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RESEARCH ARTICLE

Comparison of flue gas desulphurization processes based on life cycle assessment

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Abstract

In this work, the environmental impact assessment is prepared for three different flue gas desulphurization (FGD) processes: (1) intra-furnace sulphur removal during coal combustion with limestone addition, (2) FGD with wet lime scrubbing, (3) regenerative copper oxide flue gas clean-up process. The evaluation and ranking of the three processes according to their environmental impacts is completed for the treatment of as much flue gas that contains 1 kg sulphur. The assessment of the environmental impacts is carried out with the Eco-indicator 99 life cycle impact assessment methodology based on life cycle inventories collected from existing coal fuelled power plants. The environmental assessment is prepared for three different scenarios according to degree of the utilization of the by-products obtained during the desulphurization: (1) zero utilization, (2) full utilization, (3) utilization according to industrial statistics.

The results show that all the three investigated FGD processes have about 80% lower environmental impact than the uncontrolled release of sulphur oxides into air. Intra-furnace limestone addition and wet scrubbing processes use similar principal of physical chemistry and they have similar environmental indices. The basis of the regenerative process is a sorption/reduction/oxidation cycle that has higher SO_2 removal efficiency than the two other processes. This higher efficiency results in a significantly lower environmental impact.

Keywords

environmental performance · flue gas desulphurization · life cycle assessment

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1 Introduction

Coal plays a significant role in the generation of electricity. In 2003, about 110,000 TWh primary energy was consumed worldwide and, on a global basis, coal-fired processes provided about 26% of the net electricity generated [1]. The global coal consumption rate is about 5,100 Mt coal per year, and this value is expected even to grow over the course of this century due to its relative abundance [2]. Due to the sulphur content of coal that varies normally between 0.3 and 1.2 wt% [3], sulphur-oxides (SO_x), mainly SO_2, are formed through oxidation of sulphur during high temperature coal combustion. The atmospheric SO_2 is a long range transboundary air pollutant responsible for respiratory problems and acid rain. The uncontrolled release of SO_2 from coal-fuelled power plants would raise the amount of anthropogenic SO_2 emission by about 150%, therefore, several attempts have been made on the regulation of the SO_2 emissions, i.e. the Helsinki Protocol (Protocol on the Reduction Sulphur Emissions or their Transboundary Fluxes by at least 30 per cent) and the Oslo Protocol (Protocol on Further Reduction of Sulphur Emissions) submitted by the United Nations Economic Commission for Europe or Clean Air Act Amendments 1990 (CAAA) passed by the U.S. Congress [4].

Techniques for reducing emissions of SO_2 during the combustion of fossil fuels can be distinguished as (1) pre-combustion, (2) intra-furnace sulphur removal, (3) and end-of-pipe abatement technologies (flue gas desulphurization). Pre-combustion sulphur removal includes a wide range of technologies, i.e. microbial desulphurization, halogenation, pyrolysis, electrochemical oxidation, irradiation [5], liquid phase methanol process and coal gasification [6]. Intra-furnace sulphur removal is possible with the addition of alkaline sorbent such as calcium oxide or calcium carbonate to the coal therewith removing SO_2 with dry-adsorption. Intra-furnace sulphur removal is generally done in circulating fluidized bed boilers allowing higher residence time for the sorbent particles. These processes have inherent environmental benefits over end-of-pipe flue gas desulphurization (FGD) processes, since there is no need for expensive FGD equipment; however, the retrofitting of an existing boiler is difficult and it might require a new apparatus.

End-of-pipe FGD is an effective control of SO_2 emissions from coal-fired power plants. In the last few decades, FGD processes have undergone considerable developments in terms of improved removal efficiency and reliability, as well as reduced costs. Wet scrubbers are the most commonly used FGD system, accounting for 87% of the total FGD capacity world wide and wet limestone is the predominant process, accounting for 82% of all installed wet FGD capacity worldwide. Great advantage of wet limestone scrubbing is its relative easy adaptability to existing plants and the low operating cost because of the low prices of limestone [7]-[9].

Former discussed SO_2 removal techniques are called once-through processes since continuous delivery of fresh sorbent is required for the operation. Simultaneously, a huge amount of by-products is generated that has to be disposed of. FGD processes with regenerable sorbent offer a solution for that problem.

A relative new technology developed at the US Department of Energy Federal Energy Center is called Copper Oxide Technology which is able to reduce SO_x and NO_x in a single unit. The copper oxide process (CuO process) is a dry regenerable process that has many advantages over wet scrubbers: it does not produce landfill waste, thus it avoids concerns over the limited landfill space and it does not increase landfill costs related to SO_2 removal, moreover, it calls public awareness of the environmental impact. CuO process also provides an effective way to obtain a concentrated SO_2 stream that can be used to produce sulphuric acid, elemental sulphur, fertilizer, etc. The valuable by-product partially compensates the operating costs [11].

The selection between SO_2 control technologies is usually made on the basis of economic considerations, and environmental performance of the technologies are usually characterized by concentration of the SO_2 remaining in the end gas. However, the control technologies have also a significant environmental load. Life cycle assessment (LCA) provides a framework for identifying and evaluating environmental burdens associated with the life cycles of materials and services in a "cradle-to-grave" approach therewith providing the possibility of an environmental-focused comparison end evaluation. Chevalier et al. [12] studied flue gas cleaning processes (a typical wet process and the new transported droplets column) of municipal solid waste incinerators with LCA approach. They found that the global environmental burden is similar between the two processes which conforms the viability of the transported droplets columns process. Benetto et al. [13] investigated the environmental issues of electricity production scenarios promoting the design of new production scenarios. Meyer [14] studied the rate of greenhouse gas emission evolving by electricity generation offering accurate means for evaluating greenhouse gas emission reduction strategies for U.S. electricity generation. Environmental consciousness has to be integrated into process engineering too; however, it requires the numerical expression of the environmental impacts. Several attempts have already been made in this field [15]-[19]; however, a comprehensive solution for the integration of LCA results into the process engineering has not yet been presented.

In this paper, the comparison of three SO_2 removal techniques used by coal-fuelled power plants is presented. The three options are: (1) intra-furnace sulphur removal with limestone addition in an atmospheric circulating fluidized bed combustor (ACFBC); (2) FGD with wet lime scrubbing, and (3) FGD with the regenerable CuO process. The three options use different physical and chemical principles by the flue gas clean-up. The comparison is made on the basis of environmental impacts derived from energy and mass balances of the investigated processes considering the cradle to grave approach. Environmental impacts are related to 1 kilogram sulphur entering the studied control equipments. The environmental impact is assessed by the Eco-indicator 99 life cycle impact assessment methodology supported by the software SimaPro 6.0. [20, 21].

2 Discussion

Comparison of the selected SO_2 removal options is carried out on the basis of annual input-output inventories [11, 22, 23]. Functional unit for the LCA is defined as the treatment of flue gas containing 1 kg sulphur in form of SO_2. System boundary includes:

- sorbent production (mining of raw materials, manufacturing and transportation of sorbents);
- electricity consumption;
- discharge of the purified flue gas;
- disposal of FGD by-products.

Life cycle inventories of sub-processes, like sorbent production, electricity generation etc. are obtained from the built-in inventories (ETH-ESU 1996) of SimaPro 6.0 [20]. In our assumption, auxiliary energy requirement of the studied processes is covered by lignite-fuelled power plants. Air contaminants considered in this study are: SO_2, NO_x and particulate matter, since these are important air pollutants of coal-fuelled power plants, and the studied processes have great influence on these emission ratios. In our consideration, disposal of flue gas cleaning by-products can be done by landfilling in high active chemical landfills or by its industrial utilization (material recycling). FBC ashes (bed- and flying ashes from ACFBC systems) can be used by cement production. FGD gypsum (by-product of wet-limestone FGD) can be utilized by building industry. By-product of the CuO process is pure SO_2 which can be utilized by the chemical industry. Three possible disposal scenarios are considered and investigated which represent the less and the most desired situations in the field of the utilization; the third scenario aims to represent today's common situation:

- 0% of the by-products is utilized by the industry;
- 100% of the by-products is utilized by the industry, and
- utilization rate according to industrial statistics.

Industrial statistics about utilization rate of FGD by-products is obtained from the USGS Minerals Yearbook [10]. An extract form the yearbook is shown in *Table 1*. In the environmental evaluation, utilized by-products reduce the total environmental impact, since they replace the production of new materials consuming the Earth's resources. These valuable and utilizable by-products are called avoided products which have negative environmental impacts in our calculations.

The environmental evaluation of the studied processes is done with aggregated environmental impact indicators (Eco-indicator points) calculated by EI-99 method, egalitarian version with egalitarian weighting set. The expression of the environmental impacts requires the preparation of the life cycle inventories of the studied systems referring to the functional unit. Product systems (collection of materially and energetically connected unit processes which perform one or more defined functions) of the studied processes are discussed one by one for each process.

2.1 LCA and Eco-indicator 99

The definition of the Life Cycle Assessment (LCA) by the International Organization for Standardization (ISO) is "a systematic set of procedures for compiling and examining the inputs and outputs of materials and energy and the associated environmental impacts directly attributable to the functioning of a product or service system throughout its life cycle" [24]. The method is developed to evaluate the mass balance of inputs and outputs of systems and to organize and convert those inputs and outputs into environmental themes or categories relative to resource use, human health, and ecological areas. The set of data on all the energy and material input flows required by a process or product and all the output emissions to air, water and land, including solid waste is called life cycle inventory (LCI). Life cycle assessment is an environmental decision-making tool that can help an organization to estimate the environmental performance of its product or service from cradle to grave [25]. The outcome of an LCA is the quantification of the environmental impacts associated with a product throughout the entire production life cycle which makes possible the identification of the more environmental-friendly product, service or process [24]. The frames of the preparation of an LCA are even standardized and are given among others in the ISO 14040-43 standards.

Eco-indicator 99 life cycle impact assessment methodology (EI-99) is specially developed for making easier the preparation of LCA studies. The method contains a damage model (fate-, exposure-, effect- and damage analysis), a normalisation and weighting step, which make possible the expression of the environmental impact regarding a substance and even a product system with a single score, the so called Eco-indicator point. EI-99 is chiefly relevant to European industrial pollutions [20, 26]. Eco-indicator points are used to calculate potential damages occurring to the "Human Health" "Ecosystem Quality" and "Resources". Damage category Human Health includes impact categories: *carcinogens, respiratory organics, respiratory inorganics, climate change, radiation and ozone layer depletion*; category Ecosystem Quality includes *ecotoxicity, acidification/eutrophication* and *land use*; and category Resources include the impact categories *minerals* and *fossil fuels*. Environmental impacts calculated according to the three damage categories are converted into dimensionless figures by the normalization step; weighting step makes possible to evaluate damages from several aspects (i.e. long term impacts get as high priority as short term impacts or vice versa). There is no absolute value of the indicators; they have only a relative value: similar processes might be compared based on the Eco-indicator scores. The scale of Eco-indicators is chosen in such a way that the value of 1 pt is representative for one thousandth of the yearly environmental load of one average European inhabitant. The Eco-indicator 99 is acknowledged as a standard investigation tool of LCA and applied in 45 countries [20].

Egalitarian version of EI-99, which is used in this study, means that environmental impacts are approached in a precautionary manner. The time perspective is long, theories are not accepted that predicts that future problems can be avoided, resources are assumed to be depleting, long-term effects of chemicals are considered in nature. The weighting set is also egalitarian that means that impacts regarding the impact categories Human Health, Ecosystem Quality and Resources are considered with 30%, 50% and 20% weighting, respectively.

2.2 SO_2 Removal in ACFBC

In the investigated atmospheric circulating fluidized bed combustor limestone is contacted with the flue gas in a circulating fluidized bed and SO_2 is captured by the calcinated limestone in a sulphation reaction, as shown in *Eqs.* 1 and 2a-b. The process is schematically shown in *Fig. 1*. The fluidized bed is formed as a result of pure air and/or flue gas flowing upward through a bed of sorbent solids. ACFBC provides a long contact time between the sorbent and flue gas because sorbent passes through the bed several times. The flue gas laden with reaction products then flows to a particulate control device, in this case electrostatic precipitator (ESP). Bed ash produced in the furnace is removed and sent to disposal. An additional benefit of the fluidized combustion is that low furnace temperature and air factor makes possible the reduction of thermal NO_x formation. Based on *Eqs.* 1 and 2a-b, the theoretical Ca/S ratio is 1. Because of steric problems, the inner parts of the CaO particles can not be utilized by the sulphation reaction, therefore the Ca/S ratio should be set at least between 1.4 and 2 [27]. Due to the relative low furnace temperature, sulphation reaction is strongly shifted to the right side and the product (gypsum) is thermally stable. The obtained gypsum either gets to fly or bottom ash.

$$\text{calcination:}\quad CaCO_3 \rightarrow CaO + CO_2 \tag{1}$$

$$\text{sulphation:}\quad CaO + SO_2 \rightarrow CaSO_3 \tag{2}$$

$$CaSO_3 + 0.5\,O_2 \rightarrow CaSO_4 \tag{3}$$

Tab. 1. Utilization ratios [%] of several coal combustion by-products, based on the data of the European Coal Combustion Products Association [10].

	Fly ash	Boiler slag	FBC ashes	FGD gypsum
Cement raw material	20.6	–	–	–
Blended cement	10.6	–	2.2	–
Concrete addition	29.9	6.6	6.7	–
Aerated concrete blocks	3.7	–	–	–
Nonaerated concrete blocks	3.2	–	–	–
Lightweight aggregate	1.3	–	–	–
Bricks and ceramics	0.4	–	–	–
Grouting	2.9	6.6	–	–
Asphalt filler	1.0	–	11.1	–
Subgrade stabilization	1.8	–	–	–
Pavement base course	1.2	51.7	–	–
General engeneering fill	7.2	–	–	–
Structural fill	7.6	–	–	–
Infill	7.6	–	80.0	–
Blasting grit	–	30.2	–	–
Plant nutrition	–	1.7	–	–
Set retarder for cement	–	–	–	7.1
Projection plaster	–	–	–	9.4
Plaster boards	–	–	–	61.0
Gypsum blocks	–	–	–	3.6
Self levelling floor screeds	–	–	–	18.9
Other uses	1.1	3.7	–	–

Environmental evaluation of an ACFBF system is carried out on the basis of the annual input-output database of an existing European power plant. Operational and technical parameters of the investigated ACFBC system correspond to the generic technological descriptions found in the Oslo Protocol. Operational data are obtained from the literature [23, 30]. In the reference year (1999), 295 kilotons of lignite were fired with an average sulphur content of 1.7%. The molar Ca/S ratio was 1.7/1 in the reference year. NO_x control is done with primary measures: low combustion temperatures and air staging reduce the formation of thermal NO_x to the desired low level. For dust control an ESP with separation efficiencies of $>$99.9% is installed. In our consideration, bed ash and fly ash forms FBC ash.

2.3 FGD with Wet Lime Scrubbing

A generalized flow diagram of a baseline wet FGD system is shown in *Fig. 2*. Fly ash is removed from the flue gas by a particulate control device, in this case ESP. The SO_2-containing flue gas is then contacted with limestone slurry in an absorber. Limestone slurry is prepared in two consecutive steps. First, limestone is crushed into fine powder with a desired particle size distribution. This takes place in a crushing station. Next, this fine powder is mixed with water in a slurry preparation tank. Sorbent slurry from this tank is then pumped into the absorber reaction tank where limestone slurry is sprayed downwards by an array of spray nozzles. In the absorber, SO_2 is removed by both sorption and reaction with the slurry. Reactions initiated in the absorber are completed in a reaction tank, which provides retention time for finely ground limestone particles to dissolve and to react with the dissolved SO_2.

The main reaction in the absorber and in the reaction tank can be summarized in *Eq.* 3.

$$SO_2 + {}^1\!/_2O_2 + CaCO_3 + 2H_2O \rightarrow CaSO_4.2H_2O{+}CO_2 \quad (4)$$

Normally, the required stochiometry of a limestone wet FGD system varies from 1.01 to 1.1 moles of $CaCO_3$ per mole of SO_2. Spent sorbent from the reaction tank (slurry bleed) is dewatered and disposed.

In this study, a power plant applying a pulverized lignite-fired dry bottom boiler is investigated. Operational data are obtained from the literature [23, 30]. Operational and technical parameters of the investigated ACFBC system correspond to the generic technological descriptions found in the Oslo Protocol. In the reference year (1999) 12,068 kilotons of lignite were fired with an average sulphur content of 0.73%. Sulphur oxides are removed with wet limestone scrubbing consuming limestone, water and auxiliary energy and producing FGD gypsum. Waste water produced by the FGD unit is utilized by fly ash sedimentation. NO_x control is performed with primary measures which mean fuel and air staging. Dedusting is done with ESP that consumes auxiliary energy and produces fly ash. FGD gypsum and fly ash can be utilized by the building industry.

2.4 Regenerable FGD with Copper Oxide System

The investigated regenerable FGD technology with CuO system is developed by DB Riley Inc. [28]. The technology is being researched under the Department of Energy's sponsorship. The technology uses copper oxide flue gas clean-up process that utilizes a regenerable sorbent, removing both SO_2 and NO_x from the flue gas and producing pure SO_2 instead of creating a solid waste. The process is shown schematically in *Fig. 3*. The basis of this SO_2 removal technology is that CuO can readily react with SO_2 in the flue gas at temperatures around 350 to 400°C to form $CuSO_4$, see *Eq.* 4. $CuSO_4$ then can be reduced to Cu with methane or other reducing gases, releasing SO_2 in a concentrated form that can be used in various processes, see *Eq.* 5. The regenerated sorbent is exposed to the flue gas so that the elemental copper is converted to copper oxide that can be again used to react with SO_2, see *Eq.* 6. The temperature in reaction unit is 350°C while the regeneration takes places at 500°C. Practically, the CuO is layered on an Al_2O_3 carrier with approx. 7% CuO content [11]. The main reactions can be expressed as follows:

$$\mathrm{CuO{+}SO_2} + {}^1\!/\!_2\,\mathrm{O_2} \rightarrow \mathrm{CuSO_4} \tag{5}$$

$$\mathrm{CuSO_4} + {}^1\!/\!_2\,\mathrm{CH_4} \rightarrow \mathrm{Cu{+}SO_2} + {}^1\!/\!_2\,\mathrm{CO_2} + \mathrm{H_2O} \tag{6}$$

$$\mathrm{Cu} + {}^1\!/\!_2\,\mathrm{O_2} \rightarrow \mathrm{CuO} \tag{7}$$

An interesting feature of the process is that both $CuSO_4$ and CuO can serve as catalyst for reducing the NO_x content of the flue gas to N_2 using NH_3. By injecting NH_3 into the flue gas before it contacts CuO impregnated sorbent, both NO_x and SO_2 in the flue gas can be removed. The NO_x reduction reactions can be written as:

$$4\,\mathrm{NO} + 4\,\mathrm{NH_3} + \mathrm{O_2} = 4\,\mathrm{N_2} + 6\,\mathrm{H_2O} \tag{8}$$

$$6\,\mathrm{NO} + 4\,\mathrm{NH_3} = 5\mathrm{N_2} + 6\,\mathrm{H_2O} \tag{9}$$

$$2\,\mathrm{NO_2} + 4\,\mathrm{NH_3} + \mathrm{O_2} = 3\,\mathrm{N_2} + 6\,\mathrm{H_2O} \tag{10}$$

$$6\mathrm{NO_2} + 8\,\mathrm{NH_3} = 7\,\mathrm{N_2} + 12\,\mathrm{H_2O} \tag{11}$$

Operational data of this CuO process applied in this study is obtained form the literature [11, 22, 28, 29]. The CuO technology studied here was applied as a secondary FGD and DeNOx measure at a Low Emission Boiler System (LEBS). LEBS is projected to have significantly higher thermal efficiency, better performance, and a lower cost of electricity. Emissions for this system are those forecasted from future plant utilizing a LEBS. Sulphur oxides and nitrogen oxides coming from the boiler are removed in the CuO absorber unit, dust is removed in the fabric filter following the absorber unit. Theoretically produced CO_2 emission is also considered, see *Eq.* 5. Fly ash is recycled to the boiler. The LEBS system uses U-fired slagging boiler which converts the coal ash and fly ash into slag. As it is quenched, the slag converts into a low volume, inert, vitreous granulate [29]. The system consumes electricity for the pneumatic transport of the sorbent, NH_3 and CH_4 as reducing agents and air. By-products of the system are high concentrated SO_2 and boiler slag generated in the bed of the boiler. There are no statistical data about the utilization rate of the recovered SO_2; however, it is a valuable product in our consideration. Fly ash is recycled to the boiler and leaves the system in form of boiler slag.

3 Results

First, LCIs of the studied processes are collected and prepared on the basis of operational data obtained from the literature sources. Input and output streams refer to as much flue gas that contains 1 kg sulphur. LCIs are shown in *Table 2*. Amount of avoided products is calculated on the basis of the three disposal scenarios, explained above. *Table 3* shows the utilization rates of each by-product generated by the processes in the three considered disposal scenarios. At the third scenario, statistical data obtained from the USGS Minerals Yearbook [10] are applied. In the case of SO_2 generated by the CuO process no statistical data is available for utilization. However, landfilling of SO_2 is not realistic option for disposal (Scenario 1), therefore it is omitted form the evaluation. Moreover, we assume a 100% utilization rate for SO_2 in the third scenario.

Based on the life cycle inventories (*Table 2*) completed with the disposal scenarios of the by-products, the environmental impacts of the studied processes are assessed by the EI-99 methodology. The results are shown in *Table 4*. Environmental impacts caused by the systems vary between 0.189 and 0.467 EI-99 points per 1 kg sulphur for the three systems. According to EI-99, the simple release of SO_2 into air causes 2.32 EI-99 points/kg sulphur damage to the environment. This shows the reasonability of the flue gas treatment: at least 80% reduction in the environmental impacts can be achieved by the application of flue gas desulphurization.

Total environmental impacts of the ACFBC process are signed by 0.467 EI-99 points referring to the functional unit, if 0% utilization is considered. Energy and material inputs, and airborne emissions are responsible for 35% and 56% of the total impacts, respectively. Contribution of by-products disposal (landfilling) is 9%. If the utilization of the by-products is considered, the total environmental impact is lower: 0.372 and 0.424 EI-99 points at the disposal scenarios 100% and general utilization rates, respectively.

Total environmental impacts of the FGD process with wet lime scrubbing are characterized by 0.425 EI-99 points per kg sulphur entering the system. About 61% of the total environmental impacts is generated during the energy and material consumption. However, efficiency of flue gas cleaning is higher which results that only 31% of the total impacts is linked to the airborne emission. Contribution of by-products disposal is 8%. If utilization of the by-products is considered, the total impact reduces by 27 and 17% at the two disposal scenarios. It can also be noted that the environmental impacts of the ACFBC process are lower than those of the FGD with wet lime scrubbing (0.425 EI-99 points), if the utilization of the by-products is only at the ACFBC process (0.372 and 0.424 EI-99 points) is considered. This may occur in a site specific case if i.e. FBC ash can be utilized while fly ash and FGD gypsum can not.

The novel flue gas desulphurization process, the CuO process, has the lowest total environmental impact: 0.216 EI-99 points if

Tab. 2. Input-output database of the studied systems, referring to the treatment of flue gas containing 1 kilogram of sulphur.

	ACFBC	FGD with Wet Lime Scrubbing	FGD with CuO
Coal used:	lignite	lignite	Illinois coal
Amount, kt/a	291	12,068	1,215
S%	1.7	0.95	4.0
Plant capacity, MW	43	1,500	407
DeNOx:	primary	primary	SCR
DeSOx:	N/A	Wet scrubbing	Reg. CuO abs.
Dedusting:	ESP	ESP	fabric filter
Input			
Limestone, kg/kgS	8.1	2.1	–
Electricity, kWh/kgS	3.8	6.0	1.6
Water demand, kg/kgS		39	–
CuO, g/kgS	–	–	19.5
Al2O3, g/kgS	–	–	279
Ammonia, g/kgS	–	–	9.9
Natural gas, m3/kgS	–	–	0.3
Output			
Emission to air, g/kgS			
SO_2	116.1	21.2	10.9
NO_x	49.8	44.7	10.9
PM	2.9	0.7	1.1
CO_2	1,295	1,360	684
Solid waste, kg/kgS			
FBC ash	10.9	–	–
Fly ash	–	4.7	0.9
FGD gypsum	–	3.7	–

Tab. 3. Utilization ratios of FGD by-products considered in the study.

Scenario	Disposal	ACFBC	FGD with Wet Lime Scrubbing		FGD with CuO	
		FBC ash	Fly ash	FGD gypsum	Boiler slag	SO_2
0% utilization	utilization	0%	0%	0%	0%	0%
	landfill	100%	100%	100%	100%	0%
100% utilization	utilization	100%	100%	100%	100%	100%
	landfill	0%	0%	0%	0%	0%
statistical data	utilization	45%	48%	87%	100%	100%
	landfill	55%	52%	13%	0%	0%

no utilization of the by-products is considered and 0.189 points if utilization is considered. Material and energy requirements generate 79% of the total impacts, while airborne emissions and disposal are responsible for 20% and 1% of the total impacts, respectively. The recovered SO_2 with high purity reduces the total impact by about 10%.

In the environmental comparison of the three investigated processes, material end energy requirements of the ACFBC process causes the less environmental impact per kilogram sulphur in the flue gas. However, this process has the lowest contaminant removal efficiency resulting high impacts due airborne emissions. Environmental impacts caused by airborne emissions released by the ACFBC process reaches 0.26 EI-99 points which significantly lower at the wet lime scrubbing and CuO processes (0.134 and 0.43 EI-99 points/kg sulphur respectively). ACFBC process produces even the most of by-products; however, the CuO process produces avoided products with the lowest EI-99 points.

Environmental impacts of the investigated processes to the damage categories (Human Health, Ecosystem Quality and Resources) are also investigated. Fig. 4 shows the environmental impacts of the studied processes to the damage categories in the first disposal scenario. The diagram shows that the ACFBC process causes the less damage in the category Resources. This can be explained by the relative low energy consumption and with the lower impact of limestone production in contrast to copper and alumina production. The CuO process has the lowest impact in the damage categories Human Health and Ecosystem Qual-

Tab. 4. Environmental impacts of the studied systems referring to 1 kg sulphur contained in the flue gas [10^{-3}EI point/kgS].

	ACFBC			FGD with Wet Lime Scrubbing			FGD with CuO		
Input									
Limestone	3			0.75			-		
Electricity	164			260			69		
Water demand	-			0.001			-		
CuO	-			-			27		
Al_2O_3	-			-			36		
Ammonia	-			-			1		
Natural gas	-			-			37		
Output									
Emission to air									
SO_2	135			25			13		
NO_x	114			102			25		
PM	6			1			2		
CO_2	5			6			3		
By-products	0% util.	100% util.	stat. data	0% util.	100% util.	stat. data	0% util.	100% util.	stat. data
Utilized									
FBC ash	0	-55	-25	-	-	-	-	-	-
Fly ash	-	-	-	0	-49	-24	-	-	-
FGD gypsum	-	-	-	0	-35	-30	-	-	-
Boiler slag	-	-	-	-	-	-	0	-2	-2
SO_2	-	-	-	-	-	-	0	-21	-21
Landfill	40	0	22	31	0	11	3	0	0
TOTAL	**467**	**372**	**424**	**425**	**311**	**351**	**216**	**189**	**189**

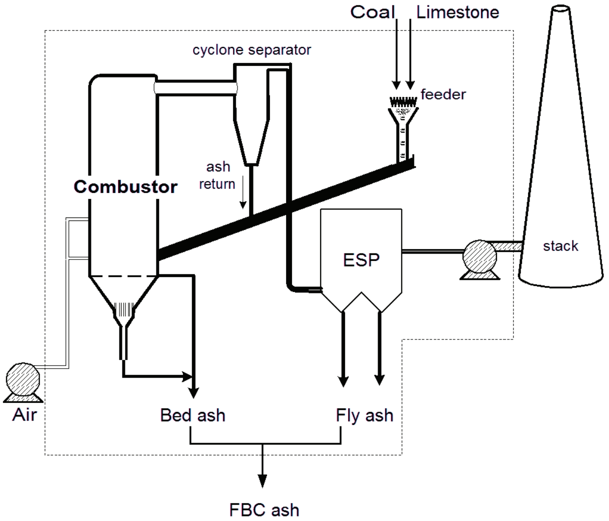

Fig. 1. Schematic diagram of the atmospheric circulating fluidised bed combustion

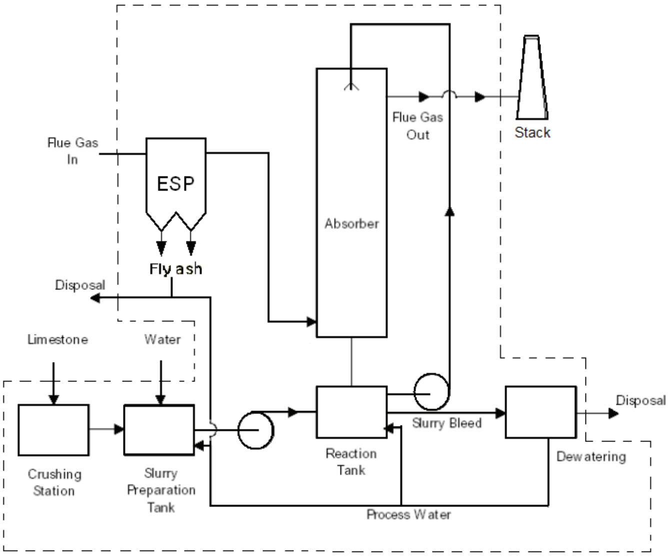

Fig. 2. Schematic diagram of the wet limestone scrubbing.

ity. The reason for the advantage of the CuO process in these two categories is the high contaminant removal efficiency of the process. The CuO process consumes a lot of raw materials; however this results in a low contaminant concentration in the flue gas leaving the process. The FGD process with wet lime scrubbing consumes the most of raw materials and energy; therefore, this process has the highest impact in the category Resources.

There is a change in the ranking if 100% utilization rate is considered. According to Fig. 5, the CuO process causes the less damage in the category Human Health and Ecosystem Quality; however, it causes the highest damage to the damage category Resources. In the comparison of the ACFBC and the FGD with wet lime scrubbing processes, ACFBC process is better in the categories Ecosystem Quality and Resources, but worse in category Human Health.

Fig. 6 shows the environmental impacts in the three damage categories if disposal of the by-products follows statistical data.

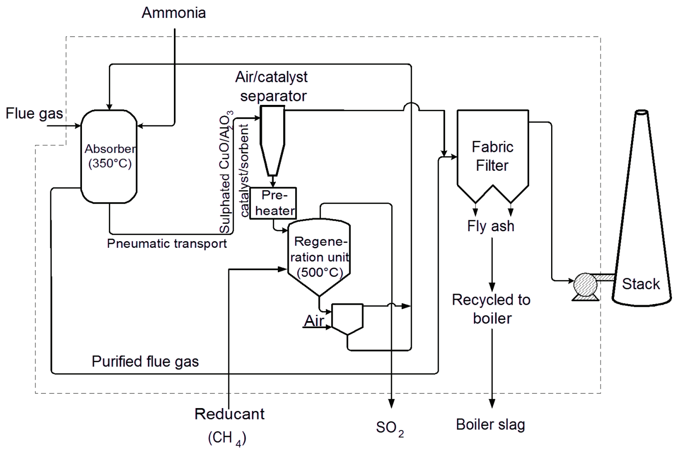

Fig. 3. Schematic diagram of flue gas desulphurization with regenerable copper oxide sorbent.

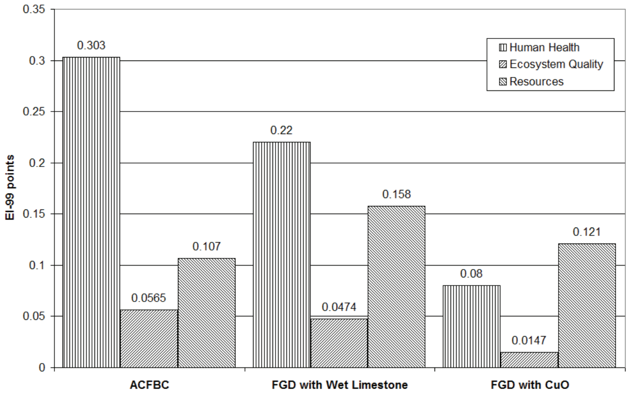

Fig. 4. Environmental impacts caused by the studied systems in the several damage categories if 0% by-products utilization rate is assumed.

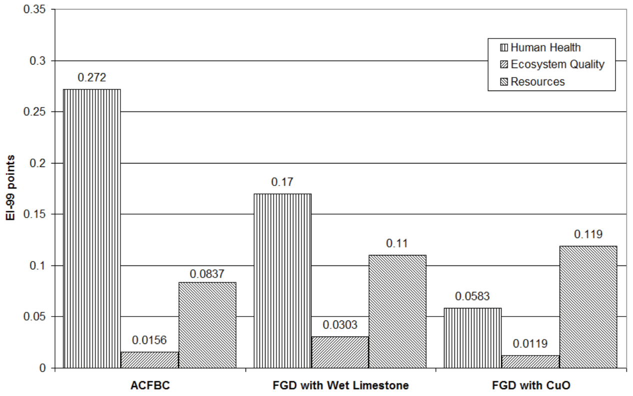

Fig. 5. Environmental impacts caused by the studied systems in the several damage categories if 100% by-products utilization rate is assumed.

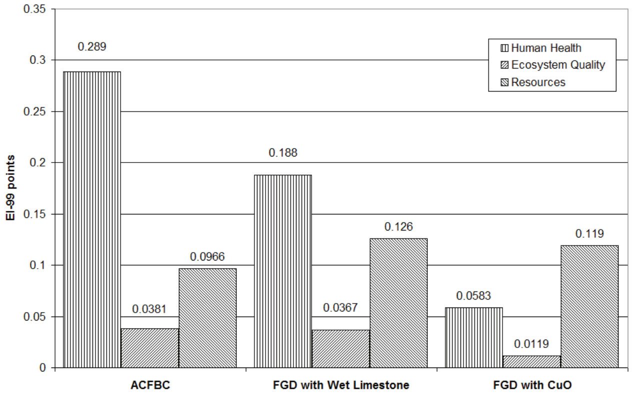

Fig. 6. Environmental impacts caused by the studied systems in the several damage categories; by-products utilization rate is based on statistical data.

Ranking of the three FGD options shows similar results as those of the first scenario (0% utilization rate): in categories Human Health and Ecosystem Quality, the CuO process has the lowest damage; in category Resources, the ACFBC process causes the lowest damage.

The results show that the conventional processes (ACFBC and FGD with wet lime scrubbing) capturing the SO_2 in the form of gypsum have similar environmental impacts during the treatment of the same quantity of SO_2. The most common used flue gas desulphurization process, the FGD with wet lime scrubbing, is slightly more environmental friendly than the ACFBC process; however, the difference of the two processes is from environmental viewpoint not significant. The lower contaminant removing efficiency of the ACFBC process results in a high impact to human health and ecosystem quality which is not balanced by the lower material and energy demand. The novel flue gas cleaning process, the CuO process, uses a regenerable sorbent and the captured SO_2 is not converted into a new chemical compound in the end of the removal. The higher removal efficiency, the higher value and the easier way of utilization of the by-products of the CuO process in comparison of those of the two other processes investigated, results in significant a lower total environmental impact. The CuO process causes lower damages to human health and ecosystem quality; however, this process consumes the natural resources on the same level then the conventional processes.

4 Conclusions

Three basically different flue gas clean-up processes have been compared based on their environmental impact caused during the treatment of flue gas containing 1 kilogram of sulphur. The first investigated process (atmospheric circulated fluidized bed combustion) is a conventional rarely applied intra-furnace dry flue gas clean-up process using limestone to capture SO_2 from the flue gas. During the removal process, SO_2 is converted to valuable gypsum which is, however, not separated from the fly ash and boiler slag. The second investigated process is the flue gas desulphurization with wet lime scrubbing using limestone slurry. This is a conventional, widespread applied secondary treatment option for SO_2 removal from flue gases. Great advantage of this technique is the easy way of retrofitting existing plants. This FGD process captures the SO_2 from the flue gas with chemical absorption and generates a valuable gypsum containing by-product. FGD gypsum is generated separated from other by-products and under appropriate running conditions high purity of the gypsum can be achieved. The third investigated process is a novel design. The Copper Oxide Process is dry secondary flue gas treatment option applying a regenerable CuO sorbent. Contrary to the former two processes, the captured SO_2 is not converted into a new chemical compound; it leaves the system as SO_2 with high purity.

The assessment of the environmental impacts is done with the Eco-indicator 99 life cycle impact assessment methodology based on life cycle inventories collected from existing coal fuelled power plants. Application of FGD processes reduces the total environmental impacts by at least 80%.The environmental based comparison of the three different processes shows that the conventional processes have similar environmental impacts during the treatment of the same quantity of SO_2. FGD with wet lime scrubbing is slightly better than the ACFBC process from environmental viewpoint; however, the difference of the two processes is not significant. The study shows that FGD with wet lime scrubbing is preferred if human health aspects are featured and ACFBC if raw material reserves are featured. The higher efficiency and the easier way of utilization of the by-products of the novel technique result in a significant lower environmental impact than those of the conventional techniques. The CuO process causes lower damaged to human health and ecosystem quality; however, it consumes similar amount of raw materials for the treatment of the same amount of sulphur in the flue gas as the other investigated processes.

The evaluation shows that the commonly used wet limestone scrubbing is slightly better than the rarely applied intra-furnace flue gas desulphurization process, however, the most attractive option is the flue gas cleaning system using regenerable sorbent where SO_2 is not converted into a new compound.

References

1 *BP Statistical Review of World Energy 2004*, available at `www.bp.com/statisticalreview2004`.

2 **McFarland J R, Herzog J, Jacoby H D**, *The future coal consumption in a carbon constrained world*, 7^{th} international Conference on Greenhouse Gas Control Technologies (September 2004).

3 *EMEP/CORINAIR Emission Inventory Guidebook, Combustion in Energy and Transformation Industries, European Environment Agency 2006*, available at `http://reports.eea.europa.eu/EMEPCORINAIR4/`.

4 **Metz N**, *World wide Emissions 1980 to 1996 and Future Trends*, Alliance Internationale de Tourisme & Fédération Internationale de l' Automobile; AIT&FIA Public Commission Week (Salsomaggiore, Italy, November 7).

5 **Thoms T**, *Developments for the Precombustion Removal of Inorganic Sulphur From Coal*, Fuel Processing Technology **43** (1995), 123-128.

6 *U.S. Department of Energy Assistant Secretary for Fossil Energy*: *Clean Coal Technology Programs: Project Fact Sheets 2003*, available at `www.netl.doe.gov/cctc`.

7 *The Royal Academy of Engineering*: *The Costs of Generating Electricity*, 2004. Westminster, London, SW1P 3LW.

8 **Wu Z**, *Air Pollution Control Costs for Coal-Fired Power Stations*: *IEA Clean Coal Centre Report CCC/53*, 2001.

9 **Rajaram S**, *Next Generation CFBC*, Chemical Engineering Science **54** (1999), 5567-5571.

10 **Kalyoncu R S**, *Coal Combustion Products 2000*: *United States Geological Survey Minerals Information*, 2000.

11 **Darguzas J N, Penrose L C, Litka A F, Chen Z**, *A Flue Gas Cleanup Technology of Next Generation - Copper Oxide Process*, 1999.

12 **Chevalier J, Rousseaux P, Benoit V, Benada B**, *Environmental Assessment of Flue Gas Cleaning Processes of Municipal Solid Waste Incinerators by Means of the Life Cycle Assessment Approach*, Chemical Engineering Science **58** (2003), no. 10, 2053-2064.

13 **Benetto E, Rousseaux P, Blondin J**, *Life Cycle Assessment of Coal By-Products Based Electric Power Production Scenarios*, Fuel **83** (2004), no. 7-8, 957-970.

14 **Meyer P J**, *Life-cycle Assessment of Electricity Generation Systems and Applications for Climate Change Policy Analysis*, PhD Dissertation, 2002.

15 **Chen H, Shonnard D R**, *Systematic Framework for Environmentally Conscious Chemical Process Design: Early and Detailed Design Stages*, Ind. Eng. Chem. Res. **43** (2004), 535-552.

16 **Burgess A A, Brennan D.J.**, *Application of Life Cycle Assessment to Chemical Processes*, Chemical Engineering Science **56** (2001), no. 8, 2589-2604.

17 **Alexander B, Barton G, Petria J, Romagnoli J**, *Process Synthesis and Optimisation Tools for Environmental Design: Methodology and Structure*, Computers & Chemical Engineering **24** (2000), no. 2-7, 1195-1200.

18 **Azapagic A, Clift R**, *The Application of Life Cycle Assessment to Process Optimisation*, Computers & Chemical Engineering **23** (1999), no. 10, 1509-1526.

19 **Azapagic A**, *Life Cycle Assessment and its Application to Process Selection, Design and Optimisation*, Chemical Engineering Journal **73** (1999), no. 1, 1-21.

20 **Goedkoop M, Spriensma R**, *The Eco-Indicator 99: a damage oriented method for life cycle impact assessment – Methodology Report*, third, PRé Consultants B.V., Amersfoort, Netherlands, 2001.

21 *SimaPro 6.0. Details*, available at `http://www.pre.nl`.

22 **Spath P L, Menn M K, Kerr D R**, *Life Cycle Assessment of Coal-fired Power Production*, 1999.

23 **Rentz O K, Gütling U.K.**, *Exemplary investigation into the state of practical realisation of integrated environmental protection with regard to large combustion plants in Germany*, 2002.

24 *ISO 14040: Environmental Management - Life Cycle Assessment - Principles and Framework*, 1997.

25 **Wenzel H, Hauschild M, Alting L**, *Environmental Assessment of Products: Methodology, Tools and Case Studies in Product Development*, Chapman & Hall, London United Kingdom, 2000.

26 **Hofstetter T B, Capello C, Hungerbühler K**, *Environmentally preferable treatment options for industrial waste solvent management - A Case Study of a Toluene Containing Waste Solvent*. B, Trans IChemE. **81** (2003), 189-202.

27 **Srivastava R K**, *Controlling SO_2 emissions*, 2000.EPA/600/R-00/093

28 **Ruth L A**, *The U.S. Department of Energy's Combustion Program: Clean, Efficient Electricity from Coal*, Energy Conversion Management **38** (1997), no. 10-13, 1249-1257.

29 **Ake T, Beittel R, Lisauskas R**, *Slag Tap Firing System for a Low Emission Boiler*, DB Riley, Inc., Worcester, MA, United States, 1999.

30 **Rentz O, Gütling K, Karl U**, *Erarbeitung der Grundlagen für das BVT-Merkblatt Großfeuerungsanlagen im Rahmen des Informationsaustausches nach Art. 16(2) IVU-Richtlinie*, 2002. Deutsch-Französisches Institut Für Umweltforschung Universität Karlsruhe Forschungsbericht 200 46 317.