Improving the efficiency of flue gas desulphurization of TPS

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Abstract. The effects of sulfur oxides on the environment and human health have negative consequences. A significant amount of sulfur oxides is emitted into the air with flue gases from the TPS. Modern Russian TPS are not equipped with sulfur-trapping units due to their significant cost. The study is devoted to the research of low-cost methods that increase the efficiency of the flue gas desulphurisation process at coal-fired TPS. Existing methods are considered from the perspective of the presence of advantages and disadvantages. The potential of the use of purge water from boilers for flue gas desulfurization of TPS.

Key word. TPP flue gases, desulfurization, seawater, coal-fired thermal power plants, benz(a)pyrene, coal, flue gases, sulfur oxides, ecology, TPP.

1. Introduction

At the present stage of development of the fuel and energy complex, the problem becomes very urgent, consumption of energy resources, but also with the results of the impact of energy facilities (in particular, coal-fired power plants) on the environment. In the conditions of constantly growing energy consumption on the background of an increase in the amount of solid fuel burned, the lack of modern technologies and equipment for flue gas cleaning and recycling of man-made wastes to be introduced to the environment becoming a serious problem.

Energy development in any country can not do without taking into account the environmental situation: both the present and projected for the future. At the same time, requirements for development activities should be developed in accordance with the recommendations of WHO and environmental organizations.

Production of heat and electricity power, combined with a mass fuel combustion, leading to an increase in environmental pollution, energy complex is one of the major sources of air pollution. Prospects for changes and increases in the fuel and energy complex should be evaluated in terms of possible harm to public health and be accompanied by a set of measures to reduce it.

With the increase in energy production and the volume of fuel burned, the problems of human health and pollution of the biosphere can be significantly aggravated. Modern TPS plants, consuming up to 20,000 tons of coal per day, instead emit a quarter of the total coal dust of industrial enterprises, two-thirds of sulfur dioxide, over 90 tons of ash and soot, over 600 tons of sulfur oxides and nitrogen,
which leads to persistent pollution of water and air basins of cities. One of the consequences of these emissions is acid rain.

Today, the majority of TPPs in Russia are equipped only with ash catchers, the remaining harmful emissions, including sulfur and nitrogen oxides, are emitted into the atmosphere without being captured, and therefore the share of emissions into the atmosphere of energy enterprises is about one fourth of the emissions of other industrial enterprises in the Russian Federation.

Abroad, beginning in the second half of the 20th century, industrial sulfur traps for flue gas cleaning are successfully introduced and operated. In the USA, Germany, Japan, Austria, the commissioning of TPS without flue gas desulfurization module is impossible. Foreign suppliers offer sets of flue gas cleaning systems, successfully implemented at foreign energy enterprises. In addition to the high cost of the installation itself, they are quite expensive to operate, while fines for emissions into the atmosphere in Russia are much lower. Implementation of projects for the installation of desulfurization facilities at existing TPPs requires the reconstruction of power units, and the construction of new stations increases the project cost by 10-20%, and therefore, Russian sewerage plants do not find use of sulfur-trapping plants, with the exception of pilot plants. [1]

2. Materials and Methods

One of the main pollutants emitted with flue gases TPPs are sulfur oxides. The emission of flue gases at thermal power plants reaches, on average, several million cubic meters per year. Sulfur dioxide adversely affects human health, inhibits the animal and plant world, increases the corrosion of metal structures, and under the action of ozone in the atmosphere oxidizes to trioxide and combines with water vapor and forms sulfuric acid vapors that precipitate with the sediments, acidifying the soil and water resources. [2]

When burning solid fuel, which is the predominant energy from the pyrite and organic sulfur dioxide formed and sulfur trioxide:

\[
S + O_2 = SO_2; \\
2FeS_2 + 5O_2 = 2FeO + 4SO_2; \\
SO_2 + \frac{1}{2} O_2 = SO_3. \]

According to G.F. Bystritsky, «... in natural types of fossil solid fuels, sulfur is found in three varieties:

1. organic SO, associated with other elements of fuel C, H, N and O in the form of complex organic compounds;
2. pyrite SC in the form of pyrite, FeS2 pyrite;
3. sulfate Sculfate in the form of sulfuric acid salts (gypsum, FeSO4, etc.).» [1]

Only the first and second types of sulfur are subject to burning, sulphate sulfur does not burn. So, G.F. Bystritsky writes: “Sulfates are oxides of sulfur, therefore the sulfur in them cannot burn. Organic and pyritic sulfur present in the fuel is burned to form toxic sulfur dioxide SO2 and (in small quantities) even more toxic sulfur dioxide SO3. Emission from combustion products causes air pollution. Organic and pyritic sulfur together form volatile combustible sulfur Sl. [1]

Flue gas desulphurisation methods, despite their diversity, can be divided into three large groups: dry, wet (regenerative), semi-dry or wet-dry (non-regenerative). These methods can be classified as follows: absorption (absorption of the removed components by the liquid), adsorption (methods based on the ability of certain solids to selectively absorb gaseous components), chemisorption (a group of methods based on the chemical interaction of gases and vapors with solid or liquid absorbents with the formation of poorly soluble chemical compounds). [2]

3. Results of modern research

The classical methods of flue gas desulfurization include various methods of using lime and milk of lime. Desulfurization methods, which are based on wet technologies, are characterized by intensive irrigation of flue gases with lime milk, which is a solution of water and lime, and the lime in this solution is divided into ions during the dissociation process. The contact of this solution with flue gases
leads to the fact that sulfur dioxide also passes into the ionic form and binds to lime ions, after which it is removed from the flue gases along with the solution.

To dry methods cleaning can be related:
- Dry additive method (chemisorption), alkaline-earth compounds (mainly limestone) are directly blown into the furnace or fed there with fuel. The method has a relatively low efficiency, processes "bergbau frschung", Germany; HOKCO, USA, etc.;
- \( \text{SO}_2 \) chemisorption with the use of copper oxide ("wop-shell" method);
- Catalytic oxidation of \( \text{SO}_2 \) to \( \text{SO}_3 \), to produce sulfuric acid as a result of the process, the «WSA» process developed by Haldor Topse Denmark;
- \( \text{SO}_2 \) adsorption using activated carbon or coke, to obtain the final product of dilute sulfuric acid or gypsum;
- Radiation-chemical cleaning of flue gases from sulfur oxides and nitrogen (radiolysis), developed by Steinmuller, Germany, also in Russia, Institute of Nuclear Physics. The flow of flue gases after the introduction of ammonia into it is irradiated by a beam of accelerated electrons. The final product is ammonium sulphate and ammonium nitrate.

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Due to the availability of this material and the possibility of using the resulting semi-finished product (gypsum) [3] in construction, methods based on the use of lime are widespread, however, are not without drawbacks. Thus, the use of dry limestone gas cleaning method, despite its simplicity and accessibility, worsens the performance of boilers and has low efficiency. Wet limestone methods can impair the operation of electrostatic precipitators and increase abrasive wear of surfaces. At the same time, the potential in the field of desulphurisation suggests a possible modernization and maintenance of existing methods, despite the shortcomings described.

One of the ways to improve the wet methods of flue gas desulphurisation is ash leaching technology, which is relevant for coal-fired power plants with a high content of calcium oxides in ash emissions. Such TPPs in irrigation water are characterized by an increase in the content of calcium ions due to the additional leaching of ash trapped in the ash collector.

Free lime is converted to bicarbonate \( \text{Ca} (\text{HCO}_3) \) with greater solubility than lime. The separation of the flow from the lixiviant is divided by means of a cyclone into condensed pulp and clarified water. Clarified water is used in scrubber irrigation to trap sulfur oxides. The pulp is discharged to the ash dump, compacting and sealing its bed and preventing the ingress of contaminated water into groundwater and water bodies. [4] Such use of a wet ash collector allows to catch about a third of the total flow of sulfur oxides and almost complete trapping of fly ash.

A more complete trapping of sulfur oxides is shown by a double alkaline technology, which allows scrubbing flue gases from sulfur oxides by 50-60% due to the use of soda \( \text{Na}_2\text{CO}_3 \) as a reagent. When using this purification technology, sulfur dioxide is neutralized by soda, resulting in the formation of sodium sulfites and sodium bisulfites, which, in turn, are treated with lime used as the second alkaline reagent. The combination of sodium sulfites and sodium bisulfites with lime forms gypsum and lye, which can be reused in the irrigation of the ash collector. [5]

Another possible way to improve the methods of wet gas cleaning is the use of boiler blowdown water that has not been subjected to a decrease in alkalii. Ph quicklime is 12, whereas the pH of the boiler water is between 7.1 and 14.0, opening up an additional way to dispose of the purge water - mixing it with a solution of water and lime. Increasing the alkalinity of the solution accelerates the binding of sulfur dioxide to it. Thus, the use of alkaline purge water boilers for irrigation of flue gases can increase the efficiency of desulfurization for any of the methods described above at minimal cost.
However, in addition to the low cost of this method, there is a danger of deterioration of the internal surfaces of the boiler from the use of alkaline water and an increase in the brittleness of the metal, therefore, this approach requires further detailed study.

A separate direction can be distinguished desulfurization of fuel.

![Figure 1. Schematic of a lixiviant.](image)

Before any burning, any significant volumes of desulfurization can be discussed only with respect to fuel oils, which are desulfurized by hydrocracking, that is, when the fuel oil is saturated with hydrogen at high temperature, about 350–430°C, and high pressure, about 10–15 MPa, as which cobalt, nickel and molybdenum sulfides can be used. Hydrogen sulfide formed in the hydrocracking process is removed from the fuel oil, being separated by the adsorption method from the residual hydrogen.

Due to the high cost of desulfurizing solid fuel, it is practically not used in modern energy, except in cases where the processing of coal is necessary. [5]

More than half of the sulfur can be removed by physical cleaning (crushing and grinding). Technologically justified use of flue gas cleaning from sulfur oxides, but two-stage cleaning (fuel and flue gases burning it) significantly increases the cost of fuel in comparison with single-stage cleaning (only flue gases).

In addition to physical cleaning, coal is cleaned in other ways:
- adding limestone during coal gasification in a fluidized bed and into the lower vortex zone of the combustion chamber of a low-temperature vortex furnace (allows you to bind up to 80% of sulfur);
- processing of coal into gaseous / liquid hydrocarbons.

However, in spite of the potential of the methods described above, their practical value is low, which is due to the high cost of the final product, which is commensurate in value with oil. [5]

Desulfurization with seawater, the use of activated carbon, and the use of zeolites can be distinguished among the less common methods of flue gas desulfurization.

In the process of using seawater, its natural alkalinity is used to remove sulfur dioxide from flue gases [6, 7]. This method compares favorably with the use of lime, firstly, the use of water at much lower temperatures (30 - 40 °C), and secondly, the lack of use of the suspension. However, seawater increases the corrosive wear of surfaces, which requires additional protection with a spray-coated flake coating or rubber lining, which increases installation costs.
Sea water is highly mineralized and contains various soluble compounds of many, including alkaline, elements with a total average concentration of about 35 g/kg sodium - 10.76 potassium - 0.38 calcium - 0.40 magnesium - 1.29 bicarbonate - 0.14, etc. The presence of the listed ions allows the use of seawater to neutralize the sulfur dioxide of flue gases. [8, p.82]

The principle of operation of desulfurization plants using seawater is similar to the principle of operation of wet limestone desulfurization, however, given the absence of reagent-preparing units and units of gypsum production, the installation cost is reduced by simplifying the technological scheme. [5, p.83]

![Figure 2. Simplified schematic diagram of desulfurization with seawater.](image)

The simplified technological scheme reduces the cost of operation, including for additional wastewater treatment, due to the fact that the pulp obtained at the outlet of the sulfur treatment plant does not require treatment before discharge into the sea, since its composition practically does not differ from the composition of the marine water. [8, p.83]

For the desulfurization of gas is often used activated carbon [8] as an adsorbent. In particular, such purification is carried out by coke oven gas, from which various sulfur compounds are removed. The technology allows to achieve 75-80% gas purification. [9]

In addition to gas purification, there is a method of dust and gas cleaning of flue gases using the «Dusseldorf system» and activated carbon.

This cleaning system belongs to the class of wet-dry technologies based on the injection of lime mortar into the gas flow to suppress SOx. After that, the stream is dedusted in the electrostatic precipitator, thereby, in addition to ash, calcium compounds are also removed from the stream. The next stage of this system is cleaning of heavy metals, chlorine and fluorine derivatives of hydrocarbons, including dioxins and furans, as well as SOx and HCl breakthroughs. The final stage of purification is the removal of nitrogen oxides in a DeNOx reactor in a moving layer of activated carbon at a temperature of 130 °C. [10]

The potential of this method is not studied sufficiently directly in the desulfurization of flue gases. It is known that the adsorption activity of activated carbon is maintained at a high level even after repeated regeneration cycles, in connection with which the desulfurization process can be cyclical in nature with regeneration, which can significantly affect resource saving. [5, p.44]

So, I. N. Shmigol notes that «in recent years, these methods have been increasingly used for cleaning air-gas mixtures from mercury vapor and organo-mercury compounds. Previously it was believed that activated carbon has a small adsorption capacity for mercury vapor and therefore it is rarely used...
in its pure form to remove mercury vapor from ventilating emissions. However, if activated carbon is pre-treated with chlorine, iodine, potassium permanganate, hydrogen sulfide or some other substances, its adsorption capacity increases dramatically, and when it comes into contact with mercury-contaminated gas, it is almost completely purified from mercury.» [8]

Also of interest is the method of using zeolites as fuel additives. This method stands out favorably among others for its versatility, allowing for complex desulfurization of flue gases, using natural zeolite-containing raw materials as a reagent. [11, 12, 13]

This technology is relatively simple in comparison with alternative ones and does not require the construction of additional technological equipment. A feature of this approach is the continuous supply of zeolites to the flameless zone of the boiler directly during its operation to absorb sulfur oxides. [14, 15]

A. G. Batukhtin and V. V. Pinigin summarize: «this technology of adsorption cleaning of flue gases with the help of natural zeolites allows simultaneously with the removal of harmful gas emissions from products of combustion of fuel to improve the economic performance of heat-generating equipment. [16] So, for example, wiping natural zeolites in the convective shaft of the boiler in the amount of 15% in relation to fuel consumption at the nominal mode of operation allows reducing the emission of sulfur oxides by 70% and increasing the efficiency of the boiler gross by 1.6% by reducing the outgoing dew temperature gases». [17, 18, 19]

4. Discussion
The selection of an effective method of desulfurization, as a rule, is guided by the following criteria:

1. Baseline data: sulfur content in fuel, volume and temperature of flue gases;
2. Factor availability and prevalence of the sorbent;
3. Disposal of the final product of desulfurization: the possibility of its industrial use or the need to transport to the dump;
4. Installation of desulfurization equipment: the amount of investment in installation and maintenance;
5. The payback period. [20]

In practice, the following ways to reduce sulfur emissions are used:

1. Removal of sulfur directly from the fuel by thermal, mechanical or biological cleaning.
2. Regime-technological measures: the use of the fluidized bed, mixing of fuel with limestone before burning, elimination of fuel with high sulfur content from the fuel balance, reduction of equipment load, gasification, etc.
3. Flue gas desulfurization: dry, wet, semi-dry (wet or mixed). [21]
However, it should be borne in mind that the use of various technological measures related to the suppression of NOx and SO2, as a result, leads to an increase in the amount of benzo (a) pyrene in the flue gases, which poses a rather serious complex problem to minimize harmful emissions.

Benz (a) pyrene - an aromatic compound, a member of the family of polycyclic hydrocarbons, formed during the combustion of hydrocarbon fuels - causes a high level of air pollution. [22]

Benz (a) pyrene is one of the most toxic combustion products belonging to the group of polycyclic aromatic hydrocarbons. It is a substance of the first class of hazard, with an extremely high level of dangerous effects on the environment and human health (carcinogenicity and blastomogenicity are noted upon contact with benzo (a) pyrene). At the same time, the mechanism of the occurrence of polycyclic aromatic hydrocarbons during fuel combustion has not been sufficiently studied today. [23]

In addition to the special danger, benzo (a) pyrene is extremely widespread, chemically and thermally stable, and when it enters the body it can accumulate. [24]

The relationship between the decrease in the sulfur content in the fuel and the increase in Benzo (a) pyrene emissions is most pronounced. An example of this relationship for the BKZ-220 boiler with various sulfur contents in the fuel is shown in figure.

![Figure 4. The dependence of emissions of benzo (a) pyrene on the sulfur content in the fuel.](image)

5. Conclusion

Thus, the relationship between a decrease in the content of sulfur oxides and nitrogen and an increase in the content of benzo(a) pyrene poses a rather serious complex problem of minimizing harmful emissions in exhaust gases, among which solutions you can specify:

1. Introduction of energy-saving technologies for heat consumers (including control and metering devices);
2. Reduction of heat loss in networks;
3. Optimization of boiler operation modes; [25]
4. Improving the efficiency of fly ash trapping (including fine fractions).

In practice, an extremely unfavorable ecological situation has developed in Chita today. So, according to the results of the first half of 2018, Chita entered the list of Russian cities with the most polluted air. In addition to Chita, Petrovsk-Zabaykalsky was included in this list. Information about this was published on the website of Rosstat. Based on these data, 8 facts of air pollution were recorded, with benzo (a) pyrene exceeding the maximum permissible concentration by 57 times. [6]

The President of the Russian Federation, V. V. Putin, has set the task of reducing pollutant emissions by 20% by 2024. This task has been set for 12 Russian cities, including Chita. The main air
emissions in the city of Chita are accounted for by energy enterprises (CHP-1, CHP-2, numerous boiler houses), a dense stream of city transport and seasonal forest fires. [2]

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