### ANALYSIS OF THE MEASURES FOR NO<sub>x</sub> EMISSIONS REDUCTION RESULTING FROM FOSSIL FUELS BURNING AT DEVA THERMAL POWER PLANT FOR COMPLIANCE WITH THE EU ENVIRONMENTAL STANDARDS

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**ABSTRACT:** In order to line up with European environmental and energy efficiency standards as a result of Romania's accession to the European Union, it is required technological rehabilitation of coal plants, respectively the implementation of flue gas desulphurization installations and modernization of electrostatic de-dusting (Electrostatic Precipitators), as well as the decrease of the nitrogen oxides amount from flue gases and revision of the current transport, storage and disposal systems of products resulting from the combustion of coal into boilers.

The efforts of Deva Thermal Power Plant to meet EU environmental requirements are particularly important; their effect will be that of emissions lowering, with direct impact on human health, on flora and fauna in the area, having at the same time a social character, by maintaining jobs in a sensitive area of our country (Deva and Jiu Valley mining basin). Nitrogen oxides (NOx), an important complex of pollutants are generated in significant quantities in spite of an optimized combustion. For large power plants, compliance with different emissions is important from two points of view: it is mandatory to comply with the requirements imposed by environmental authorities relating to emissions of NOx and NH3 secondary emissions (so-called NH3 leaks) which may arise from the application of measures for NOx reduction, either by the SNCR or SCR method. Compliance with emission limits has an economic importance because some power plants residues (ash from electrostatic precipitators, gypsum from desulphurization installations) can be used as raw material in construction industry where these residues must meet certain purity requirements, such the contents of the ammonium salt [5].

**KEYWORDS:** Nitrogen oxides, Selective catalytic reduction (SCR), Selective non-catalytic reduction (SNCR), Urea, Pollutants.

#### 1. INTRODUCTION

Energy is a vital aspect of humanity, both in terms of consumption, but rather in terms of its generation. Most power plants use fuels and other raw materials from natural resources of the earth, transformed into useful energy. Fossil fuels are energy sources most often used today. However, the result of their burning leads over time to a relevant and significant impact on environmental as a whole.

The combustion process leads to the generation of emissions into air, water and soil; emissions generated into air are considered to be one of the main environmental problems. The main air emissions from burning fossil fuels are:  $SO_2$ ,  $NO_X$ , CO, particulate matter ( $PM_{10}$ ) and greenhouse gases, such as  $N_2O$  and  $CO_2$ . Other substances such as heavy metals, halogenated compounds and dioxins are generated in smaller quantities. [1]

Clean air is a basic requirement for human health and for the environment as well as for maintaining biodiversity. The general direction of humanity is moving towards clean energy generation. Romania's accession to the European Union puts domestic energy generation on one of the worst position in terms of competitiveness on well regulated global and European energy markets.

The form of energy generation by burning coal is the most polluting of those known and Societatea Complexul Energetic Hunedoara S.A cannot cut back on this issue: the obligations assumed by the Romania during EU accession process puts pressure on it in terms of environmental compliance. Moreover, the generation of electricity from fossil fuels, through their high costs lead to a high price of generated MW. Such price can hardly cope with a competitive energy market while environmental costs lead to a further increase of it.

At European and international level there is an increasing concern about optimal strategies and actions to reduce environmental pollution. Directive 2008/1/ EC (IPPC) – on Integrated Pollution Prevention and Control and Directive 2001/80/EC (LCP)- on limitation of air pollutants emissions from Large Combustion Plants are the two of the main directives which regulates the legal framework of environmental pollution reduction.

These two directives, plus another five are reunited in Directive 2010/75/EC (IED) –Industrial Emissions Directive, transposed into Romanian legislation by Law 278/2013, whose overall objective is to reduce emissions into the air, water and soil as well as wastes coming from agriculture and industry, in order to achieve the highest level of environmental and health protection.

The limit values that LCP within Deva TPP shall comply with are the following:  $SO_2$  –200 mg/Nm<sup>3</sup>;  $NO_X$  – 200 mg/ Nm<sup>3</sup>; Particulate matter – 20 mg/ Nm<sup>3</sup>.

The rehabilitation of power units is actually a shortterm priority of Complexul Energetic Hunedoara S.A – Deva TPP, whose final purpose is to protect the environment and the health of the personnel operating on site, as well as the health of population in the surrounding area and apart from it.

#### 2. DESCRIPTION OF DEVA THERMAL

#### **POWER PLANT**

Complexul Energetic Hunedoara S.A. was set-up on 01.11.2012 by the merger of SC Electrocentrale Deva S. A. and SC P.E.E.T. Electrocentrale Paroşeni S. A. On 01.08.2013 was registered the merger by absorption between Complexul Energetic Hunedoara S.A as the acquiring company and the National Coal Company SA (SNH), as the company being acquired, in whose administration are four mines in the Jiu Valley: Lonea, Livezeni, Vulcan and Lupeni. Complexul Energetic Hunedoara S.A provides about 5% of Romania's electricity generation, with a current installed capacity of 1.435 MW, being the largest single power generator in the center and northwest of the country. (Figure 1) [2]







**General layout – Deva TPP** 



**Deva TPP** 

Figure no. 1 – Presentation of Deva TPP [6].

#### Deva TPP:

• provides about 4% of electricity generation in Romania, with an installed capacity of 1.075 MW in five power units and generating thermal energy in cogeneration which is delivered to the inhabitants in Deva, being an important power generator from the center and northwest of the country.

• has a strategic importance to the National Power Grid due to its geographical location, on the 400 kV line connected to the Western European network, providing power injection required for proper operation of the interconnected power system. [2], [3].

Power aggregates operating within Deva TPP are divided into the following LCPs, according to G.D. No.

440/2010- on setting measures to limit air emissions of certain pollutants from Large Combustion Plants: LCP 1 (boilers 2A, 2B); LCP 2 (boilers 3A, 3B and 4A, 4B); LCP 3 (power boilers 5A, 5B, 6A, 6B).

#### 3. GENERAL CONSIDERATIONS NITROGEN OXIDES (NO<sub>x</sub>)

In contrast to sulfur oxides which are closely connected to the sulfur content of the fuel used, the amount of NOx generated during combustion, although it depends to some extent on the type of fuel used, depends at the same time on the conditions in which combustion occurs. The main NOx generated during fossil fuels firing are: nitrogen oxides (NO), nitrogen dioxide (NO<sub>2</sub>) and nitrogen protoxide (N<sub>2</sub>O). The first two of these form a mixture known as NO<sub>x</sub>, constituting more than 90% of NO in the main types of LCP. NO<sub>x</sub> formation is prevailed by three key mechanisms characterized by the origin of nitrogen and the environment where the reaction takes place:

• *thermal*  $NO_X$  results from the reaction of oxygen with nitrogen form air (at t > 1000<sup>0</sup>C);

•  $NO_X$  fuel is formed from the nitrogen found in the fuel (at t < 1000<sup>0</sup>C);

• Sudden  $NO_X$  formed at the conversion of molecular nitrogen into the flame in the presence of intermediary components of hydro-carbon (small quantity) (figure no 2.a). [5].

The amount of NOx generated from fuel is higher in power plants firing coal, because nitrogen amount is higher is his structure than in the other type of fuels (coal: -  $0.5 \div 2\%$  nitrogen in the fuel, dry, ash-free basis).

Thermal nitrogen oxides are mainly formed of oxygen monoxide (NO) by means of a slow mechanism applicable to the nitrogen in the air, starting from the next area of the flame front or in gas burners where the temperature is high enough to excite the particles to a level higher than the binding energy. Reduction of NO emissions can be achieved by:

• decrease of temperature in the boiler or in the burning chamber;

• decrease of the residence time in the areas with high temperatures.

• reduction of excess air for decreasing the concentration of oxygen atoms.

To highlight the previous, it will be taken into consideration a temperature increase by 50K which will lead to a doubling of nitrogen oxides NOx quantity, for an equal residence time (Figure 2b).



# Figure no. 2: a) The character of dependency between the temperature inside the flame and formation of NOx according to the 3 mechanisms

#### b) – Temperature effect on NOx formation [5]

#### 4. MEASURES OF NOX REDUCTION WITHIN DEVA TPP

Within Deva TPP there is a compliance program to reduce emissions of nitrogen oxides (NOx) for large combustion plant no. 2 (LCP 2) as well as for unit no. 3 and unit no. 4 to values that will not exceed 200 mg /Nm<sup>3</sup>. Reduction of nitrogen oxides (NOx) from the combustion of fossil fuels in boilers of the thermal power plant involves the execution of important works which include:

✓ Primary measures (control and reduction of  $NO_X$  in the burning process);

 $\checkmark$  Secondary measures (technics applied at the end of burning process);

It is considered that the NO<sub>X</sub> emission values below 200 mg/Nm<sup>3</sup>, on all thermal load range of the boiler can be obtained through the implementation of primary measures associated with secondary measures. Reducing emissions from 600 to 700 mg/Nm<sup>3</sup> (maximum values) to a minimum of 200 mg/Nm<sup>3</sup> only through primary measures, even with advanced technology, it is not possible, considering the relatively high nitrogen content in the fuel (0,5 – 0,7%.)

#### 4.1. PRIMARY MEASURES OF NITROGEN OXIDES REDUCTION

Generally, the primary measures aim to limit the formation of thermal NO, through a management of the combustion, so that the oxidation speed of the fuel to be limited to the extent possible so as to avoid the formation of high temperature centers. When introducing primary measures, it is important to avoid occurrence of adverse effects or the formation of other pollutants. Therefore, it should be considered:

 $\checkmark$  Aspects related to safety operation (stable ignition of the fuel on the total load range);

✓ Operational safety (to prevent corrosion, erosion, deposition, slagging, overheating of pipes, etc.);

 $\checkmark$  Ability to burn the total range of fuel for which the power plant was designed;

 $\checkmark$  Complete combustion so that the carbon in the ash is below the limit imposed by ash marketing (5%) and avoid high emissions of carbon monoxide;

✓ Avoid the formation of other pollutants, such as organic matter and  $N_2O$ ;

✓ Low maintenance costs. [5]

Power unit no. 3 was rehabilitated and completed in 2009, improving its operational and economic performances so that electric power increased to 235 MW while maintaining steam consumption non-rehabilitated, as in previous situation.

Power unit no.3 is used for electrical service of secondary control system and for high-efficiency cogeneration scheme. Heat generation capacity through cogeneration increased from 58 MW to 140 MW. The two boilers belonging to power unit no.3 have been modernized and adapted to limit NOx generation based on the application of the primary methods. Essentially, these measures consisted of:

 $\checkmark$  Totally replacing the system for preparation and burning of coal dust;

 $\checkmark$  Installation of new mills, with rolls, operating on coal crushing principle;

 $\checkmark$  Replacing old burners with new swirling burners, but designed for low emission of NOx (Low NOx) with combustion in stages;

 $\checkmark$  Addition to the air supply system belonging to combustion installation of new OFA ports (controlled air insertion into the furnace).

Each of the two boilers is equipped with a coal dust preparation system comprising 4 mills (3 in operation -1 mill used as back-up). Grinding is based on the crushing principle. Each mill supplies coal dust to the two burners which are located on one of the two side walls, both on the same floor (at the same level).

There are 8 burners totally, 4 on each side walls and placed on every 2 floors (level 11,00 m and 14,00 m) (Figure no. 3).



Figure no. 3 – Location of burners with low NO<sub>X</sub> and coal dust supply system of boilers at power unit **no.3**[6].

Coal is the primary fuel burned in the boilers, while flame support is made with methane gas. The modernized pulverized coal combustion system aims to decrease NOx of boiler below the limit of  $360 \text{ mg/Nm}^3$ (6% O<sub>2</sub> in flue gas output) while the ration of unburned carbon in the expelled ash to be as small as possible.

Air distribution has been adapted to this system to generate, in the burners area of a reducing-type combustion obtained by limiting the oxygen in the area. At the same time for drying and transportation of dust, it is used, as in the initial technology (project), the air, which later is used as a part of the combustion fuel.

Low levels of NOx are based on combustion air phasing which occurs in low NOx burners in three parts: the primary, secondary and tertiary air (Figure no. 4). Implementation of low NOX burners can increase the level of *carbon in the ash*, which must be kept between some limits not to compromise the management of these combustion residues. Improvement of *pulverized coal graining* in the mills is an effective way to prevent this problem.

According to the project, for boiler operation at loads higher than 80% (over 280t), NOx reduction is achieved below 400 mg/Nm<sup>3</sup> (compared to 6%  $O_2$  in dry gas) (Figure 4).



Figure no. 4 – Low NOx burner

 $NO_X$  emissions at partial loads increases due to high air flow (excess) required to be circulated through mills which are then blown into the furnace by the primary mixing of burners, and which by the excess air, it leads to firing in an oxidizing atmosphere, generating more NOx. Such operating condition occurs at boiler's lower loads (below 280t/h) when operational safety considerations require to operate three coal mills (6 burners).

In such cases the requirements on dust transport conditions and burning speeds impose a percentage increase of primary air unbalancing the distribution structure optimal for limiting NOx formation. Moreover, safe operation of coal mill requires that the temperature at separator's output to be within restrictive temperature range (70 ... 95 ° C). At lower values, clogging can occur at mills and at temperatures exceeding the upper limit, there is risk of dust selfignition. At the same time, safe operation of burners requires lower speed limits blowing a primary mixture.

Simultaneous fulfillment of the two conditions is achieved by providing a certain air flow entering the mill so that the temperature in the separator complies with the temperature range stated above, through appropriate injection of preheated hot air flow, and of cold air taken directly from the boiler air fan discharge. In these circumstances, the requirement regarding a certain distribution of combustion air into the primary air, secondary air and OFA may not be met, having negative consequences in limiting the  $NO_x$  generation. Basically, it is a conflict between the requirements for safe operation of the coal preparation and burning system and those for limiting NOx emissions. Thus, the primary measures implemented to reduce the formation of NOx emission values have no effect, as the values of emissions reaching  $500 \div 700 \text{ mg/Nm}^3$  (Figure 5).

In these circumstances, additional works are necessary to expand primary NOx reduction measures in order to provide emission values below 400 mg/Nm<sup>3</sup> for all thermal loads of the boiler. 200 mg/Nm<sup>3</sup> value

will be obtained by applying secondary measures and the deadline for compliance will be 01.01.2016.



## Figure no. 5 – Evolution of $NO_x$ emissions in 3 test conditions according to the boiler's thermal load [3]

In order to limit the formation of NOx, the air quantity in the main combustion area need to be limited, therefore burning has to be made under substoichiometric conditions or at the unit value limit. At the same time, it is necessary to preserve the existing diagram of mill control, keeping the gas flow through the mill and its variation in relation to the mill load.

Measures to reduce NOx emissions at partial loads cannot be limited to operating measures given the fact that safety operating conditions of mills imposed by coal mills datasheet and implemented through automatic control functions must, be met.

#### Flue gas recirculation

To avoid excess air, respectively burning in an oxidizing atmosphere, flue gas recirculation at mills must be applied, to the extent to which is simultaneously provided the optimum gas / fuel ratio required by the mills and the limitation of excess air in the burner area according to the system for primary measures of  $NO_x$  generation. Taking-up flue gas will be such as to provide the maximum decrease of dust load of the recirculated flue gas. Taking-up flue gas will be made after the electrostatic precipitators to minimize dust load flow in the recirculated gas. To limit the discharge height of recirculating gas fan it is proposed that sampling point to be after the fan gas, where static

pressure is around zero. The static pressure upstream of it is around 15 ... 20 mbar. Taking-up flue gas will be such as to provide the maximum reduction of dust load in the recirculated gas flow. Air flow injection for the mill is proposed to be done after the mill air fan, to avoid potential erosion of the rotor due to residual dust found in the flue gas (Figure 6).



Figure no. 6 – Diagram showing the flue gas recirculation system for each boiler [4].

In order to reduce primary air flow, but keeping the flow of the primary agent required by the mill, a certain amount of cold flue gas is recycled which, mixed with combustion air, will form the transport and drying agent from mills. In this way, a part of the combustion air is replaced in the mill and then is redistributed to other air flows so as to meet the requirements for optimum distribution to limit the formation of  $NO_X$ .

By lowering the total amount of combustion air through the mill, the cold airflow taken from the discharge air fan is also reduced. By this flow lowering, the thermal efficiency will be increased.

**Power unit no. 4** will get into a comprehensive process of rehabilitation which comprises works at boilers, steam turbine, electrostatic de-dusting installations system for preparing coal dust, etc., These equipment have been purchased amounting 21 million  $\notin$  including low NO<sub>X</sub> burners and OFA system.

#### 4.2. SECONDARY MEASURES FOR NITROGEN OXIDES REDUCTION

Secondary measures aim at reducing nitrogen oxides already formed, both those formed based on fuel from nitrogen and those thermal formed.

The amount of nitrogen oxides formed during combustion can be broadly reduced, by acting on the conditions under which the combustion takes place, by lowering the maximum temperature of the flame, by reducing excess oxygen, etc.

When these measures are insufficient, it is necessary to take into consideration methods of reducing nitrogen oxides from flue gases (smoke denitrification). This denitrification or reduction can be performed through "another combustion" using a hydrocarbon or by injecting into the smoke of products which, through  $NH_2$  radical release, nitrogen oxides are converted into  $N_2$  and  $H_2O$ . This transformation may be carried out in the presence of a catalyst or without it, but only under very precise temperature and mixing conditions. [5].

#### 4.2.1. Selective non-catalytic reduction (SNCR)

Selective non-catalytic reduction (SNCR) is a secondary method of reducing nitrogen oxides already formed. To conduct the lowering reaction, it is not necessary to use a catalyst. The reaction between the nitrogen oxides and the reducing reagent depends essentially on:

- flue gas temperature in the area where the latter is introduced;
- residence time in the area of optimal temperature.

The temperature range for the aqueous urea solution is between 850°C and 1150°C with optimal value of 1000°C. If ammonia is used as a reducing agent, the upper limit of the temperature range is lower, about 1000°C, with the optimum temperature of 870°C.

At boiler load decrease, the optimum temperature range is moving towards the burning area. For reagent injection to follow the optimal area, injectors will be placed on several floors, being activated, where appropriate, the floor spraying in the area corresponding to the optimal temperature. Real time knowledge of the temperature of flue gas combustion and its dynamics requires, in particular for large-capacity boilers, the installation of a temperature measurement system and generation of temperature maps (Figure 7).



Figure no. 7: Location of the spraying injectors and temperature measurement installation (SNCR)

In order to obtain a high-efficiency denitrification and a low ammonia leak, it is particularly important:

- to provide a tight mixture with flue gas;
- to be a uniform distribution of spraying in the combustion gases;
- obtaining an optimal spraying, i.e., a medium droplet size to provide a large area for the reaction of spraying but which avoids premature vaporization.

SNCR equipment is simple and very easy to install. Except the reactor tank which may be located anywhere, even far from the injection area, the rest of the equipment takes up very little space. The temperature measurement is mandatory at large boilers, because here can occur high temperature irregularities caused by the geometry of the burners' sprays that are in operation. The command and control system provides the management of the SNCR system, controlling according to the boiler load, the NOx emissions, the temperature map (isothermal geometry) etc, activation of injectors and their operating parameters (flow, pressure). This system is integrated in the boiler management system and automatically generates all controls necessary for proper operation of the SNCR installation.

Compared to the SCR, SNCR technology provides lower reduction efficiency. Lately, efficiencies of 55 ... 65%, have been achieved even for large capacity plants (over 500 t/h). A major drawback is that the high denitrification efficiencies, the specific consumption of reagent (the amount of reagent per reduced NO unit) increase considerably.

From Figure 8 it can be seen that up to 50% efficiency, the excess of reagent is comparable to that existing at SCR. For this reason, SNCR technology is economically limited to approx. 50% efficiency.

Investment costs for SNCR system for a power unit of 200 MW amounts to approx. 2 million Euros, being lower than for a similar SCR installation. Operating costs (it is considered particularly the cost of reagent) are approx. 2500 Euro/ton NOx depending on the DeNOx efficiency limitation to 50%.



Figure no. 8: Reagent excess according to the DeNOx efficiency

#### **4.2.2.** Selective catalytic reduction (SCR)

Selective catalytic reduction (SCR) is a process that involves catalytic reduction of nitrogen oxides with ammonia or urea in the presence of a catalyst (expendable with 6 ... 10 years of lifetime for coal fired power plants). NO<sub>X</sub> conversion takes place on the surface of the catalyst at a temperature which can range from 170°C to 510°C. There are three basic configurations for the integration of the SCR reactor in the chain of the flue gas cleaning, the main factor being the temperature of the flue gas:

a) **"high-dust" method** is preferred when it is desired to avoid reheating the flue gas (necessary for the catalyst operation) or to install a high temperature dust remover." This is the technology used in coal fired boilers, especially for the new boilers.

b) **"low-dust" method**, eliminates the disadvantage of the dust-loaded gas, not being required the installation of the flue-gas reheating; however, for this reason, it requires a dust remover (safe and effective) to operate at a high temperature.

c) c) SCR system is placed in the final area of flue gas flow with relatively lowered temperatures. Here, clean flue gas pass through the catalyst. [1].

In any of these configurations, the integration of SCR into the flue gas stream leads to major works in the

cold area of the boiler. The main advantages of SCR technology are:

- ✓ it can be used for boilers firing a wide variety of fuels;
- ✓ NO<sub>x</sub> conversion does not generate secondary pollutants
- ✓ DeNO<sub>X</sub> may reach 90% efficiency.

Investment costs for SCR system are considerable and dependent on the size of the installation. The investment for a SCR system based on method a) (design, equipment, ducts, pipes, installation, commissioning, without catalyst) can be estimated with  $I = 15 \cdot V^{0,7}$ , million Euro, where V is the flue gas flow in terms of millions m<sup>3</sup>/h. [1]. (Figure 9).

Catalyst cost can be estimated at about 7.000 euros for  $1m^3$  of catalyst. Catalyst quantity requirements are determined based on the flow of flue gas to be treated and on the denitrification efficiency.

For a boiler with flue gas flow of 500.000  $\text{Nm}^3/\text{h}$ , for which is necessary to reduce the NOx concentration from 250 ... 400 mg /  $\text{Nm}^3$  at 100 mg /  $\text{Nm}^3$  the necessary of catalyst needed is approx. 180 ... 200 m<sup>3</sup>. For the same conditions, the specific operating cost is about 1600 ... 2500 euro/ton of reduced NOx (lower value corresponds to higher DeNOx efficiency).



Figure no. 9: SCR investment related to the electric power of the plant.

#### 5. CONCLUSIONS

A brief comparison of the performances of the two technologies, SCR and SNCR, provide us advantages for implementation of SNCR system using urea as reagent. Benefits consist of an investment with lower

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[4] ECOTECH PROIECT București, Audit tehnic și implementarea măsurilor primare privind arderea huilei cost and reduced space for implementation, which compensate the lower denitrification efficiency.

Deva–Mintia power plant performances aim, from environmental point of view, the integration in the requirements of sustainable development.

The combustion process method which is used influences the quantity of generated NOx, these being higher in a pulverized coal boiler, varying according to the type of the burner and to combustion chamber construction [1].

Primary measures applied should lead to sufficiently low  $NO_x$  emission values (below 400 mg/Nm<sup>3</sup>) so that the secondary measures are feasible and cost-effectiveness.

Flue gas recirculation is a method to replace excess combustion air when operating at partial loads.

Through recirculation of flue gas, the flue gas temperature at the outlet is increased, so the efficiency decreases (within 0.7%) but, by reducing the amount of cold air for mill control, there is an increase in the efficiency which compensate for the adverse effects mentioned above.

Among the secondary measures existing in the BAT, the most technically and financially viable which may be used for Deva TPP is the selective non-catalytic reduction technology (SNCR) with  $NH_2$  (urea) as donor agent which provides denitrification efficiencies of approx. 50% (decrease from 400 mg/ Nm<sup>3</sup> to 200 mg/Nm<sup>3</sup>).

The conclusion was based on a comparison of SCR and SNCR technologies resulting in the large difference in favor of SNCR investment and its opportunity, given the fact that the primary measures to reduce  $NO_X$  are already implemented.

The main environmental impact caused by the implementation of these measures will be positive. The direct consequence of the reduction of NOx emissions *is the improvement of air quality in the region*, with beneficial effects on human health, flora and fauna.

The implementation of the project will also generate a positive social impact, both locally, by *supporting existing jobs* due to the possibility of power units continuous operation as a result of compliance with legal provisions and also *by creating new jobs* during implementation works and nationally, by supporting the mining sector which provides fuel used for the power units

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