



# Self-anticorrosion for the combustion tower of heat recovered thermal process phosphoric acid production



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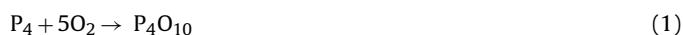
## ABSTRACT

When a combustion boiler is used to recover heat released from yellow phosphorus combustion for phosphoric acid production, serious corrosion of the inner wall of the combustion tower caused by high temperature acid substances becomes a very critical problem. Based on pilot experiments, we found that a layer of anti-corrosion protective coating can be generated and adhered on the inner wall of the combustion tower by controlling the process parameters, including  $R_{H_2O-P_4O_{10}}$  (the molar ratio of moisture of feeding air to the phosphorus pentoxide produced from reaction) and  $T_A$  (the temperature of the outlet gas from the combustion tower). This coating can prevent high temperature acid gas from directly contacting the inner wall of the combustion tower, so that the tower can be effectively protected from corrosion. This work innovatively realizes the self-anticorrosion of the combustion tower through controlling the reaction conditions. The main composition and preliminarily conjectured molecular structure of this coating was firstly identified by the analyzing its chemical components, physical and chemical properties, crystal form, and other detection data. Finally, operating conditions for stabilizing this coating were proposed and verified based on an industrialized production process.

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## 1. Introduction

Phosphoric acid is widely used in the chemical and other industries, including food (Shen et al., 2016; Hao et al., 2014), electronics (Jiang et al., 2011), catalyst (Zhang et al., 2015; Li et al., 2016), biomass treatment (Wang et al., 2018), activated carbon (Marzbali et al., 2016; Mahmood et al., 2017) etc. Besides, phosphoric acid can also be further processed into a variety of phosphate to be used into various fields, such as coating (Knohl et al., 2013; Peres et al., 2014), flame retardant (Shan et al., 2011; Liao et al., 2014), drug (He et al., 2017; Yuan et al., 2013), food (Minh et al., 2012; Doan et al., 2015), animal food (Willis and Montgomery, 1994), fire protection. Thermal process phosphoric acid (TPA) production has been widely applied for a long term. In this process, phosphoric acid is produced by using water (or dilute phosphoric acid) to absorb phosphorus pentoxide ( $P_4O_{10}$ ) which is obtained through burning yellow phosphorus under well-oxygenated conditions (Babrauskas, 2017). The reaction equations are as follows:



At present, there are principally two process routes to produce TPA by yellow phosphorus combustion in the world, i.e. one-step process (combustion and hydrated reactions are completed in one reaction tower) and two-step process (combustion and hydrated reactions are separately done in two reaction towers).

Yellow phosphorus will release massive heat during combustion. Theoretically,  $24.335 \times 10^3$  kJ of heat can be released by burning 1 kg of yellow phosphorus (Mei et al., 2005). Thus, it is considerably significant to recover and reuse such massive heat. In order to effectively recover this heat, Yunnan Research Institute of Chemical Industry has conducted a lots of works and made great achievements since 1990s (Mei et al., 2005). This technology adopts a two-step process, i.e. combustion and hydrated reactions are done separately in two towers. A special design has been made for combustion tower to provide a structure similar to a steam boiler, so that heat released from yellow phosphorus combustion can be recovered to generate steam.

With this heat recovery technology, 50% of heat released by yellow phosphorus combustion can be recovered at least. It means that 4.57 t (0.5 MPa) of steam can be produced in combustion of 1 t of yellow phosphorus (Yang et al., 2005). As a result, the cost for producing phosphoric acid can be reduced by 126 RMB/t based

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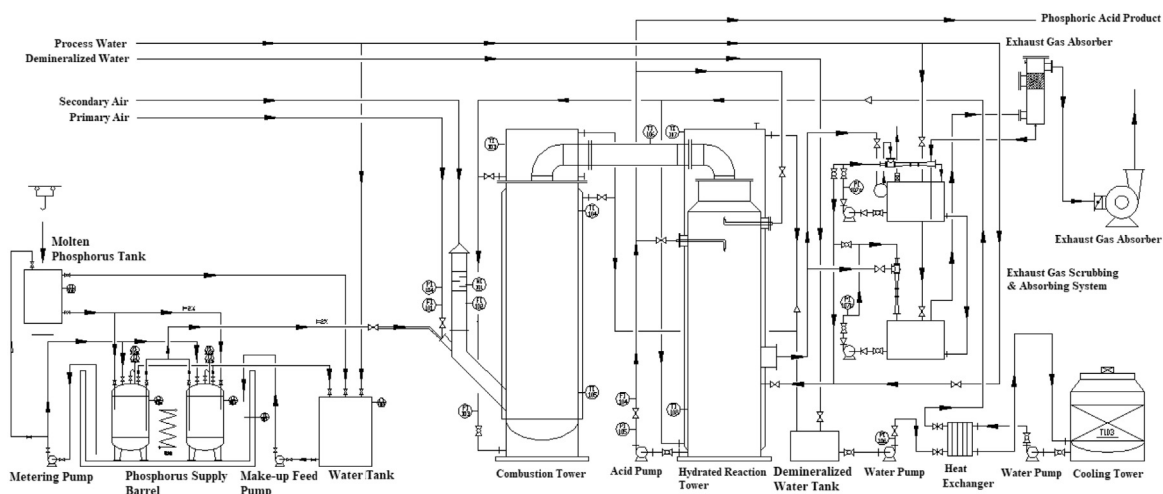


Fig. 1. Process flow chart of the phosphoric acid preparation part of the pilot plant.

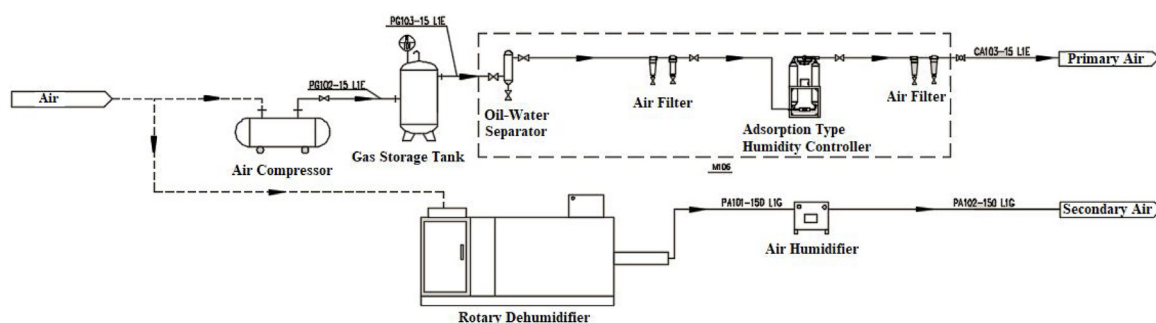


Fig. 2. Process flow chart of the air humidity control part of the pilot plant.

on a given steam price of 100 RMB/t (Yang et al., 2005). For a TPA plant with a capacity of 50,000 ton/a, the two-step process could reduce coal consumption from 6640 ton/a to zero, electricity consumption by  $2.05 \times 10^6$  kWh per year, and water consumption by 262,600 ton/a compared with the traditional one-step process (Yang et al., 2008). Therefore, the two-step process has a positive effect on energy conservation and emission reduction in China.

The produced phosphorus pentoxide, phosphoric acid and other phosphoric substances can be heated to an extremely high temperature by the heat released from yellow phosphorus combustion in the combustion tower. According to the heat energy recovery technology provided by Yunnan Research Institute of Chemical Industry, the inside temperature of the combustion tower calculated based on the numerical simulation can reach about 1500 °C in the case of low pressure steam production, and may exceed 1700 °C at some places (Mei et al., 2005). The measured temperature of the reaction materials in airway tube of the combustion tower can reach 650–750 °C in actual production. Based on the upgraded technology to produce medium pressure steam, the temperature in the combustion tower can reach 2200–2400 °C, and the temperature in airway tube can exceed 800 °C (Yang et al., 2005). At such high temperatures, the acidic mixture will cause extremely strong corrosiveness to the combustion tower, which can seriously affect the stability and consistency of production as well as the quality of product (Mei et al., 2005). Therefore, how to realize corrosion prevention in the combustion tower of yellow phosphorus is one of the research hotspots in the TPA industry. The key point of traditional corrosion prevention methods is to control the temperature of the inner wall of the combustion tower or combustion – hydrated reaction tower (one-step process) below 80 °C, so that the high-temperature gas containing phosphoric acid and phos-

phoric substances can be cooled down on the surface of the inner wall and therefore its corrosiveness can be reduced to realize the anti-corrosion function (Mei et al., 2005). However, this process generally requires lots of circulating water or circulating phosphoric acid to take away heat, resulting in a complex process and high energy consumption. Moreover, when the temperature of the inner wall is below 80 °C, the combustion tower cannot be used as a steam boiler to produce steam anymore, and thus heat arising from yellow phosphorus combustion cannot be recovered.

There are two patents from Japan and Germany, which mentioned that they can prevent the corrosion of the combustion tower by burning yellow phosphorus with dried air (Mei et al., 2005). However, several major problems exist in use of dried air. Firstly, the use of dried air increases energy consumption, which leads to high production cost. Secondly, an absolutely dry condition in the tower cannot be truly realized because of the limited degree of air drying as well as the moisture of yellow phosphorus, which may easily result in serious corruptions at some places with higher moisture contents. Therefore this technology cannot fully solve the corrosion problem. Moreover, plasma spraying was studied to solve this problem, but the results are far from the expectation at last (Yang et al., 2008).

Shute and Rosenhouse (1984) mentioned that a layer of metaphosphoric acid protective coating with less corrosive on the inner wall of the tower will form when the  $R_{H_2O-P_4O_{10}}$  is between 0.2 and 1. Hudson (1982) believed that a layer of ultra-phosphoric acid protective coating will form on the internal wall of the combustion tower only by keeping  $R_{H_2O-P_4O_{10}}$  between 0.6 and 0.9 for protecting the tower from corrosion. Based on the above results, Mei et al. (2005) proposed that formation of a layer of anti-corrosion protective coating ( $P_4O_{10}$  is 93%–97%) could be realized on the inner

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