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Thermal Power Plants - Flue Gas Purification

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Abstract: One of the most important problems today is the formation of greenhouse gases, mainly caused by the emission of flue gases in thermal power plants: carbon dioxide (CO_2), sulfur dioxide (SO_2) and nitrogen oxides (NOx) ("acid rain"). CO_2 emissions are inevitable and related to the fuel combustion process (carbon capture techniques). Sulfur dioxide emissions are reduced by cleaning coal and by applying wet and dry flue gas desulfurization. The injection of absorbents can be applied in the combustion chamber or in the flue gases. Nitrogen oxide emissions are reduced by proper fuel selection and advanced solutions in the construction of the combustion chamber. Burners with low NO*x* emissions are used.

Keywords: environmental aspect; flue gases; greenhouse gases; reducing emissions

1 INTRODUCTION

Since the beginning of the 19th century, with the first industrial revolution and the introduction of the steam engine, the concentration of greenhouse gases in the atmosphere has been continuously increasing. This phenomenon has been particularly significant in the last 50 years [1]. When talking about greenhouse gases, carbon dioxide (CO_2), methane (CH_4) and nitrogen oxides (N_2O) are primarily investigated. Greenhouse gases are mainly produced by the conversion [2] and use of energy in buildings consumption [3], transport [4-5], production process [6] and different types of heating systems [7]. During this process as much as 85 % of carbon dioxide is produced. Greenhouse gases result in global warming, the occurrence of bad weather conditions in certain areas, and then indirectly affect human life and health, ecology, water supply and crops, [8]. In order to slow down the impact of global warming, it is necessary to reduce energy generation based on fossil fuels, which also reduces the share of energy obtained through the combustion process of primary energy, to increase the efficiency of energy conversion, to improve fuel quality and conversion technology to reduce emissions (e.g. coal gasification), and also to use technologies for the separation and storage of emitted gases, especially carbon dioxide. So, there is a growing need to reduce harmful emissions from flue gases that directly affect the ecosystem (greenhouse gases and acid rain). Harmful emissions include emissions of sulfur dioxide, nitrogen oxides and particulate matter. There are methods for reducing harmful emissions from flue gases that have been in conventional use for years, but there are also several methods that are still in the development process. This paper deals with the methods used to reduce sulfur dioxide emissions, such as: coal cleaning, injection of absorbents into the flue gas duct, injection of absorbents into the firebox and also wet and dry flue gas desulfurization and methods for reducing nitrogen oxide emissions.

In the second chapter, the authors provide an overview of the legislation in the world and in the Republic of Croatia and references to the literature used in the work. Chapter 3 describes technologies for reducing sulfur dioxide emissions based on coal cleaning and flue gas desulfurization. Chapter 4 discusses technologies for reducing nitrogen oxide emissions based on primary steps such as changes in the combustion method, ways of influencing the reduction of nitrogen oxide emissions during boiler construction, and the use of selective non-catalytic and selective catalytic reduction. Chapter 5 generally considers combined techniques for simultaneously reducing emissions of both sulfur and nitrogen oxides in flue gases. Finally, the conclusion provides an overview of the effectiveness of individual methods.

2 LEGISLATION DOCUMENTS

The Kyoto Protocol and the United Nations Framework Convention on Climate Change is an addendum to the international agreement on climate change. The protocol is a complex agreement with significant economic implications that obliges developed countries to reduce greenhouse gas emissions by an average of 5 % below the 1990 level in the first period until 2012. The protocol was opened for signature on December 11, 1997, in the Japanese city of Kyoto organized by the United Nations Framework Convention on Climate Change (UNFCCC). According to the obligations arising from the Kyoto Protocol, countries are divided into two groups according to their industrial development: developed and underdeveloped. And while the highly industrialized countries, grouped in the so-called Annex 1, must reduce the emission of harmful gases by 5-10 %, countries outside that group do not have such an obligation, but must therefore participate in the Clean Development Program. Kyoto Protocol entry into force (ratification by at least 55 members of the Framework Convention, which must account for at least 55 % of all greenhouse gas emissions in industrialized countries) on February 16, 2005. The Kyoto Protocol shows the clear commitment of most countries to take long-term measures that will initiate the resolution of the problem, [9-11].

Further progress was achieved in 2010 at United Nations Climate Change Conference Cancun, Mexico and after confirmed by Agreement in Paris, France 2015. A political agreement has been reached to a key step to achieve the goal of reducing greenhouse gases and helping developing countries from climate impacts with the "right to development". It was the world's largest joint effort to reduce emissions, with national plans set at the international level through the UNFCCC. Also, the most complex package of agreed government measures to support developing countries: financial, technological and support in capacity building to combat the consequences of climate change.

Standards for the emission of harmful substances in Republic of Croatia are defined by the "Air Protection Act". It determines the measures, the way of organizing and implementing protection, improving air quality, and also prescribes air quality monitoring. There are also 2 subordinate legislations: Regulation on recommended and limit values of air quality and Regulation on limit values of emissions from stationary sources. Recommended and limit values of air quality represent only values for determining air quality categories, and not maximum permitted quantities of pollutants. The Regulation on limit values of emissions from stationary sources prescribes maximum permitted emissions and the method of adapting existing facilities to new regulations. Emission limit values for SO_2 are determined by the Regulation on limit values of emissions from stationary sources in Croatia. Regulations on SO_2 emissions for large combustion plants are dependable on the age and size of the power plant. Stricter rules and regulations apply to large plants, as well as to new plants compared to existing ones. Large burning devices can operate without a waste gas desulphurization device for a maximum of 240 hours a year, of which 72 hours continuously. Emission limit values for NO_x are also determined by the Regulation on emission limit values from stationary sources in Croatia. When determining the limit of NO_x emissions for gas turbines, it is necessary to make a correction with regard to the degree of operation of the plant by taking the value from defined maximum value multiply by the factor $\eta/30$ (where η is the percentage of heat supplied by the fuel that is converted into useful mechanical work in %). The degree of action η is determined for the most unfavorable mode of operation of the turbine when η is the smallest.

For determining the composition of flue gases, norms and standards are prescribed, which describe the measuring devices used for such measurements. The composition of combustion flue gases is measured in accordance with the HRN EN ISO 10396 standard, which describes the measurement methods and procedures.

3 TECHNOLOGIES FOR REDUCING SULFUR DIOXIDE EMISSIONS

Sulfur dioxide emissions depend on the amount of sulfur in the fuel. Natural gas contains less than 0.1 % sulfur (mainly in the form of H_2S) and emissions are relatively low. Light fuel oils, such as those used in gas turbine plants, usually contain little sulfur and emissions are also low. Coal and heavy oils (fuel oil) can contain a lot of sulfur and therefore combustion produces large emissions of sulfur oxides. Methods for reducing sulfur dioxide emissions are briefly described below.

3.1 Coal Cleaning

Sulfur in coal is of organic and inorganic origin. The origin of organic sulfur is related to plant matter (especially proteins). Inorganic sulfur is divided into pyritic and sulfate sulfur, of which pyritic sulfur makes up a significant portion, so when talking about pyrite, inorganic sulfur in coal is usually meant in general. Initially, the main task of coal cleaning was to remove ash and moisture, in order to reduce transportation costs and increase the efficiency of the power plant. In the United States, the Clean Air Act was adopted in 1970, which obliged new coal-fired power plants to significantly reduce sulfur dioxide emissions. Since then, a number of procedures and methods have been developed with the aim of achieving the best possible results in this field. Thus, in the 1970s, the potential emission of sulfur dioxide in new power plants was reduced to 80 %, by the 1990s, standards prescribed a 90 % reduction in emissions, and today the efficiency of such systems is as high as 99 %, [8]. Recently, the focus has shifted to the removal of sulfur from coal, due to the growing problem of acid rain caused by sulfur dioxide emissions from the coal combustion process. Since current coal extraction methods are non-selective, they result in a large amount of impurities in the coal. Physical cleaning of coal, which removes ash and some of the pyrites, is a method that has been used for many years.

Conventional cleaning methods are mostly based on gravitational separation of ash and sulfur components before the coal is converted into coal dust and introduced into the boiler. It is important to note that this method also significantly reduces the particulate content in the flue gases. Conventional cleaning usually begins with crushing the coal into pieces with a diameter of less than 50 mm, followed by separation into coarse, medium and fine particles. The crushing releases the materials that form the ash and inorganically bound sulfur (e.g. pyrites, FeS_2). The smaller the particles, the better the separation. Since mineral substances have a higher density than coal particles, they can be removed from coarse and medium particles by physical cleaning methods. Separation of fine particles can be done by flotation, using the surface difference between coal and ash, [2].

3.2 Flue Gas Desulfurization

SO_2 emission reduction by flue gas desulfurization can be divided into two basic processes: regeneration and non-regeneration processes. In regeneration processes, SO_2 is obtained, which can be further used to produce sulfuric acid, elemental sulfur, or liquid SO_2. Such devices are in the minority because they are significantly more complex and expensive. In non-regeneration processes, a by-product is obtained that is either permanently disposed of or used as a raw material in the cement or construction industry. Both processes are further divided into wet and dry processes. From wet to dry processes, the efficiency of desulfurization and the price of the device decrease. The costs of flue gas desulfurization plants depend on market and other conventional conditions. In addition, the price also depends on technical factors such as the amount of flue gases, the concentration of SO_2 in the flue gases, the required degree of desulfurization, environmental restrictions, and the amount of wastewater, [12].

3.2.1 Wet Process of Flue Gas Desulfurization

The flue gases enter a large vessel and an aqueous solution containing 10 % lime or limestone is injected into it. The calcium in the solution reacts with SO_2 to form calcium sulphate (gypsum) or sulphite. Part of the solution goes into a settling vessel where the solids settle before going to a filter where the water is removed and 50 % of the solids remain. The waste calcium sulphite is mixed with ash (1:1 ratio) and disposed of in landfills. Mist eliminators are also installed to collect the solution and remove moisture from the flue gases. They are located at the outlet of the firebox. After leaving the particle removal plant, the gases enter a spray tower or absorber. An aqueous calcium-based solution is injected here, which forms calcium sulphite or calcium sulphate with SO_2, which is removed by releasing water and settling in a designated vessel. The resulting waste is usually mixed with ash collected in a filter or electrostatic precipitator and with lime in a mixer, and disposed of in a landfill, [13]. The advantages of wet desulfurization are good efficiency and additional particle removal. The disadvantages are scale deposition and a tendency to clog, a significant drop in flue gas pressure, and significant investment and operating costs. In terms of efficiency, the standard version allows for sulfur removal of between 80 % and 90 %. Additives (magnesium-enriched lime) increase the efficiency by 5 % to 10 %, ultimately reaching 95 % to 99 %. Installation of this technology on an existing plant takes 3 to 6 weeks, provided there is sufficient space, [2].

3.2.2 Dry Process of Flue Gas Desulfurization

Dry process of flue gas desulphurisation is carried out by feeding a solution of calcium hydroxide (lime mixed with water) into a spray tower. The solution is sprayed and injected into the flue gases. As they evaporate, the droplets react with the SO_2 in the vessel. The amount of water is chosen so that all the water must evaporate before the absorbent falls to the bottom of the vessel, which is why this process is called a dry process. The by-product that is formed is collected at the bottom of the drying tower and in particle removal equipment. The by-product is unreacted lime and fly ash, and is removed by a filter or electrostatic precipitator. In the case of coal with a low sulphur content, 70 % of the sulphur dioxide is removed. Investments in the dry desulphurisation method are lower than investments in the wet desulphurisation method, and they are also simpler to operate and maintain. It takes 3 - 6 weeks to install a dry flue gas desulphurisation unit in existing plants. The advantages of this method are simplicity and lower operating and investment costs, while its only disadvantage is lower efficiency (approx. 70 %), [14].

3.2.3 Injection of Absorbent into the Combustion Chamber

When injecting into the combustion chamber, the absorbent (dry powdery material) is injected together with air above the combustion area through special injection inlets. Lime, which reacts with SO_2 and forms calcium sulfate (gypsum), is separated from the absorbent in the combustion chamber by calcination. The absorbent can be limestone or lime. Calcium sulfate (gypsum) and fly ash are removed using an electrostatic precipitator or bag filters. SO_2 removal efficiency is 30 – 60 %. Some other difficulties that arise with this method must be solved, for example, the efficiency of the absorbent must be increased to make the method more economically attractive. Another problem is that the injection of the absorbent has a negative effect on the operation of the clarifier. This method requires less investment than wet and dry gas desulfurization, requires less space and is simpler to operate and maintain.

3.2.4 Injecting the Absorbent into the Flue Gas Duct

Injecting the absorbent into the flue gas duct is injection in front of the precipitator, similar to injection in the combustion chamber. Similar equipment is required. A reaction vessel (absorber) is required as well as particle removal equipment (electrostatic precipitator, bag filter). Calcium hydroxide is injected into the reaction vessel, where it mixes with the flue gases. As the gases enter the absorber, they are cooled to the "approach temperature" by injecting water. Unreacted absorbent and ash are captured in a factory filter located behind the absorber and disposed of in a landfill, while the clean flue gases are discharged into the stack. Injection in front of the electrostatic precipitator can remove 30 – 70 % of SO_2, and injection after the precipitator can remove 80 – 90 %. The efficiency also depends on the amount of absorbent and the approach temperature.

4 TECHNOLOGIES FOR REDUCING THE EMISSION OF NITROGEN OXIDES (NO_x)

Emissions of nitrogen oxides are associated with acid rain and photochemical smog and ozone in the troposphere. For this reason, technologies were introduced to regulate NO_x emissions from new, as well as from old, power generation plants. NO_x emissions can be reduced in the following ways:

- primary steps (change in combustion)
- choice of fuel
- construction of the combustion chamber.

4.1 Primary Steps (Change in Combustion)

These are changes in the boiler, or changes in combustion, and include: changes in operating conditions and changes in combustion devices. Changes in operating conditions include reducing the amount of combustion air, reducing the temperature of the combustion air, reducing the amount of heated air and reducing the load on the combustion chamber. Air reduction is a frequently used method for reducing NO_x emissions, but it is also used as a method for reducing energy consumption. A significant reduction in nitrogen oxide emissions can be achieved by primary steps during combustion, which is achieved by appropriate burner design and stepwise air and fuel supply. This reduces the maximum temperatures in the flame core and reduces the oxygen concentration in the combustion zone. The amount of NO_x emissions into the atmosphere can be reduced to a value of less than 300 mg/m^3 (up to 40 %), [14, 15]. The method of reducing the amount of air has a number of advantages:

reduction of flue gas heat losses and reduction of fuel consumption (both of which result in higher energy efficiency), no need for additional investments and reduction of low-temperature corrosion, but also disadvantages: requirements for precise combustion control, possible increased particle emissions and more intense deposition on the inner walls, which reduces heat transfer and increases maintenance costs. In this case, the power plant is out of use more often and for longer. The method of reducing the temperature of the combustion air has its advantages: simplicity of implementation and no need for investments, but also disadvantages: reduced energy efficiency and increased fuel consumption. The situation is similar with the primary step of reducing nitrogen oxide emissions by reducing the load on the combustion chamber, which is simple and without additional investments (advantages), but at the same time the available power is reduced and the specific fuel cost is increased (disadvantages), [16].

4.2 Low Emission NO*x* Burners

Burners, by their design, have a significant impact on NO*x* formation. Modifications to the burners regulate the combustion of carbon and hydrogen with acceptable NO*x* formation. The basic ways of improving burners are:

- burner with improved mixing
- burner with flame splitting
- self-recirculating burner
- burner with substoichiometric combustion
- burner with two-stage combustion.

4.3 Selective Non-Catalytic Reduction (SNCR)

SNRC is a chemical process that converts nitrogen oxides NO*x* into molecular nitrogen N_2. In this process, there is no catalyst, and the reducing agent (ammonia, amides) is injected directly into the flue gas in the high-temperature zone. This chemical reaction occurs at high temperatures (870 - 1200 °C). In this process, the temperature must be carefully controlled. A gaseous or liquid nitrogen-based reactant is injected and mixed with the hot flue gases. The reactant reacts with the NO*x* in the flue gas stream and converts it into atmospheric nitrogen and water vapor, which are harmless. The reactant primarily reacts with NO*x*, not with oxygen or other components of the flue gases. This technology does not produce any solid or liquid waste. In this way, NO*x* emissions can be reduced by 35 - 70 % without affecting the operation of the plant. At temperatures below optimal, the reaction is weak, so the reduction of NO*x* is very small, almost negligible. The technology was first developed for fuel oil and natural gas boilers, and is still in the testing phase for coal-fired furnaces. Due to the very simple components that need to be installed, installing this technology is much easier than others. Installation in existing plants takes 2 - 5 weeks, [2].

4.4 Selective Catalytic Reduction (SCR)

It is widely used for low-sulfur coal, but it is very expensive and there is a need to adapt to different types of coal. The working principle of this method is based on the reduction of NO*x* by converting it to N_2 and H_2O by the reaction of NO*x* and ammonia with the help of a catalyst. The primary reaction that occurs in this method requires oxygen, and the catalyst works best at oxygen levels above 2-3 %. This technology is based on the injection of ammonia into the flue gases to convert NO*x* into elemental nitrogen and water. It works in almost the same way as selective non-catalytic reduction. The only difference is that selective catalytic reduction requires a reagent that accelerates the chemical reaction. The SCR system contains equipment for measuring NO*x*, which is necessary for precise dosing of the reagent, so that ammonia is not released into the environment. The catalyst is needed because the process takes place at significantly lower temperatures than in selective non-catalytic reduction. Typical temperatures for this process are 340 - 380 °C. The most common catalysts are a combination of vanadium and titanium and zeolite materials. This technology was first used in Japan in the mid-1970s, in Western Europe in the 1980s in coal-fired power plants, and in the USA in the early 1990s also in coal-fired power plants. The installation of a hot system on an existing plant takes 2 - 3 months, and for a cold system 3 - 6 weeks. The efficiency of this method is 70 – 90 %, [14].

5 TECHNOLOGIES FOR THE COMBINED REMOVAL OF SO_2 AND NO*x*

Sulfur dioxide SO_2 and nitrogen oxides NOx are present in flue gases. It is desirable that the same process be used to reduce SO_2 and NO*x* emissions. In practice, separate methods are used for control. Conventional flue gas desulfurization methods are used to reduce SO_2 emissions, while selective catalytic reduction is used to reduce NO*x* emissions. Combined systems should operate in such a way that their SO_2 and NO*x* removal efficiency is equal, [13]. The emission limit values for new coal-fired plants with a capacity of >100 MW for the EU (including the Republic of Croatia) for sulfur and nitrogen oxides are 200 mg/m^3, [15].

6 CONCLUSION

The operation of a thermal power plant also produces sulfur dioxide SO_2, and one of the measures to reduce the production of this gas is the use of natural gas as a primary energy source, which has an almost negligible amount of sulfur (gas-fired thermal power plants have SO_2 emissions of 0,02 - 0,42 g/kWh, liquid fuel thermal power plants have 3,77 - 4,61 g/kWh, and coal-fired thermal power plants have 0,68 - 24,51 g/kWh). Furthermore, the reduction of SO_2 emissions is also achieved by cleaning coal with an efficiency of 60 %. After the coal cleaning process, the percentage of coal is 60 - 90 % of the initial mass, and the percentage of heating value is 85 - 98 %. The wet desulfurization method achieves an efficiency of 80 - 90 % in removing sulfur. By adding lime, the efficiency of SO_2 removal rises to 95 - 99 %. The dry desulfurization method is used in coal-fired thermal power plants with a low sulfur content in the coal itself. The efficiency of the method is 70 - 90 %. This method is the most investment-friendly and easiest to install and maintain. In order to reduce the emission of nitrogen oxides NOx, burners with low NO*x* emissions are used (e.g. flame splitting burners

- emission reduction of 20 - 40 %, used in large liquid fuel plants; self-recirculating burner - efficiency of 20 %; burner with two-stage reduction - efficiency of 30 - 55 %). In general, the method of using low NOx burners achieves an efficiency of reducing NO*x* emissions by 40 - 50 %. Selective non-catalytic reduction achieves NO*x* removal of 35 - 70 %, and is used for fuel oil and natural gas. The selective catalytic method is used for coal with low sulfur emissions, the efficiency is 70 - 90 %, but this method is more expensive than the non-catalytic method with installation time in the range of 2 - 3 months, while for non-catalytic reduction this time is 2 - 5 weeks. During the installation, the plant does not work, and thus additional costs accumulate.

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