products is also CaSO₄, which has a smaller particle size and fewer impurities and toxic substances compared to FGD gypsum. Due to these characteristics, utilizing dechlorinated FGD gypsum and wastewater evaporation products in construction materials can save the cost of handling impurities and can solve the problem of waste being generated by using FGD gypsum.

**CaCO₃ production**

Many different procedures have used FGD gypsum to produce calcium carbonate:

1. Byproduct of elemental sulfur formation (de Beer et al.¹¹²):

\[
\begin{align*}
\text{CaSO}_4 \cdot 2\text{H}_2\text{O}(s) + 2\text{C} (s) & \rightarrow \text{CaS}(s) + 2\text{CO}_2 (g) \\
+ 2\text{H}_2\text{O}(l) & \text{[thermal reduction process]} \\
\end{align*}
\]
\[
\text{CaS (s) + H}_2\text{O (l) + CO}_2\text{ (g) → H}_2\text{S (g)} + \text{CaCO}_3\text{(s) [carbonation process]} (6)
\]

\[
2\text{H}_2\text{S (g) + O}_2\text{ (g) → 2S (s) + 2H}_2\text{O (l)} [\text{recovery of elemental sulfur}] (7)
\]

2. Utilizing atmospheric carbon dioxide to produce calcium carbonate Lee et al.\textsuperscript{112}:

\[
\text{CaSO}_4\cdot2\text{H}_2\text{O (s) + CO}_2\text{ (g) + 2NH}_4\text{OH (l) → CaCO}_2\text{(s)} + (NH}_4\text{)}_2\text{SO}_4\text{(aq)} (8)
\]

The CaCO\textsubscript{3} generated in Equation (6) can be reused in the flue-gas desulfurization process, while (NH\textsubscript{4})\textsubscript{2}SO\textsubscript{4} can be reused as fertilizer; this reaction can be carried out in the field. Therefore, utilizing the dechlorinated FGD gypsum and wastewater evaporation products using this technique can provide sustainable waste management for power plants. In addition to controlling the level of carbon dioxide in the atmosphere, the formation of calcium carbonate can also increase the market value of FGD gypsum.

**Production of calcium sulfate hemihydrate**

Flue-gas desulfurization gypsum is typically used to produce calcium sulfate hemihydrate. This ingredient often requires high temperatures processing and further purification and can be used in building and design, pharmaceuticals, ceramics, and molding configurations.\textsuperscript{115} As shown in Figures 9 and 11, wastewater evaporation products are heat-treated and their main ingredient is calcium sulfate hemihydrate; the emission reduction process includes high-temperature treatment and purification of FGD gypsum. Therefore, using the emission reduction process in coal-fired plants can save costs and energy consumption for producing calcium sulfate hemihydrate.

### 3.4.2 Land applications

Flue-gas desulfurization gypsum is widely used as a soil amendment. Thus, it is used to improve the quality, permeability, and porosity of soil, and to aggregate its soil particles. In addition, FGD gypsum can be used as a fertilizer.

**Soil amendment**

In the past few decades, the use of FGD gypsum as a soil amendment has been thoroughly investigated.\textsuperscript{114,115} Recent studies have mainly focused on improving sodic soils.\textsuperscript{116,117} In addition, the ratio of sodium: (calcium + magnesium) in the soil solution is essential for the desalination process. When Ca\textsuperscript{2+} ions replace the Na\textsuperscript{+} in the soil colloids, it results in increased soil porosity and enhanced water infiltration.\textsuperscript{114} Due to the high Ca\textsuperscript{2+} content of FGD gypsum, it can easily be used to replace the exchangeable Na\textsuperscript{+} in soil. Thus, FGD gypsum plays an important role in soil desalination, and the dissolution-exchange reaction in the soil colloids can be expressed as:

\[
\text{NaX + CaSO}_4\rightarrow\text{CaX} + \text{Na}^+ + \text{SO}_4^{2-} (9)
\]

where X is the exchangeable form of Na compounds.

The particle sizes of the wastewater evaporation products with high concentrations of Ca\textsuperscript{2+} and Na\textsuperscript{+} ions are smaller than those of FGD gypsum. The main component is similar to that of FGD gypsum and does not contain many impurities. Its small particle size is good for penetrating the soil, and its high Ca\textsuperscript{2+} content is good for replacing Na\textsuperscript{+}. Therefore, the wastewater evaporation products are suitable for soil amendment use.

**Fertilizer**

Flue-gas desulfurization gypsum is used to provide calcium and sulfur for plant growth. In addition, there are large amounts of Mg, K, and Se in FGD gypsum. Even if excessive selenium is toxic, it is considered a dietary essential.\textsuperscript{118}

However, neither the dechlorinated FGD gypsum nor the wastewater evaporation products contain Se. If using these two products as a fertilizer in large quantities, they will not cause toxicity to plants due to Se accumulation. Therefore, a large amount of fertilizer made from FGD gypsum and wastewater evaporation products could be beneficial to the growth of some crops and increase their yields.

**Reduction of soil erosion and eutrophication**

Many studies have reported that FGD gypsum reduces surface runoff by decreasing surface sealing/crusting, increasing infiltration, and reducing the effectiveness of sediment transport,\textsuperscript{116,118} which indirectly helps to prevent eutrophication. The higher Ca\textsuperscript{2+} content in FGD gypsum-modified soil promotes flocculation of clay particles, which increases the porosity of the soil. The soil structure with higher porosity can increase water permeability and reduce surface runoff and sediment transportation, thereby preventing eutrophication. In order to achieve the same purpose, applying FGD gypsum and wastewater evaporation products can be beneficial for promoting the flocculation of clay particles, which increases the porosity of the soil. In addition, the small particle size wastewater products are more likely to enter higher porosity soil so they can further exert its functions.

**Mine reclamation**

Compared to natural soils, mineral soils usually have lower organic matter and nutrient contents, higher Fe-oxide and toxic metal contents, and lower pH and water retention capacities.\textsuperscript{115,119} Mine drainage and acidic soils can be neutralized by the alkaline nature of CCPs, thus reducing the solubility of
metals by precipitating metal hydroxides. Comparative studies have been conducted using different materials (FGD gypsum, fly ash, bio-solids, and zeolites) to amend mine soil.\cite{120} The highest pH increments were associated with FGD gypsum amendment, while the highest water retention capacity was associated with bio-solid amendment. A recent study has investigated the long-term effects of FGD gypsum application on coal mine reclamation, both alone and when mixed with compost.\cite{121,122} This study has revealed that using only FGD gypsum increased the pH of mine soil from 3.1 to 6.9 after one year and maintained the pH at ~6.4 after 15 years of amendment. In addition, after 16 years of FGD gypsum amendment, there was increased bacterial populations and diversity in mine soil.

Furthermore, both the dechlorinated FGD gypsum and wastewater evaporation products have high water content. Using these two wastes to replace the use of FGD gypsum in mine reclamation could further improve the low water retention capacity and pH value of the mineral soil. In conclusion, the use of the dechlorinated FGD gypsum and wastewater evaporation products either alone or mixed with compost can effectively remediate long-term abandoned mines, thereby improving revegetation and water quality in the reclaimed areas.

Remediation of heavy metal-contaminated soil
As shown in Figure 12, Wang and Yang (2018)\cite{64,43} have shown that FGD gypsum can be used for several remediation applications. They introduced the effects and mechanisms of the application of FGD gypsum in degraded soils.

To stabilize contaminated soils, fly ash and bottom ash have been wildly utilized.\cite{123,124} Recently, several studies reported that a mixture of calcite and FGD gypsum can successfully remediate soil contaminated by pyritic minerals.\cite{125} The influence of FGD gypsum and natural gypsum on soil chemistry has been compared by Kost et al.\cite{42} After one year at an addition rate of 20 Mg/ha, the pH of the soil treated with FGD gypsum was lower than that of untreated soil, and lower than that of soil treated with natural gypsum, because the CaCO$_3$ content in natural gypsum is higher.

In addition to the chemical and physical properties of the soil, the presence of soil microbial communities is also very important for agricultural productivity and nutrient balance. Chen et al.\cite{126} assessed the ability of FGD gypsum and natural gypsum to alter the effects of the Hg, Se, and As content in earthworms and agricultural soil when amended with them. The earthworm population density (expressed in number of earthworms/m$^2$) and biomass density (expressed as the dry weight of earthworms in g/m$^2$) of FGD gypsum- and natural gypsum-amended soil were greatly reduced, compared to the control soil. The decrease in earthworm density and biomass can be attributed to the increase in the soluble salt content of the soil by gypsum application. The effect of FGD-CaSO$_3$ on the soil microbial community was evaluated by measuring the enzyme activity of FGD-CaSO$_3$; There were no major differences of FGD gypsum and FGD-CaSO$_3$ amendments on soil enzymatic activities.\cite{127}

A new trend in remediation of contaminated soil is the use of waste to increase the pH value of the soil, which can also reduce the amount of waste that requires disposal. As the dechlorinated FGD gypsum and wastewater evaporation products are solid wastes from coal-fired power plants, both have similar physical and chemical properties to several other CCPs. Thus, the dechlorinated FGD gypsum and wastewater evaporation can replace the application of FGD gypsum in the remediation of heavy metal-contaminated soil in some aspects. According to the abovementioned factors, we should focus on how to efficiently use these two wastes to repair heavy metal-contaminated soils to improve the utilization of desulfurization waste for the benefit of coal-fired power plants.

3.4.3 Water treatment processes
Due to FGD gypsum’s high Ca$^{2+}$ and SO$_4^{2-}$ contents and alkaline pH, water treatment is one of its important new applications. As shown in Table 1, Koralgedara et al.\cite{118} summarized and discussed the beneficial uses of FGD gypsum in water treatments in detail. In addition, both the dechlorinated FGD gypsum and wastewater evaporation products have similar characteristics with FGD gypsum, and the impurity content of these two products is extremely low. Using these two wastes to replace the use of FGD gypsum in water treatment processes could further improve water treatment efficiency.

3.5 Feasibility of the craft enlargement experiment
This study proposed an emission reduction process for the dechlorination of FGD gypsum and its wastewater treatment and analyzed the feasibility of the emission reduction process according to laboratory experiments. The laboratory experiment has certain limitations and cannot fully show the actual situation as it would occur in industrial production. We can only qualitatively analyze the emission reduction process through laboratory experiments to judge the feasibility of the process. Thus, we performed these laboratory experiments so that they can play a guiding role in process amplification.

In addition, alkaline flue duct spray can improve the dechlorination efficiency and further reduce the amount of desulfurization wastewater.\cite{128} Furthermore, these factors reduce the chlorine content of the desulfurization slurry, the production of dechlorination wastewater, and the chlorine
content in FGD gypsum. Due to the limitations of this study, the effects of saturated steam with different temperatures, compositions, pH, and spray time on the dechlorination efficiency of FGD gypsum and its wastewater volume can be further evaluated; this will be completed in future work. Moreover, the availability of wastewater evaporation products under the above conditions can also be investigated.

As shown in Figure 13, the emission reduction process couples the dechlorination process of saturated steam spraying FGD gypsum and the “zero emissions” wastewater process of flue duct spray evaporation. The laboratory experiments conducted in this study were either based on the conventional process currently used on site or the proposed emission reduction process that could be used on site. The conventional process utilizes process water and steam to dechlorinate FGD gypsum. The resulting dechlorination wastewater is removed by heat from the flue duct, which in turn reduces chloride ions in the flue gas. A similar cycle of dechlorination can be achieved in the emission reduction process, thus improving the dechlorination conditions on site.

According to these laboratory test results, the average chloride ion content of FGD gypsum can be reduced to below

**FIGURE 12** Mechanisms and effects of flue-gas desulfurization gypsum remediation in degraded soils, reproduced with modifications from Wang and Yang.

**TABLE 1** Details of the application of flue-gas desulfurization gypsum in the treatment of different types of water from Nadeesha H. Koralegedara et al.

| Application                        | Water type                  | Mechanism                                                                                     | References                                    |
|-----------------------------------|-----------------------------|----------------------------------------------------------------------------------------------|-----------------------------------------------|
| Heavy metal removal               | Industrial wastewater       | Precipitation, stable mineral formation, adsorption with the aid of SO$_4^{2-}$, OH$^-$, CO$_3^{2-}$, Ca$^{2+}$, and Fe-Al oxides | Jayaranjan and Annachhatre; Yan et al        |
| Cyanobacterial growth removal     | Lake water                  | Charge neutralization by Ca$^{2+}$ addition and promotes coagulation                          | Whangchai et al                              |
| Cyanobacterial growth removal     | Lake water                  | Charge neutralization by Ca$^{2+}$ addition and promotes coagulation                          | Whangchai et al                              |
| Dye removal                       | Industrial wastewater       | Adsorption                                                                                    | Deniz and Saygideger                         |
| Dissolved organic carbon removal  | Lake water, agricultural drainage | Ca$^{2+}$ cross-linked organic matter with clay particle in sediment                        | Varcoe et al                                 |
| Reduce fecal bacteria contamination | Poultry litter drainage   | Charge neutralization by Ca$^{2+}$ addition and coagulation. Reduced bacterial growth by increasing pH | Jenkins et al                                |
300 mg/L by using the emission reduction process to treat high-chlorine FGD gypsum and its dechlorination wastewater. The dechlorination rate of the saturated steam spray dechlorination test was 97.5% of that of the conventional process, while the wastewater output was 4.9% of that of the conventional process. The saturated steam spray dechlorination process can therefore meet the dechlorination demands of the on-site FGD gypsum and can further reduce the output of dechlorination wastewater. The flue duct spray evaporation for dechlorination wastewater tests used the residual thermal energy of the flue duct to evaporate the wastewater and then discharge bottom ash separately or with the fly ash, which solves the wastewater treatment and discharge problems present in the conventional dechlorination process of FGD gypsum. The emission reduction process has a more efficient wastewater reduction and dechlorination efficiency in addition to a more economical process cost when compared with the conventional processes. The results of this study provide theoretical and engineering guidance for further research on dechlorination, pollution reduction, and utilization of the solid waste of FGD gypsum.

4 | CONCLUSIONS

In this study, given that utilities are committed to reducing the environmental impact of coal-fired power generation in the future, an emission reduction process was proposed for processing FGD gypsum and reducing its wastewater output. The average dechlorination rate of the saturated steam spray dechlorination process was 80.4%, and its average dechlorination rate reached 97.5% of the conventional process of water spray dechlorination. The entire steam spray dechlorination process used very little water; the average wastewater volume was 0.22 L. When the dechlorination was the same, the output of the dechlorination wastewater accounted for 4.9% of the conventional dechlorination process. Therefore, dechlorination with saturated steam in the emission reduction process can effectively remove chlorine from FGD gypsum and reduce its wastewater output. It also effectively reduces the chlorine content of FGD gypsum and related wastewater. In addition, this emission reduction process can lead to greater recovery of renewable resources and can result in a cleaner energy conversion process. Based on these results, the craft enlargement experiment of the emission reduction process is feasible and can be further explored. Moreover, dechlorinated FGD gypsum and wastewater evaporation products are available to material synthesis, land applications, and water treatment processes.

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CONFLICTS OF INTEREST
The authors declare no conflict of interest, and the manuscript is approved by all authors for publication. I would like to declare on behalf of my co-authors that the work described was original research that has not been published previously, and not under consideration for publication elsewhere, in whole or in part. All the authors listed have approved the manuscript that is enclosed.

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