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USE OF AMMONIA SEMIDRY TECHNOLOGY FOR FLUE GAS DESULFURIZATION IN COAL POWER PLANTS

The environmental protection legislation of Ukraine and the European Union requires a significant reduction in sulfur dioxide emissions at thermal power plants. Therefore, the need to design, to produce, and to build an installation for flue gas desulfurization in compliance with European best practice. The necessary condition is to determine optimal parameters for variable modes of purifying installation. The article presents the results of numerical modeling of semidry desulfurization plant to bind sulfur dioxide in flue gases coming from the boiler TPP-210A of 300 MW power unit. The desulfurization plant works on the semidry method using ammonia water as sorbent. Upon the results of research, the size of reactor operating area as main part of desulfurization plant has been established, the method for feeding ammonia and water used to prepare the solution and to irrigate flue gas flow has been defined. The consumption rate of ammonia solution and water has been estimated depending on volume of flue gas and water and inlet temperature of gases.

Keywords: desulfurization, ammonia, chemical reactor, and power unit.

Ukraine's thermal power plants (TPPs) are among the largest sources of atmospheric air pollution by sulfur dioxide as a result of significant emissions from combustion of several dozen million tons of sulfur-containing coal annually [1]. The environment protection legislation of Ukraine and the European Union requires a significant reduction of sulfur dioxide emissions which are the main pollutants [2, 3]. In Ukraine, 42 power units with a capacity of 300 MW each have been installed at the TPP [1]. EU Directive 2010/75/EU on industrial emissions establishes that for such boiler units, sulfur dioxide concentration in flue gases shall not exceed 200 mg/Nm^3 and 400 mg/Nm^3 in the case of high-sulfur coal combustion, provided the efficiency of sulfur purification plant is, at least, 95% [3].

At present, there are no desulfurization plants at the Ukrainian TPPs. An urgent condition for Ukraine's membership in the Energy Community is the construction of plants with an effective technology for sulfur dioxide retention. Currently, there have been many commercial engineering solutions for removing SO_2 from waste gases generated by combustion of fossil fuels [4]. Chemical binding of sulfur dioxide is possible with reagents containing calcium Ca (lime, limestone), magnesium Mg, sodium Na, manganese Mn, ammonia NH_3, etc. [5]. An important factor for selecting the purification technique is rate and conditions of the reagent dissolution, which significantly affect the SO_2 removal efficiency and the size of chemical reactor where the binding of sulfur dioxide occurs. The semi-dry ammonia desulphurization method is one of advanced technologies [6].

The simulation of SO_2 removal is based on TPP power unit with an electric power of 300 MW, which has a twin boiler unit TPP-210A where anthracite is burned. The purpose of mathematical research is to obtain the data necessary for developing the terms of reference for the design of flue gas desulphurization plant and technical specifications.

During the study, it is necessary to determine the size of sulfur purification plant (chemical reactor) work area, the way of supplying the fluid (solution of reagent and irrigation water) to the reactor, the type of liquid injectors, and the regime parameters: solution and water consumption, velocity and temperature of gases in the reactor work area depending on the power unit load and meteorological conditions. The data are obtained by the calculation method based on original mathematical model [7]. The model takes into account the absorption of gases by liquid, the evaporation of water from droplets, protolytic reactions in the liquid phase and the combination reaction in the gaseous phase of the stream. This way, it enables to determine the concentration of chemical compounds in both phases.

RESULTS AND DISCUSSION

The semi-dry ammonia technology is proposed for binding of sulfur dioxide of flue gases. It will enable, firstly, to get an output product, powdered ammonium sulfate $(NH_4)_2SO_4$ that can be used as mineral nitrogen fertilizer. Secondly, this method makes it possible to exclude the sorbent dissolution in suspension, i.e. to eliminate the process affecting the plant size and efficiency, since the rate of calcium sorbent dissolution is low. Thirdly, the method enables to avoid the formation of solid deposits in nozzles spraying the liquid and on the reactor surface.

In the gas path of the coal power unit, the desulphurization plant is located after the ash removal, for example, an electrostatic precipitator (ESP). To capture fine solid particles of the by-product resulting from SO_2 chemical binding, a sleeve filter is installed behind the reactor. Chemical binding of sulfur dioxide occurs on the sleeve surface as well [5]. Thus, the sleeve filter is the second level of sulfur purification. Depending on the sleeve filter design and the frequency of shaking the alkaline dust accumulated on the sleeves, the additional efficiency of sulfur dioxide capture varies within 5—20% [5].

One body of TPP-210A steam generator, at nominal load, after the ESP, generates flue gases at a flow rate of 180.8 Nm^3/s. At 150 °C, it makes up 280.2 m^3/s or 1.008 million m^3/h. The reagent consumption is determined by sulfur dioxide content in flue gases. To ensure an efficiency of, at least, 90% at a SO_2 concentration of 1181 ppm, the specific consumption of NH_3 should be 1.615 g/Nm^3. The work reagent solution is prepared from 25% aqueous ammonia and service water. The total water consumption should provide the maximum possible process efficiency and be such as to ensure that after drying of liquid droplets, the temperature of exhaust gases at the outlet exceeds the water dew-point temperature, at least, by 15 °C to prevent chemical corrosion of ducts [4]. In this case, the total water consumption is 51.611 g/Nm^3. For such consumption rates of NH_3 and H_2O, the ammonia content in the solution will be about 3%.

The work area of the designed chemical reactor is shaped as cylinder. The reactor inlet and outlet are shaped as a diffuser and a confuser, respectively. The flue gases are fed from the lower part, and the purified gases leave the reactor in the upper part. In the lower part of the work zone, ammonia solution is injected into the gas stream through the nozzles.

The diameter of chemical reactor work area is determined by the area of cross section which the average velocity of flue gas flow depends on. The stay of gases in the work area depends on their velocity, while water evaporation rate and duration of droplet drying depend on the velocity of liquid droplets with respect to the gas velocity. Calculations have showed that if the work area diameter of chemical reactor increases from 7 to 11 m, the gas velocity ranges within 2.9—7.3 m/s (Fig. 1) and 2.5—6.1 m/s at the entrance to the area and at the exit from it, respectively. The velocity of flue gases has been established to have a small effect on the efficiency of sulfur dioxide binding. This is explained by very low relative velocity of droplets at these gas velocities. In 200 ms after the liquid is injected into the stream, the droplet velocity becomes almost equal to the gas velocity. Consequently, there is no convective component of water molecule transport from the drop boundary layer to gas. Thus, the water molecules are transported from drop to gas only by diffusion mechanism. In the case of single-stream method, a drop of 100 μm is dried for about 5 seconds regardless of the velocity, within the abovementioned ranges. The height of the reactor work area is determined by time of droplet drying. Under these conditions, the required height of the area varies from 33.1 m to 13.5 m, depending on the area diameter. For this type of plants (for example, scrubbers) the ratio of height to diameter varies from 4 to 6. Based on the results of numerical studies, it is assumed that the work area internal diameter is 9 m and the height is 40 m.

The research [8] presents the results of a numerical study of processes in an industrial sulfur purification plant, the prototype of which is the plant used for a group of coal boilers having a thermal capacity of 50 MW, at the Lublin TPP, Poland. The plant is designed for a lower consumption of flue gases with a lower sulfur dioxide content as compared with the plant under consideration. The present research continues the study [8]. In particular, the plant for sulfur dioxide removal from flue gases coming from the boiler that is a part of 300 MW unit of Ukrainian TPP with a higher gas consumption and higher SO_2 content has been considered. In addition, more parameters that affect the performance of the chemical reactor have been identified. The results of numerical studies [8] have showed that at the nominal mode, the single-stream method of supplying liquid (solution), when it is all fed at the entrance to the reactor work area, is less effective than the double-stream one, when a weak reagent solution and processing water are fed separately. With the double-stream method, the solution and water can be fed in various sequence. In the case of water-solution method, the water is fed first, with the solution following it. In the case of solution-water, on the contrary, the solution is introduced first, with the water fed after it. It should be noted that in the second case, the efficiency is higher than in the first one.

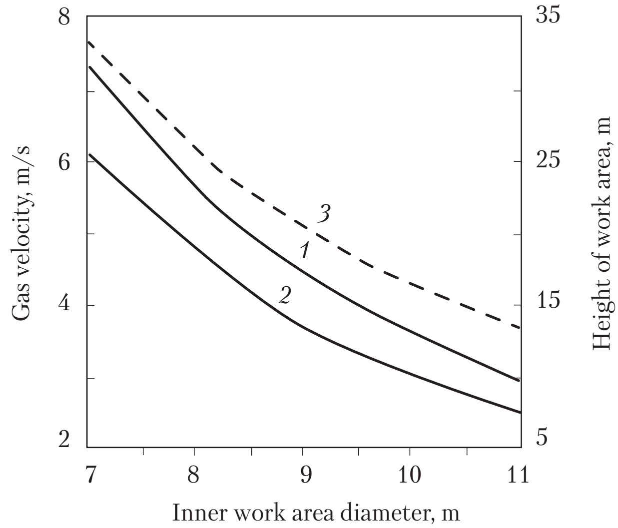

Fig. 1. Effect of work area diameter on gas velocity at the inlet (*1*), gas velocity at the outlet (*2*), and height of work area (*3*)

This effect is explained by the following factors. If the irrigation water is supplied first, then droplets absorb SO_2 without chemical binding in the absence of a reagent. In addition, as the mass of droplets decreases due to water evaporation, sulfur dioxide returns to the gaseous phase of the stream. The further introduction of ammonia solution drops to a partially cooled gaseous phase leads to a lower emission of NH_3 gas from the solution to the gaseous medium. Therefore, SO_2 is absorbed and chemically bound only by drops of ammonia solution, but in this case there is a risk of ammonia gas emissions at the exit from the reactor.

After the sorbent solution drops are fed, sulfur dioxide is chemically bound with ammonium hydroxide NH_4OH and ammonia releases from the solution droplets to the gaseous phase, in accordance with the Raoul law [9]. Further, as irrigation water drops are injected they absorb not only SO_2 but also NH_3. As a result, the chemical bind-

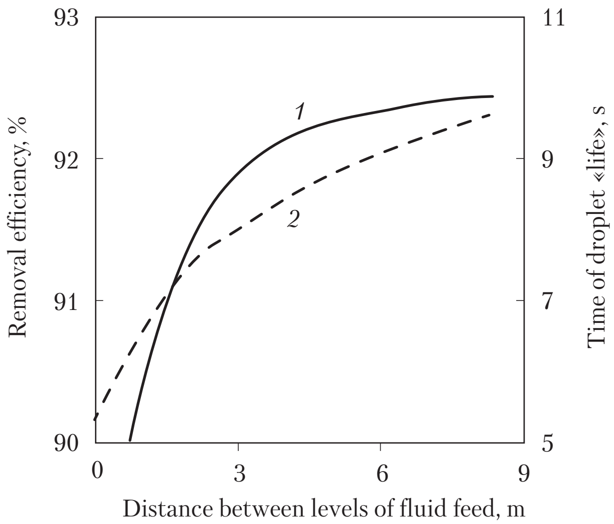

Fig. 2. Effect of distance between levels of fluid feed on effectiveness of reagent binding (*1*) and time of droplet «life» (*2*)

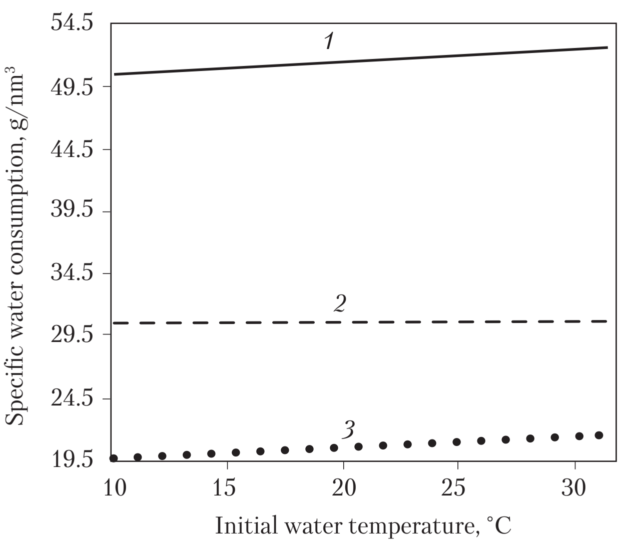

Fig. 3. Water temperature effect on water consumption: *1* — total, *2* — solution, *3* — processing water

ing of sulfur dioxide occurs in droplets. Having simulated the proposed chemical reactor, it is found that for the single-stream method the effectiveness of SO_2 binding is about 76%, for the water-solution method it is 81%, and for the solution-water method it can reach over 92 %.

Fig. 2 shows that in the solution-water method, the effectiveness of sulfur dioxide binding is influenced by the distance between the levels (along the reactor height) of nozzles for injection of the reagent solution and the irrigation water. As the distance decreases, so does the effectiveness. This is explained by a reduction in the time of water evaporation from droplets, i.e. in the time of their existence. Thus, the «life» of drops decreases by almost 45%, with the effectives falling by 4%. At the beginning, it decreases slowly, but as overlapping of solution droplets and water ones increases it goes down sharply. It is also established that at the nominal mode of the plant, the optimum distance between the levels of liquid supply is 8.3 m. In this case, 60% of water is introduced into the reactor with ammonia solution and 40% with irrigation water. Further increase in distance does not lead to increase in effectiveness. It should be noted that in the double-stream method, the initial mass content of ammonia in the solution should be about 5%.

Seasonal fluctuations in atmospheric air temperature (winter-summer) lead to changes in temperature of service water and flue gases. Calculations for the chemical reactor operating modes with double-stream fluid supply using the «solution-water» method have showed that varying initial temperature of service water within the range of 10—30 °C almost does not affect the plant effectiveness that is approximately equal to 92.4%. The weak effect is explained by the fact that the mass consumption of water is 27 times less than the gas consumption, although the water heat capacity is 4 times higher than that of flue gases. Water consumption should be raised as its temperature goes up (Fig. 3) in order to compensate growing evaporation rate of water from droplets. It is enough to increase only the consumption of irrigation water, while that of 5% ammonia solution should remain constant.

Similar change in effectiveness has been reported when the initial temperature of flue gases varies within the range 130—170 °C. At these temperatures, the plant effectiveness is approximately equal to 92.5 ± 0.2%. In addition, higher values correspond to lower temperatures. How-

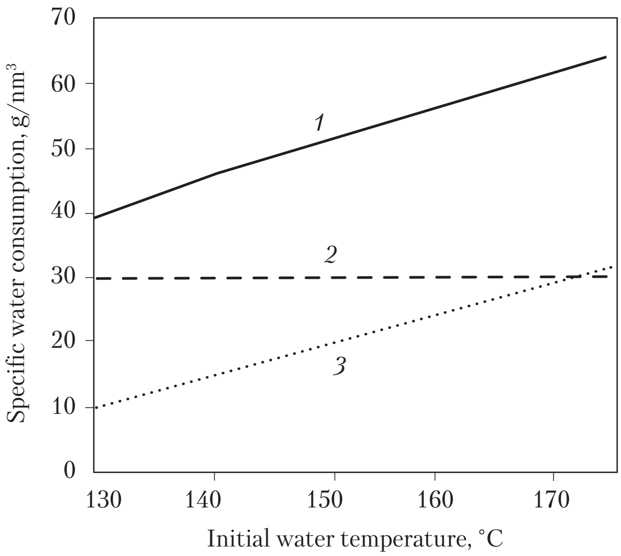

Fig. 4. Gas temperature effect on water consumption: *1* — total, *2* — solution, *3* — processing water

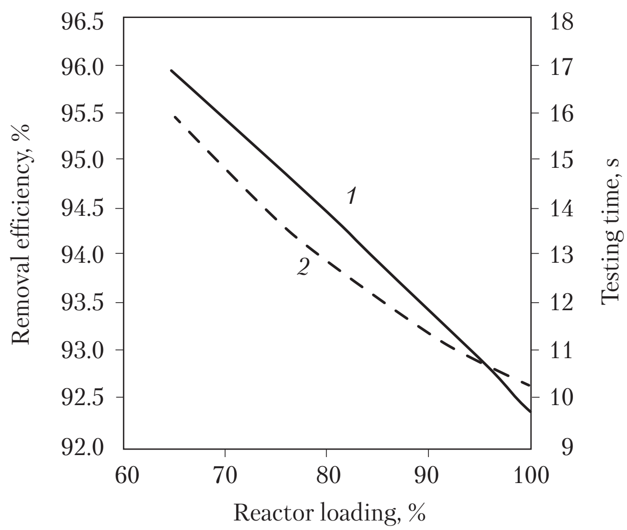

Fig. 5. Load impact on effectiveness of sulfur dioxide binding (*1*) and gas stay in work area (*2*)

ever, the specific consumption of irrigation water needs to be significantly raised as gas temperature increases (Fig. 4) to compensate growing evaporation rate of water from droplets when the heat flux introduced by gases into the chemical reactor increases. Table 1 shows water consumption at different initial gas temperatures.

During the operation of steam generator its load may deviate from the nominal one, as a result of which the volume of flue gases changes proportionally. The calculations have showed that if gas consumption decreases by 35% the effectiveness of sulfur dioxide binding rises from 92.4% to 96% or by 3.6% (Fig. 5). The gas stay in the work area increases almost 1.5 times. The initial total specific water consumption remains constant and equals to 51.611 g/Nm^3, but the absolute solution and irrigation water consumption in kg/s should decrease proportionally to load reduction. Table 2 shows the solution and water consumption at different loads of the chemical reactor. As showed above, during the operation of chemical reactor, there is a need to change the solution and/or irrigation water consumption. In the case of lower boiler load, a part of nozzles injecting the fluid into gas stream can be turned off.

Table 1

Fluid Consumption at Different Gas Temperatures

Parameter	Value				
Gas temperature, °C	130	140	150	160	170
25% NH_3 solution, kg/s	1.168				
Water for solution, kg/s	4.674				
5% NH_3 solution, kg/s	5.842				
Processing water, kg/s	1.553	2.8010	3.784	4.752	5.736
Water consumption, kg/s	6.227	7.475	8.458	9.426	10.410
Water consumption, t/h	22.415	26.909	30.447	33.933	37.473

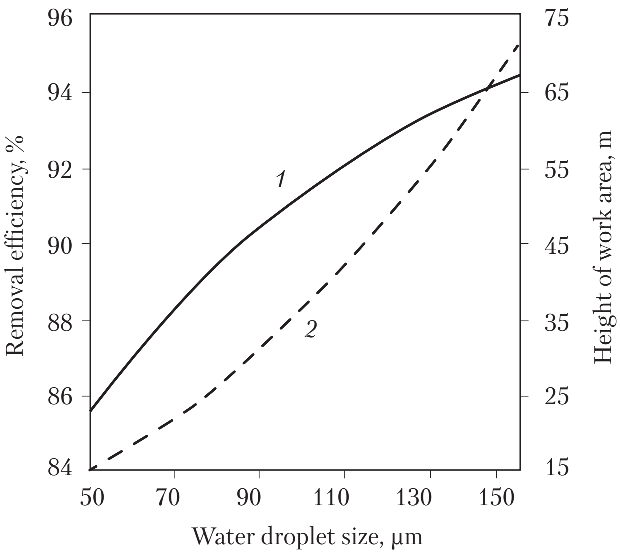

Fig. 6. Droplet size effect on effectiveness of sulfur dioxide binding (*1*) and height of work area (*2*)

The size of droplets affects the effectiveness of sulfur dioxide binding since the total surface area of droplets depends on their size. In their turn, water absorption and evaporation rates depend on the droplet area. In most cases, there is a need to change water consumption, which can lead to changing the droplet size. The effect of irrigation water droplet size in the range from 50 μm to 150 μm on the effectiveness is showed in Fig. 6. As the droplet size increases from 100 μm to 150 μm, the process effectiveness rises by 3% due to the fact that large drops «live» longer in hot gas stream. Also, an increase in the share of chemically bound sulfur dioxide in the gaseous phase is reported. If the droplet size decreases from 100 μm to 50 μm, the process effectiveness falls by 6% since the droplets evaporate faster. However, increasing droplet size can lead to a significant increase in the required height of chemical reactor work area. The previous studies have showed the optimal droplet size is 100 μm [7].

CONCLUSIONS

Based on the results of mathematical study of the plant for semi-dry ammonia purification of flue gases from sulfur dioxide formed during combustion of anthracite in TPP-210A twin boiler of 300 MW power unit the following has been established:

1. For the implementation of semi-dry sulfur purification method on 300 MW power unit, it is advisable to construct two chemical reactors with an internal work area diameter of 9 m and a height of 40 m. Since the size of liquid droplets is decisive factor for choosing the height of chemical reactor, the optimal droplet size is 100 μm. The consumption of flue gases per reactor is about 1 million m^3/h, at a temperature of 150 °C. Both reactors are installed along gas path behind the ash removal (electric precipitator). In order to capture fine byproduct particles, the sleeve filters are mounted behind the reactors.

2. In each reactor, the method of double-stream fluid supply by the «solution-water» scheme is used

Table 2

Fluid Consumption at Different Reactor Loads

Parameter	Value				
Load, %	100	90	80	70	65
25% NH_3 solution, kg/s	1.168	1.052	0.935	0.818	0.759
Water for solution, kg/s	4.674	4.206	3.739	3.272	3.038
5% NH_3 solution, kg/s	5.842	5.258	4.674	4.090	3.797
Processing water, kg/s	3.784	3.405	3.027	2.649	2.459
Water consumption, kg/s	8.458	7.611	6.766	5.921	5.497
Water consumption, t/h	30.447	27.402	24.357	21.313	19.790

as optimal one. It foresees that the droplets of 5% ammonia solution are introduced to the gas stream at the inlet of the work area, while those of irrigation water are injected at a distance of about 8.3 m. At the exit of the chemical reactor, all fluid injected should evaporate, whereas the small particles of ammonium sulfate should be captured by the sleeve filter.

3. The temperature of service water and flue gases varies as a result of seasonal fluctuations in the atmospheric air temperature. Within the range of 10—30 °C, the service water temperature almost does not affect the plant effectiveness. However, temperature of flue gases varying within the range of 130—170 °C requires adjusting the irrigation water consumption according to the calculations of Table 1.

4. The consumption of solution and irrigation water should correspond to the reactor load in order to maintain the maximum effectiveness of sulfur dioxide binding. Information on the fluid consumption depending on the plant load is given in Table 2.

5. At nominal mode, the consumption of 5% ammonia solution by one chemical reactor is 5.8 kg/s; that of irrigation water is 3.8 kg/s. The total water consumption by reactor is 33.6 t/h. The purified gas temperature at the exit from the reactor is controlled by changing the consumption of irrigation water. The control of ammonia solution consumption is based on the concentration of sulfur dioxide in flue gases at the outlet of sleeve filter. If the load changes, the fluid consumption can be reduced by shutting some nozzles injecting the reagent solution and irrigation water into the reactor work area. The fluid is proposed to be supplied using pneumatic nozzles capable of providing the required size of droplets.

6. The calculations have showed that the gas temperature at the outlet from the plant is, at least, 60 °C and exceeds the temperature of water point of dew by, at least, 15 °C, which enables avoiding chemical corrosion of gas ducts. Taking into account the sulfur dioxide binding in the sleeve filter, SO_2 concentration will not exceed 169 mg/nm^3 and the overall effectiveness of flue gas desulphurization will reach, at least, 95%.

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ВИКОРИСТАННЯ ТЕХНОЛОГІЇ НАПІВСУХОЇ АМОНІАКОВОЇ ДЕСУЛЬФУРИЗАЦІЇ ДИМОВИХ ГАЗІВ НА ВУГІЛЬНИХ ЕЛЕКТРОСТАНЦІЯХ

Законодавство України та Європейського Союзу з охорони навколишнього природного середовища вимагає суттєвого скорочення викидів діоксиду сірки на теплоелектростанціях. Для цього слід спроектувати. виготовити та спорудити установку з десульфуризації димових газів. що відповідає європейським критеріям найкращих доступних технологій. Необхідною умовою є визначення оптимальних параметрів на змінних режимах роботи очисної установки. В статті представлено результати числового моделювання роботи установки напівсухого сіркоочищення для зв'язування діоксиду сірки димових газів. які надходять з котельного агрегату типу ТПП-210А енергоблоку електричною потужністю 300 МВт. Установка десульфуризації працює за напівсухим методом з використанням в якості сорбенту амоніакової води. За результатами дослідження встановлено розміри робочої зони реактора як головної частини сіркоочисної установки. Визначено спосіб подавання та витрату амоніаку і води. що використовуються для приготування розчину і зрошення газового потоку. Виконано оцінку витрат розчину амоніаку й води залежно від обсягу димових газів і води та вхідної температури газів.

Ключові слова: десульфуризація, амоніак, хімічний реактор, енергоблок.

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ИСПОЛЬЗОВАНИЕ ТЕХНОЛОГИИ ПОЛУСУХОЙ АММИАЧНОЙ ДЕСУЛЬФУРИЗАЦИИ ДЫМОВЫХ ГАЗОВ НА УГОЛЬНЫХ ЭЛЕКТРОСТАНЦИЯХ

Законодательство Украины и Европейского Союза по охране окружающей природной среды требуют существенного снижения выбросов диоксида серы на теплоэлектростанциях. Для этого необходимо спроектировать, изготовить и построить установку для десульфуризации дымовых газов, которая отвечает критерию наилучшей доступной технологии. Необходимым условием есть определение оптимальных параметров на переменных режимах роботы очистной установки. В статье представлены результаты численного моделирования работы установки полусухой сероочистки для связывания диоксида серы дымовых газов, поступающих из котельного агрегата типа ТПП-210А энергоблока электрической мощностью 300 МВт. Установка десульфуризации работает по полусухому способу с использованием в качестве сорбента аммиачной воды. По результатам исследования установлены размеры рабочей зоны реактора, как главной части сероочистной установки. Определён также способ подачи и расход аммиака и воды, которые используются для приготовления раствора и орошения газового потока. Выполнена оценка расхода раствора аммиака и воды в зависимости от объема дымовых газов, а также температуры газов и воды.

Ключевые слова: десульфуризация, аммиак, химический реактор, энергоблок.