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# Effect of calcium formate as an additive on desulfurization in power plants

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## ABSTRACT

SO<sub>2</sub> in flue gas needs to be eliminated to alleviate air pollution. As the quality of coal decreases and environmental standard requirements become more stringent, the high-efficiency desulfurization of flue gas faces more and more challenges. As an economical and environmentally friendly solution, the effect of calcium formate as an additive on desulfurization efficiency in the wet flue gas desulfurization (WFGD) process was studied for the first time. Improvement of the desulfurization efficiency was achieved with limited change in pH after calcium formate was added into the reactor, and it was found to work better than other additives tested. The positive effects were further verified in a power plant, which showed that adding calcium formate could promote the dissolution of calcium carbonate, accelerate the growth of gypsum crystals and improve the efficiency of desulfurization. Thus, calcium formate was proved to be an effective additive and can potentially be used to reduce the amount of limestone slurry required, as well as the energy consumption and operating costs in industrial desulfurization.

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## Introduction

With the increasingly rapid development of the global economy, air pollution has become more and more serious due to the increase of annual energy consumption worldwide in recent years. Especially in developing countries, air pollution has increased along with urbanization and industrialization (Kan et al., 2009; Leung, 2015; Wang et al., 2016; Zheng et al., 2011). In recent years, serious smog pollution in China – a weather phenomenon featuring a high concentration of fine particles in the air, which leads to visibility loss (less than 10,000 m at a relative humidity (RH) lower than 90%) – has greatly hindered people's outdoor activities and also poses severe potential harm to human health (Kampa

and Castanas, 2008; Shen, 2015; Tan et al., 2009; Zhuang et al., 2014).

Clean energy and renewable energy can alleviate energy shortages and pollution, but renewable energy resources such as solar, wind and biomass energy are still under exploration and have not yet been put into large-scale use, while fossil fuels like petroleum, natural gas and coal are still the main energy sources (Forsberg, 2009; Simons, 2005). In the power plants of China, coal is widely used due to its relatively low price and abundant reserves (Hadjipaschalis et al., 2009). However, sulfur compounds in coal will inevitably generate a large amount of pollutants during combustion. One of the main types of air pollutants is sulfur oxides (SO<sub>x</sub>), which are generated when sulfur compounds react with oxygen. In

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China, the pollution caused by emission of  $\text{SO}_x$  is one of the main barriers to the efficient use of coal (Guan et al., 2003). In practice, power plants are the main sources of  $\text{SO}_x$ , which is a major cause of acid rain around the world (Lu et al., 2010; Mohanty et al., 2009; Ohara et al., 2007; Shen et al., 1991; Zhao et al., 2008). Therefore, to alleviate air pollution, flue gas needs to be desulfurized before emission. Many kinds of desulfurization technologies such as the wet process, dry process, semi-dry process, biological method and electronic method have been applied to solve the problem (Hansen and Kiil, 2012; Lin et al., 2015; Mo et al., 2007; Wang et al., 2013; Zhang et al., 2006). Among them, wet desulfurization technology is used most widely on account of its highly operational stability, high efficiency and long experience. Limestone/lime is widely applied in the wet flue gas desulfurization (FGD) process due to its relatively low price (Heidel et al., 2014; Ren et al., 2010). However, with the increased stringency of environmental standards and the gradual decrease of coal quality (Ito and Rob van Veen, 2006; Liu et al., 2005; Lin et al., 2011; Mortaheb et al., 2012; Tewalt et al., 2005), the desulfurization apparatus for coal gas is confronted with more and more challenges. An economical solution is to use additives which can effectively increase the mass transfer coefficient of the liquid phase and the pH buffer capacity between the gas-liquid interface, thus improving desulfurization efficiency (Heidel et al., 2014).

Q4 According to Heidel et al. (2014) and Ren et al. (2010), a good additive in the limestone/lime FGD process can improve the absorbent reactivity, reduce the dissolution resistance of the absorbent or diffusion resistance of  $\text{SO}_2$ , and keep the pH value of the desulfurization slurry more stable. Desulfurization additives can be classified into organic additives or inorganic ones according to their different mechanisms. The organic additives mainly include acids such as benzoic acid, adipic acid, formic acid, acetic acid and citric acid, which have a buffering effect (Dong et al., 2004a; Kong et al., 2001; Wu et al., 2003); the inorganic additives are mainly sodium salts, ammonium salts and magnesium compounds (Dong et al., 2004b; Sun et al., 2001, 2002a, 2002b, 2002c). The organic additives are widely used in power plants due to their ability to buffer the pH and improve the growth of gypsum crystals, which can thus significantly increase desulfurization efficiency. However, formic acid and acetic acid are liquid phase additives with a relatively high volatility at room temperature, for which the transportation costs have greatly increased, while the solid phase additives such as benzoic acid and adipic acid are very expensive, and their limited pH buffer capacity can significantly affect the pH value in a desulfurization system and even lead to the pH getting out of control. Furthermore, the production and usage of organic acids generate a large amount of effluents that pollute the surrounding soil and water. Also, some of these additives have the side-effect of irritating the skin, mucosa, and eyes, etc. In addition, if the organic additives carried out with gypsum in the desulfurization system are not handled in time and with care, they may cause water and soil pollution that harm many kinds of plants and even animals that eat the contaminated plants.

Calcium formate, which is widely used with livestock, the chemical industry, environmental protection and construction, is a common additive for feed, chemicals, construction

and tanneries (Antipov and Aleshin, 2002; Blinova et al., 2005; Chernoplekov, 2001). It is not only easier to transport as a solid phase than formic acid, but also cheaper than adipic acid. To the best of our knowledge, calcium formate has not been used as a desulfurization additive so far, though many other kinds of calcium compounds such as calcium chloride and calcium nitrate have been studied (Jorgensen et al., 1986). Thus, the potential for using calcium formate as a desulfurization additive in power plants is worthy of investigation.

In this work, the effect of calcium formate as an additive on desulfurization was investigated in a stirred tank. The limestone requirement and pH value of the lime-gypsum slurry were determined after different amounts of calcium formate were added. In addition, the mechanism for the desulfurization efficiency improvement with calcium formate was investigated in depth and the results were verified in an industrial power plant.

## 1. Materials and methods

### 1.1. Materials

Sulfur dioxide ( $\text{SO}_2$ , high purity, purity of 99.999%) was purchased from Tianjin Liufang Industrial Gases Co., Ltd., Tianjin, China; Calcium carbonate, adipic acid, citric acid and formic acid (AR,  $\geq 99.0\%$ ) were purchased from Tianjin Guang Fu Fine Chemical Research Institute, Tianjin, China; Calcium formate (AR,  $\geq 99.0\%$ ) for the laboratory tests was purchased from Real & Lead Chemical Co., Ltd., Tianjin, China. The calcium formate (mass fraction purity  $\geq 98.0\%$ ) for the industrial scale test was purchased from Zibo Ruibao Chemical Co., Ltd., Shandong, China, and used as an additive in the power plant. They were all used without further treatment.

### 1.2. Desulfurization efficiency measurement

The experiment aiming to study the effect of calcium formate as additive on desulfurization was conducted in a 60 mm diameter  $\times$  1000 mm tall packed tower (as shown in Fig. 1) at  $50^\circ\text{C}$ . The experiment was conducted under the following conditions: the flue gas flow rate was  $15 \text{ m}^3/\text{hr}$  with  $\text{SO}_2$  concentration of  $2200 \text{ mg}/\text{m}^3$  in air, the liquid-gas ratio was

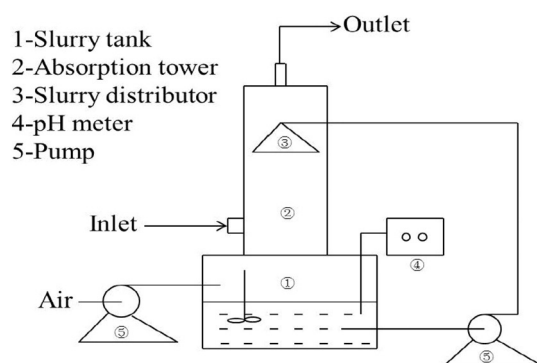


Fig. 1 – Absorptive desulfurization apparatus.

169 3.0 L/m<sup>3</sup>, the gypsum slurry contained 1 wt.% CaCO<sub>3</sub>, and the  
 170 initial pH of the slurry was 5.0. The lime-gypsum slurry with  
 171 different concentrations of calcium formate was added in a  
 172 pulp tank, then was pumped to the top of the tower and  
 173 distributed to increase the interfacial area of mass transfer;  
 174 the pH values of the slurry were recorded by a PHS-3C pH  
 175 meter (PHS-3C, Shanghai INESA Scientific Instrument Co.,  
 176 Ltd., China). The SO<sub>2</sub> concentrations of inlet and outlet gases  
 177 were analyzed on-line by a Testo 350 flue gas analyzer (Testo  
 178 350, Testo Instruments International Trading Co., Ltd., China).  
 179 The desulfurization efficiency (De.) was calculated by the  
 180 following equation:

$$\text{De.(\%)} = \frac{c(\text{SO}_2)_{\text{in}} - c(\text{SO}_2)_{\text{out}}}{c(\text{SO}_2)_{\text{in}}}$$

182 where *c* is the mass concentration of SO<sub>2</sub> in gas, mg/m<sup>3</sup>.

### 183 1.3. Power plant verification

184 The conditions of the industrial desulfurization process in  
 185 a power plant are much more complicated than that in a  
 186 laboratory. To check whether the results in a power plant  
 187 were in accordance with the results in the laboratory, the  
 188 desulfurization efficiency of the lime-gypsum slurry with  
 189 calcium formate additive was evaluated in a system including  
 190 two circular sections (as shown in Fig. 2). The first one mainly  
 191 pretreats flue gas and oxidizes calcium sulfite to gypsum at  
 192 pH value of 4.5–5.2, temperature of 50–60°C; the second one  
 193 mainly removes SO<sub>2</sub> from flue gas to the desired purity at pH  
 194 value of 5.8–6.4, temperature of 50–60°C. The experiment was  
 195 conducted at a limestone slurry spray flow rate of 14.1 m<sup>3</sup>/hr,  
 196 flue gas flow rate of 1.0 × 10<sup>6</sup> m<sup>3</sup>/hr (under standard conditions),  
 197 circulating liquid–gas ratio of 20 L/m<sup>3</sup> and various concentra-  
 198 tions of calcium formate.

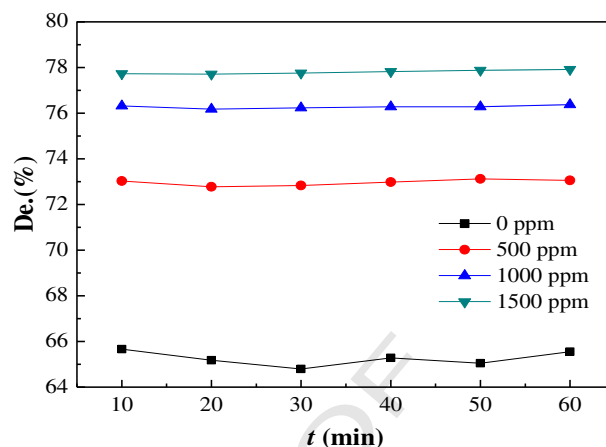


Fig. 3 – Desulfurization efficiency at different calcium formate concentrations. *t*: time; *De.*: desulfurization efficiency.

## 200 2. Results and discussion

### 201 2.1. Effect of calcium formate concentrations on 202 desulfurization efficiency

The desulfurization efficiency at different calcium formate 203  
 concentrations is presented in Fig. 3. The results indicated 204  
 that the desulfurization efficiency was improved by adding 205  
 calcium formate, and the efficiency was further enhanced by 206  
 increasing the concentration of calcium formate. After calci- 207  
 um formate was added into the slurry, the Ca<sup>2+</sup> concentration 208  
 in the liquid phase was increased, which accelerated the 209  
 formation of CaSO<sub>4</sub>. In addition, the acidity of the liquid phase 210  
 was increased, which promoted the dissolution of CaCO<sub>3</sub> in 211  
 the slurry. As reported by Ma et al. (Heikal, 2004; Ma et al., 212

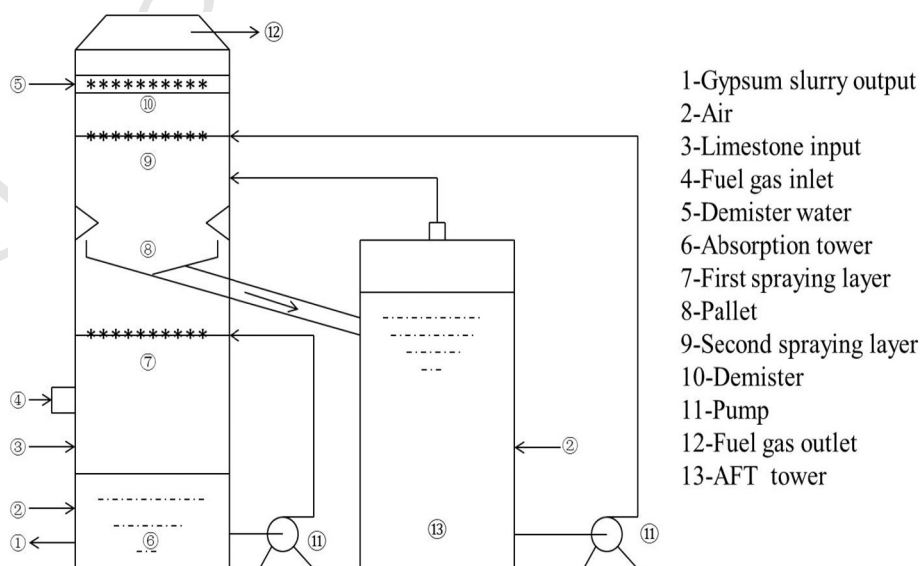


Fig. 2 – Double cycle wet desulfurization process. AFT: absorber feed tank.

213 2013; Singh and Abha, 1983), calcium formate acted as an  
 214 accelerator for the hydration of calcium salts and could  
 215 promote the formation of  $\text{CaSO}_4$ , accelerate the growth of  
 216 gypsum crystals and then facilitate their separation from the  
 217 slurry. So, adding calcium formate enhanced the reactivity  
 218 of the limestone slurry absorbent and then improved the  
 219 utilization rate of the absorbent.

## 220 2.2. Effect of calcium formate concentrations on pH value of 221 lime-gypsum slurry

222 The pH value affects many aspects of the operation of a  
 223 desulfurization system. On one hand, a relatively high pH  
 224 value contributes to the dissolution of  $\text{SO}_2$ , so that the  
 225 dissolution rate of  $\text{SO}_2$  at pH 6 is about 5–10 times as high as  
 226 that at pH 4; on the other hand, a relatively low pH value is  
 227 helpful for limestone dissolution. In practice, the appropriate  
 228 pH value in the absorption liquid in desulfurization tower  
 229 is between 4 and 6, which also leads to high oxygenation  
 230 efficiency for  $\text{SO}_3^{2-}$ .

231 The pH value of the lime-gypsum slurry at different  
 232 calcium formate concentrations is shown in Fig. 4. It was  
 233 clear that the pH value of the lime-gypsum slurry showed a  
 234 slight decline, and the decline trend became more evident  
 235 with increasing calcium formate concentration. This was  
 236 because calcium formate could accelerate the migration of  
 237  $\text{SO}_2$  from gas to liquid. This migration speed increased with  
 238 increasing concentrations of calcium formate in the slurry,  
 239 resulting in the increase of the  $\text{SO}_2$  concentration in the liquid  
 240 phase. The decline of pH can be attributed to the increasing  
 241  $\text{SO}_2$  concentration in the liquid phase. On the other hand, the  
 242 limited decline in pH was because calcium formate could  
 243 facilitate the absorption of  $\text{SO}_2$  in the liquid film and kept  
 244 the concentration of  $\text{SO}_2$  in the liquid phase relatively stable,  
 245 which made the pH value in the system controllable.  
 246 Moreover, the decline of pH helped to accelerate the oxidation  
 247 from  $\text{CaSO}_3$  to  $\text{CaSO}_4$  and the dissolution of  $\text{CaCO}_3$ . As  
 248 reported by Zhou (2005), sulfite in the slurry was oxidized  
 249 very fast at pH 4.5–5.0, in accord with the improvement of  
 250 desulfurization efficiency.

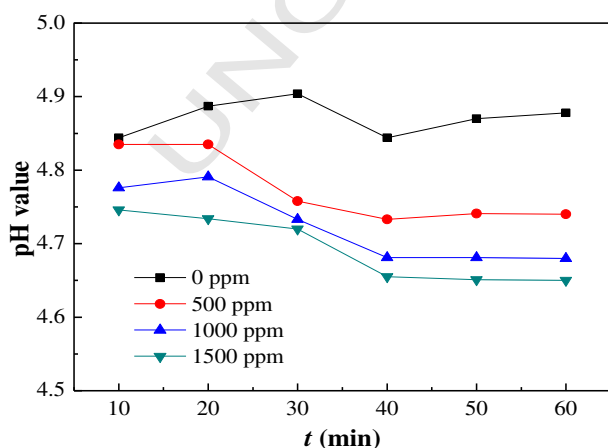


Fig. 4 – pH value of lime-gypsum slurry at different calcium formate concentrations.

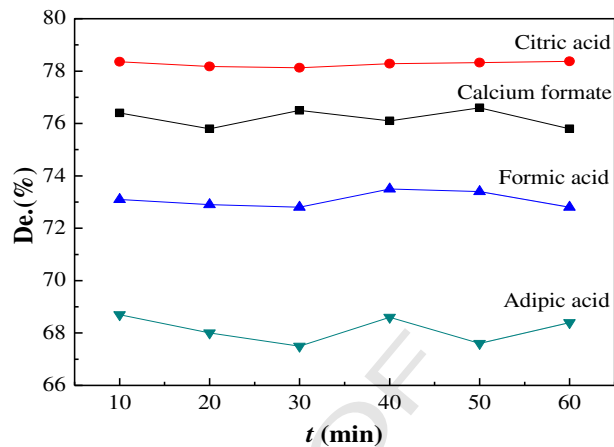


Fig. 5 – Effect of different kinds of additives on desulfurization efficiency.

## 251 2.3. Comparison of calcium formate with other additives

252 The effects of different kinds of additives on desulfurization  
 253 efficiency were studied at 1000 ppm of each additive, and  
 254 the results are shown in Fig. 5. As shown in Fig. 5, the  
 255 desulfurization efficiency of calcium formate was higher than  
 256 that of formic and adipic acids, while it was lower than that of  
 257 citric acid. Given that the price of calcium formate is lower  
 258 than citric acid, it is reasonable to infer that using calcium  
 259 formate as an additive is economical and has good potential  
 260 to improve the desulfurization efficiency and reduce  $\text{SO}_x$   
 261 emission in power plants.

## 262 2.4. Verification results in a power plant

263 The results showing the effect of calcium formate as an  
 264 additive on the desulfurization efficiency of the wet lime-  
 265 gypsum process in a power plant are shown in Figs. 6–8.

266 Fig. 6 shows the effect of calcium formate concentration on  
 267 desulfurization efficiency for different concentrations of  $\text{SO}_2$ .  
 268 It was clear that for the  $\text{SO}_2$  concentration of  $2500 \text{ mg/m}^3$ , the  
 269 average concentration of  $\text{SO}_2$  in the outlet gas declined from  
 270  $47.2$  to  $14.9$  and  $12.7 \text{ mg/m}^3$ , and the average desulfurization  
 271 efficiency over the time of the experiment was increased from  
 272  $98.05\%$  without calcium formate to  $99.48\%$  at calcium formate  
 273 concentrations of  $500$  and  $1000 \text{ ppm}$ . At a  $\text{SO}_2$  concentration  
 274 of  $3500 \text{ mg/m}^3$ , the average concentration of  $\text{SO}_2$  in the  
 275 outlet gas declined from  $78.8$  to  $68.0$  and  $46.5 \text{ mg/m}^3$ , and  
 276 the average desulfurization efficiency rose from  $97.83\%$  to  
 277  $98.08\%$  and  $98.78\%$  at calcium formate concentrations of  $500$   
 278 and  $1000 \text{ ppm}$ , respectively. Clearly, the addition of calcium  
 279 formate significantly improved the desulfurization efficiency  
 280 in the power plant. As reported by Liu and Xiao (2006), too  
 281 much  $\text{SO}_2$  in fuel gas could lead to obstruction and instability of  
 282 the desulfurization system. Fig. 7 shows that the pH value in the  
 283 absorption tower at different concentrations of calcium formate  
 284 was almost the same as the pH value without addition of  
 285 calcium formate, which demonstrated that the pH was still  
 286 under control with the addition of calcium formate, and no  
 287 negative influences were found in the system of the industrial  
 288 power plant. In accordance with the results in Fig. 4, this can

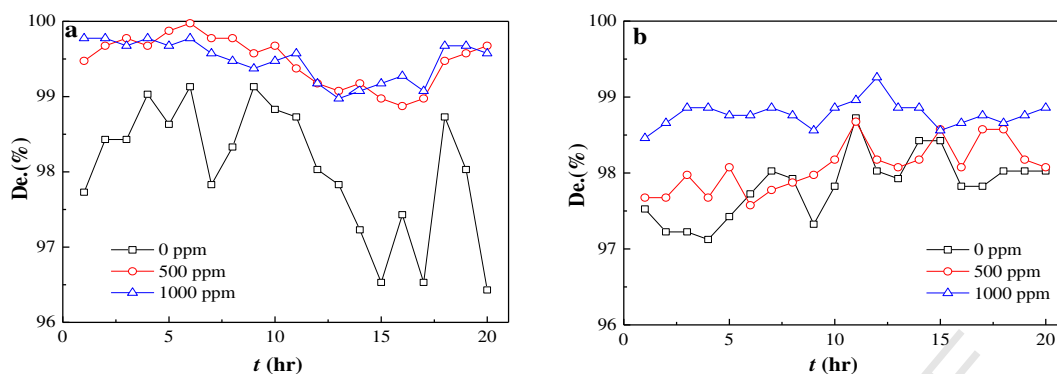


Fig. 6 – Desulfurization efficiency at different concentrations of calcium formate under  $\text{SO}_2$  concentration of (a) 2500 and (b) 3500  $\text{mg/m}^3$ .

289 further illustrate that even though the  $\text{SO}_2$  concentration in the  
 290 liquid phase was increased, the absorption tower in the  
 291 desulfurization system had the ability for self-restoration by  
 292 changing the dissolution rate of  $\text{CaCO}_3$  and the separation rate  
 293 of  $\text{CaSO}_4$ . Fig. 8 shows the amount of limestone required to  
 294 obtain the same desulfurization efficiency at different concentra-  
 295 tions of calcium formate along with the variation of the inlet  
 296 concentration of  $\text{SO}_2$ . It was clear that the amount of calcium  
 297 carbonate needed to obtain the same desulfurization efficiency  
 298 was decreased with increasing concentrations of calcium forma-  
 299 te. At the  $\text{SO}_2$  inlet concentration of 3500  $\text{mg/m}^3$ , the  
 300 limestone amount required was reduced by 4%–9%. Therefore,  
 301 adding calcium formate can promote the dissolution of calcium  
 302 carbonate and maintain a relatively high desulfurization effi-  
 303 ciency at a low limestone amount. Since the amount of calcium  
 304 carbonate slurry consumed in a power plant is directly related  
 305 to the economic benefit, the addition of the calcium formate is  
 306 beneficial for the power plant.

### 308 3. Conclusions

309 The effect of calcium formate as an additive on desulfurization  
 310 efficiency improvement was investigated both at laboratory

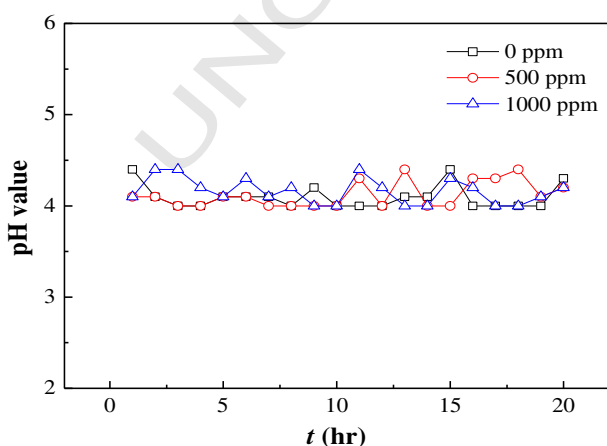


Fig. 7 – pH value of absorption tower at different concentrations of calcium formate.

scale and at industrial scale. The results indicated that the  
 311 addition of calcium formate in the lime-gypsum slurry could  
 312 not only promote the dissolution of calcium carbonate, the  
 313 formation of  $\text{CaSO}_4$  and the growth of gypsum crystals, but also  
 314 accelerate the migration velocity of  $\text{SO}_2$  from gas phase to liquid  
 315 phase and the absorption rate of  $\text{SO}_2$  in the liquid film. As a  
 316 result, a significant improvement in desulfurization efficiency  
 317 was observed after calcium formate was added. The degree of  
 318 increase was larger than 1% at the inlet gas  $\text{SO}_2$  concentration of  
 319 2500  $\text{mg/m}^3$  and nearly 1% at the inlet gas  $\text{SO}_2$  concentration of  
 320 3500  $\text{mg/m}^3$  in the power plant. The amount of limestone slurry  
 321 needed to maintain the same desulfurization efficiency could  
 322 be reduced. Thus, the energy consumption in a desulfurization  
 323 system using calcium formate will be reduced significantly, as  
 324 will the operating costs. All these findings demonstrated that  
 325 calcium formate could be used as an additive to enhance the  
 326 desulfurization efficiency in power plants. 327

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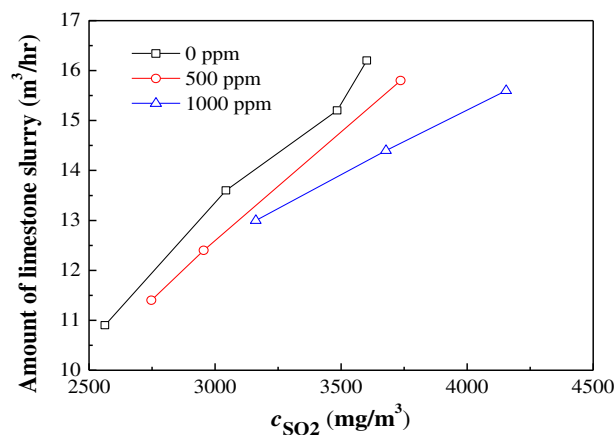


Fig. 8 – Limestone amount required for adsorption of different inlet  $\text{SO}_2$  concentration ( $c_{\text{SO}_2}$ ) at different concentrations of calcium formate.

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