

## Reduction of Sulfur Oxide Emissions by Means of Mechanical Preparation of Hard Coal

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**ABSTRACT:** A possibility for the reduction of the worldwide increasing emission of sulfur dioxide is the beneficiation of coal with high sulfur content before its utilisation. Reduction of sulfur content by means of processing methods is possible by cleminaling pyrite. The success of this way depends above all on the sulfur distribution in raw coals and on the selected specialised processing methods. In the last decades RAG and DMT carried out extensive tests in laboratory and pilot plants with a large number of different raw coals with high sulfur content. Under our assistance another pilot plant was designed and operated for sulfur reduction of extremely high-sulfurous lignites in Spain. All test results lead to the construction of numerous large-scale preparation plants for sulfur reduction in Europe (Germany, Poland, Spain). Particularly in case of three new preparation plants in Poland, more than 90% of the pyrite contained in the raw material was removed. At the same time, the coal quality as a whole has been improved, which resulted in a reduction of carbon dioxide emissions when the coal is being burnt. To reach the same reduction of sulfur dioxide, the investment for the mechanical separation is only 10% of the costs necessary for a Hue gas desulfurisation plant.

### 1 SULFUR DIOXIDE EMISSION AND SULFUR IN COAL

Sulfur dioxide results primarily from the combustion of sulfur-bearing fuels, smelting of sulfur-bearing metal ores, and industrial processes. Major sulfur dioxide emission sources are power plants, refineries, some types of chemical plants, primary metal smelters, and cement plants. The problem of acid rain is becoming serious as energy consumption

and use of fossil fuels is increasing. Although in most industrialised countries, sulfur emissions are in sharp decline over the past decades, it is in the coal dependent developing countries including China and India, where the concern for the future is potentially great. In the absence of widespread sulfur abatement measures, SO<sub>2</sub> emissions in South and East Asia would triple by 2010.

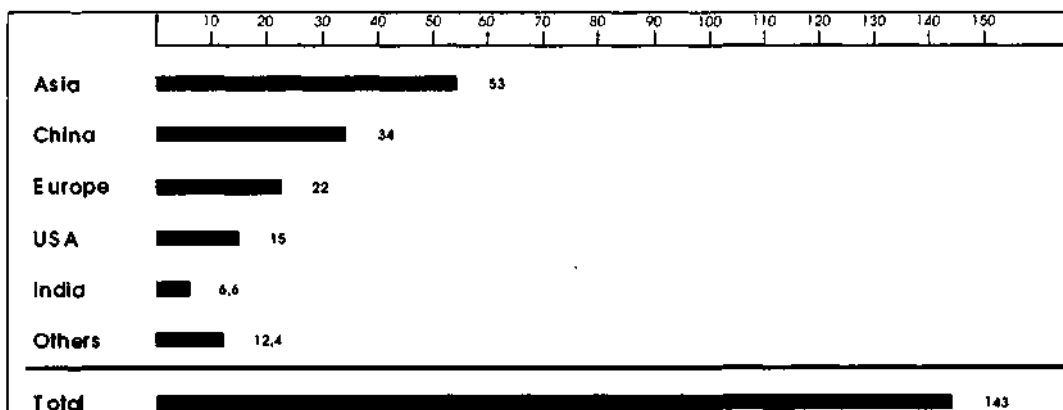


Figure 1. Assessment of global sulfur dioxide emission in year 2000 [10,000,000 t/a]

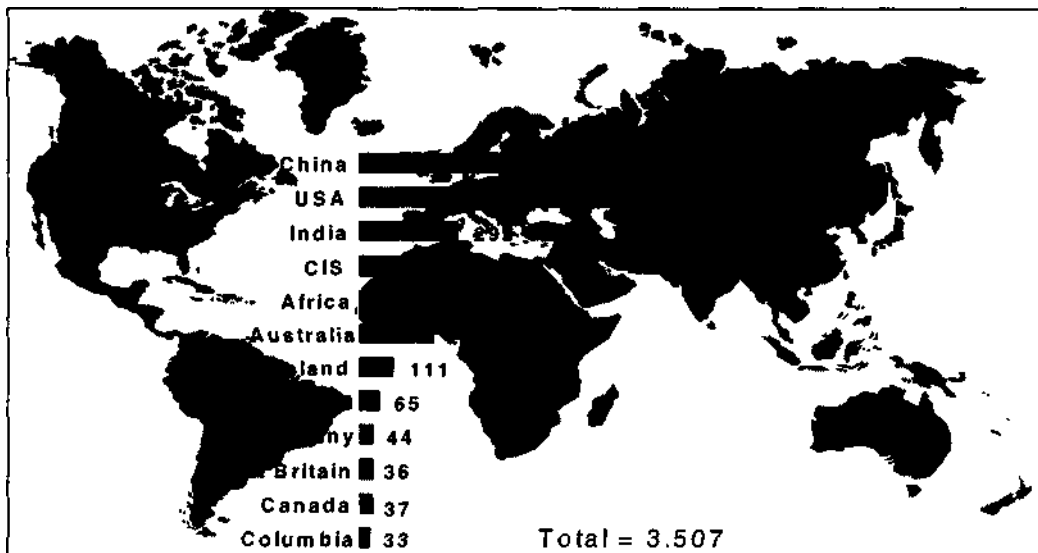


Figure 2. World coal production in year 1999 [1.000.000 t/a]

The People's Republic of China as the world's leading hard coal producer is also the world's leading consumer of coal as primary energy for the generation of electricity. During the maximum production in 1996, China consumed about 1,374 million tonnes of coal which is more than 70 % of the primary energy. Approx. 150 million tonnes of the total coal production, i.e. 12.5 % of the total sales, have medium or high sulfur contents. The average sulfur content of raw coal is 1.72 %.

From the total amount of sulfur dioxide (SO<sub>2</sub>) emissions in the year 1996 of 18.25 million tonnes, about 80 % or 14.6 million tonnes of SO<sub>2</sub> came from coal combustion.

## 2 SULFUR DISTRIBUTION IN RAW COALS

It is difficult to check emissions of sulfur dioxide during burning of coals totally because sulfur is generally present in all the coals either in small or in large proportions,

Seen world-wide, sulfur contents in hard coals and lignite can be from 0,2 %, which is very low, up to extremely high 10%. Usually, coals which are referred to as sulfurous coals contain between 1,3 % and 2,8 % sulfur. These are total sulfur contents.

In general, we find sulfurous coals in two types of formation:

### 2.1 Organically bound sulfur

This form of sulfur occurs in all biological primary substances and is imbedded in the molecular structures of all living organisms (plants, animals, human beings). Coals, which developed from plants, contain organically bound sulfur of about 0,1 % up to 1,2% as regards the dry substance, very rarely they have a higher sulfur content. In case the total sulfur content is higher than the organically bound sulfur, this indicates that there is the second form of sulfur which is

### 2.2 Anorganic Sulfur, Pyrite (FeS<sub>2</sub>)

Pyrite has either been formed during the carbonification process or was incorporated into the coal seam by natural influences. Pyrite can occur in different forms, but it usually occurs in the macrocrystalline (epigenetic) or microcrystalline (syngenetic) form.

Reduction of sulfur content by means of processing methods is only possible by eliminating pyrite. First of all, a few microscopic slides as examples for different pyrite formations are shown:

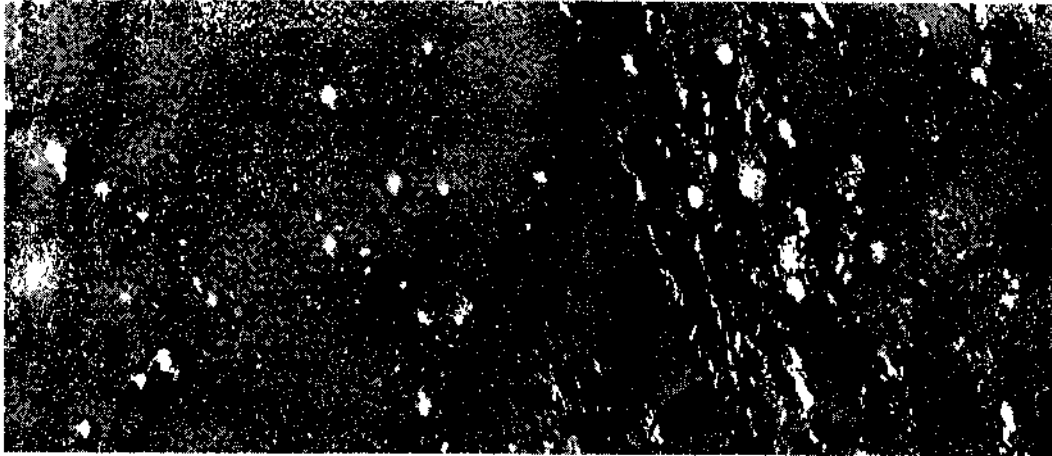


Figure 3 Syngenetic occurrence of pyrite in coal

On the upper photo a typical example of microcrystalline (syngenetic) occurrence of pyrite is shown. This is washed clean coal of a density of 1,3 - 1,4 g/cm<sup>3</sup>. The pyrite particles (white spots on dark background) are very finely distributed and embedded in the coal. On the lower photo a coal grain of a density of 1,4 - 1,5 g/cm<sup>3</sup> is shown. Here pyrite particles are finely distributed as well, but they are also a bit more concentrated than in the photo above. It is obvious that pyrite particles settlement is concentrated near small fractures in the coal grain. If the size of the coal grain is reduced, i.e. using mechanical crushing, the grain would break at the various tractures and so pyrite particles

would also be liberated, which cannot be expected in the case of the photo above.

Figure 4 shows macrocrystalline (epigenetic) occurrence of pyrite in coal grains. The photo above shows a coal particle, i.e. middlings of a density of 1,6 - 1,7 g/cm<sup>3</sup>. The concentration of pyrite particles at the fractures of the coals grain is obvious. A defined impact crushing would liberate the pyrite particles. The lower photo shows a middlings grain with a density of 1,7 - 1,8 g/cm<sup>3</sup> with gross pyrite particles which are surrounded by natural fractures. These pyrite particles could easily be liberated by crushing.

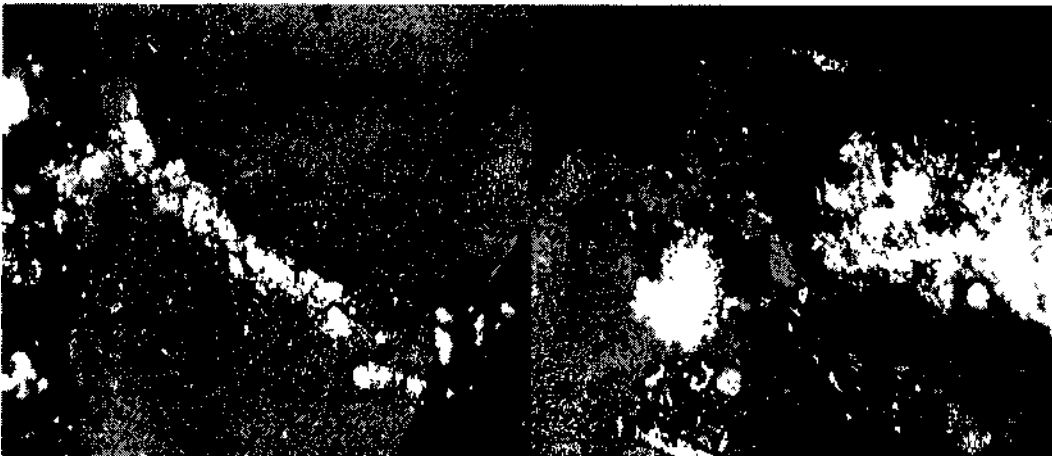


Figure 4 Epigenetic occurrence of pyrite in coal

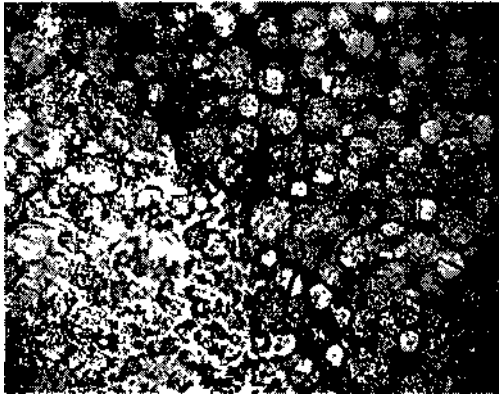


Figure 5. Pyrite in rejects

Figure 5 shows the formation of gross pyrite particles in rejects of a density of more than  $1,8 \text{ g/cm}^3$ . These particles can usually be separated already in a standard processing route.

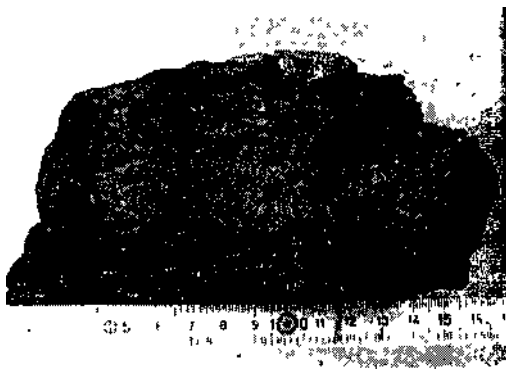


Figure 6 Large particle of pyrite in coal

Figure 6 shows a piece of coal from Shanxi province in China. The pyrite occurs in a separate layer embedded in the coal structure.

In any case, if a reduction of the sulfur content of raw coals is targeted at, first of all the pyrite particles will

### 3 MECHANICAL PROCESSING METHODS OF SULFUROUS RAW COALS

It is been common practice in the most industrialised countries to operate flue gas desulfurisation plants in

coal-fired power stations in order to avoid high sulfur dioxide emissions. But these measures require high capital and operating expenditures.

There is another method to reduce the sulfur content which can be applied even before coal firing in the power stations: the mechanical liberation and subsequent separation of sulfur components by using specialised processing methods. The success of this way of sulfur reduction depends above all on the sulfur distribution in raw coals.

There is a large number of different coal processing methods at the current state of technological development, depending on the particle size distribution, distribution of density and particle form. It depends on the individual raw coal properties which of these methods or which combinations can be chosen for classification, size reduction, separation and dewatering.

In order to develop a adequate flow sheet for sulfur reduction which is favourable under technical as well as under economical aspects, before planning a processing plant one should carry out laboratory tests particularly tailored to the possible processing combinations.

In the last decades RAG and DMT carried out extensive tests in laboratories and technical schools with a large number of different raw coal qualities. A pilot plant with a feeding capacity of 70 t/h was built.

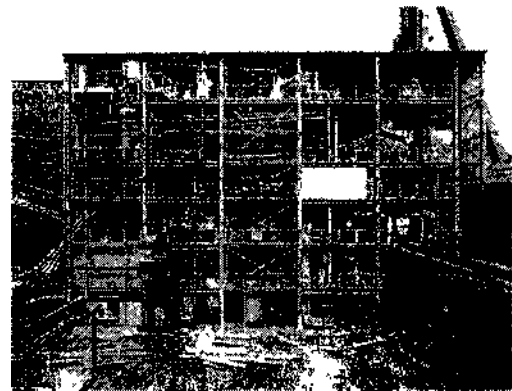


Figure 7. Pilot plant for the desulfurisation of coal

Figure 7 shows this pilot plant shortly before the end of the installation works. It comprises of all relevant processing installations and allows operation of many different processing combinations.

Under the assistance of DMT another pilot plant for the desulfurisation of high-sulfur containing



Figure 9 shows the solid pyrite inclusion of washed clean coal of the density range 1,3 - 1,5 g/cm<sup>3</sup> in the particle size traction 10-3 mm before size reduction

Figure 10 Pyrite inclusion of washed pure coal after size reduction

Washed pure coal of particle size 10-3mm contains a total sulphur content of 1,37% where 1,0% is organic sulphur and 0,37% is pyritic sulphur After size reduction < 3mm and cutting of density fraction +1,8g/cm<sup>3</sup> the total sulphur is 1,17%.

Density range (g/cm <sup>3</sup> )	pure coal size 10-3mm before size reduction		size 10-3mm reduction		pure coal after size reduction		size < 3mm reduction		pure coal after desulphurisation by cutting of density fraction +1,8g/cm <sup>3</sup>	
	mass content fraction %	cum %	fraction %	cum %	mass content fraction %	cum %	fraction %	cum %	Masse %	sulphur cum %
1-3	81	81	1-14	1-14	75	75	1-10	1-10		
13-15	15	96	2-14	1-30	19	94	1-26	1-13		
15-16	2	98	3-18	1-33	2	96	2-42	1-16		
16-18	1	99	3-43	1-35	1	97	2-48	1-17	97	1-17
+ 18	1	100	2-70	1-37	3	100	7-5	1-37		
	100		1-37		100		1-37			

Figure 11 Density structure of washed pure coal before and after size reduction

Figure 10 shows the same coal of figure 9 but after size reduction to less than 3 mm. It is quite obvious here that there are pyrite particles which have been liberated from the coal grain.

Figure 11 shows the density distribution of the washed clean coal shown in figure 10, particle size range of 10-3 mm (the part on the left of the table). There are details on quantities and sulfur contents of the respective density ranges and of the combined density ranges. As you can see here, the total sulfur content of all density fractions is 1,37%. If the size of the coal grain is reduced to below 3 mm the sulfur content in the different density ranges changes compared to the sulfur content in uncrushed material. After the size reduction the sulfur content

was reduced in all density ranges, up to the density range < 1,8 g/cm<sup>3</sup>. The density range > 1,8 g/cm<sup>3</sup> now shows a sulfur content of 7,75% after size reduction whereas before it was 2,7%. If the density range > 1,8 g/cm<sup>3</sup> is separated from the range below 1,8 g/cm<sup>3</sup> with the adequate equipment, a clean coal product with a sulfur content of 1,17% will be received. In this case the yield is 97%, i.e. the losses are about 3%. But these losses are tailings with a high sulfur content. In this connection it has to be mentioned that the organic sulfur in this coal product is about 1,1% so that the clean coal product only contains a proportion of pyrite which could not be separated of about 0,07%.

No.	Country	Colliery	Raw coal feed	Total sulphur content		Yield pure coal	Reduction in t/a	
				raw coal	pure coal,		Sulphur	SO <sub>2</sub>
1	Polen	Janina	4 250 000	1,78*	<1,0*	ca. 70	46 500	93 000
2	Polen	Jaworzno	3 250 000	2,39*	<1,0*	ca. 70	55 400	110 800
3	Polen	Jan Kanty	1 900 000*	1,91*	<1,0*	ca. 70	23 300	46 600
4	Deutschland	Monopol	3 250 000	1,37**	1,1"	ca. 50	26 700	53 400
Remark: Organic sulphur in total			*0,9%					i
			** 1,0%					
Efficiency of pyrite reduction			No.	7				
			1	92%				
			2	95%				
			3	93%				
			4	86%				

Figure 12. Examples for the operating results of mechanical desulfurisation of raw coals in preparation plants

Figure 12 shows examples for the operating results of mechanical desulfurisation of raw coals in different European preparation plants. The design concept of this desulfurisation method has been developed by DMT-Montan Consulting engineers. The plants 1, 2 and 3 are in operation in Poland since several years. Annual capacities are 19 million tonnes, 3.25 and 4.25 million tonnes. The sulfur contents of the ROM of 1.78 %, 2.39 % and 1.91 % can be regarded as high. The content of organic-sulfur which cannot be separated from the coal is in average of about 0.9 %. As a result of the preparation a clean coal product with a total sulfur content of 1 % and clean coal yield was about 70 % is produced. In the past, coal quantities which were as high as the annual capacities of the three plants, were burnt in power stations without being washed. After the preparation plants had started operation, sulfur dioxide emissions of the power plants were considerably reduced, as can be seen in the very right column. In the 3 Polish plants, sulfur reduction was relatively easy because of the properties of the raw material.

Plant No. 4 was a preparation plant in Germany with an annual capacity is 3.25 million tonnes. The total sulfur content was 1.37 %, of which approximately 1 % used to be organic sulfur. Pyrite here was more finely intergrown and its dissociation was more difficult, therefore the total pyrite content in the washed coal product was 1.1 %.

It is remarkable to mention that in case of the Polish coals, more than 90 % of the pyrite was removed (92 %, 95 % and 93 %). In case of the

German coals, the efficiency of the separation was 86 %, which resulted a reduction of sulfur dioxide emissions by 53,400 tonnes per year.

The figures demonstrate that mechanical separation of pyrite from raw coals by means of mechanical processing, can give a significant contribution to the reduction of sulfur dioxide emissions into the atmosphere.

## 5 PROCESS TECHNOLOGIES FOR MECHANICAL REDUCTION OF SULFUR IN RAW COALS

In the following, two examples of many possibilities for mechanical desulfurisation will be described.

Example 1 shows the float & sink analysis of a raw coal, particle size range 100 - 0.1 mm, which represents inclusions of high sulfur contents within the middlings range of 1.45 to 2.20 g/cm<sup>3</sup>. The organic sulfur content in this material is more or less 0.7 %. The sulfur contents of more than 0.7 % are epigenetic pyrite inclusions. The rejects of > 2.20 g/cm<sup>3</sup> contains already liberated pyrite.

To fulfill the required quality of a maximum of 0.8 % sulfur in the saleable product with the highest economical yield, the middlings part of the density range between 1.45 and 2.00 g/cm<sup>3</sup> has to be desulfurised by mechanical processing. The next figure shows a simplified flowsheet as an example for the liberation and separation of pyrite through middlings treatment.

The selected process comprises of different separation steps for three particle size ranges. Due to the fact, that the amount of in the area of near-gravity material is relatively low, the jigging processes for cleaning of the fractions 100-20 mm

and 20-2 mm has been chosen. The size 2-0.1 mm will be cleaned on spiral concentrators. The particle size fraction below 0.1 mm is high in ash content and will be rejected after thickening and press-filtration.

Table I. Gravity distribution

Specific Gravity g / cm <sup>3</sup>	Weight - %	Ash - %	Sulfur • %	Cum. Sulfur - %
-1.35	67.72	8.54	0.74	0.74
1.35-1.45	3.77	13.40	1.20	0.76
1.45- 160	5.15	26.83	1.82	0.84
1 60- 1.80	3.48	40.76	3.26	<b>0.94</b>
1.80-2.00	3.22	53.98	4.19	1.07
2.00-2.20	0.87	55.99	5.47	<b>1.11</b>
+ 2.20	15.79	73.00	9.66	2.46
	<b>100.00</b>	<b>22.84</b>	<b>2.46</b>	

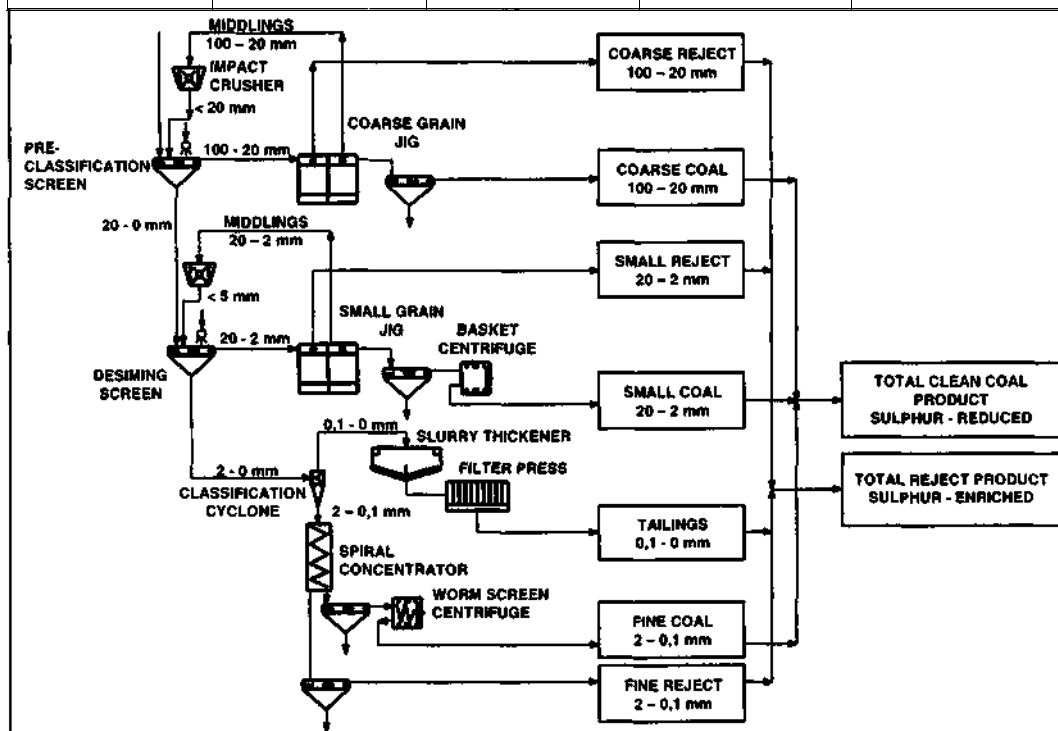


Figure 13. Example 1, Liberalisation & separation of pyrite through middlings treatment

After the jigging process of size 100 - 20 mm there are three products as follows:

Clean coal as part of the saleable product containing a sulfur content of around 0.76 - 0.7S g/cm<sup>3</sup>.

Reject containing some free pyrite, total sulfur content of around 9.5 %.

Middlings containing inclusions of pyrite pieces, total sulfur content is around 3.5 %

There is normally no sales market for the middlings. Dumping of those material can create problems due to self-ignition. As a result the treatment of the middlings with the objective to liberate and separate the pyrite becomes necessary. For this purpose the middlings fraction 100 - 20 mm will be crushed to < 20 mm by impact crushing. The crushed material will be classified by wet screening at 2 mm. The screen overflow, size 20 - 2 mm is processed together with the original ROM fraction 20 - 2 mm on the small grain jig.

By the latter jigging process three products are produced - clean coal, reject and middlings - in similar qualities as the above mentioned coarse

products. Clean coal and rejects are final products. The middlings size 20 - 2 mm will be crushed again by impact crushing to < 5 mm. The crushed material will be classified by wet screening at 2 mm. The fraction 5 - 2 mm flows for rewashing back to the feed of the small grain jig. All original raw coal and crushed middlings < 2 mm will be deslimed at 0.1 mm and then cleaned in spiral separators. Products of the spiral separators are desulfured clean coal and sulfur-enriched rejects.

At the end of the process all the clean coal products together will be in a saleable quality of the required sulfur content of < 0.8 %.

Example 2 shows the float & sink analysis of a raw coal, particle size range 100 - 0.1 mm, which represents inclusions of high sulfur contents within the middlings range of 1.45 to 2.20 g/cm<sup>3</sup> and in the clean coal range of 1.35 to 1.45 g/cm<sup>3</sup>. The organic-sulfur content in this materials is more or less 0.8 %. The sulfur contents of more than 0.8 % are epigenetic pyrite inclusions. The rejects of > 2.20 g/cm<sup>3</sup> contain already liberated pyrite.

Table 2 Gravity distribution 2

Specific Gravity g/ cm <sup>3</sup>	Weight - %	Ash - %	Sulfur - %	Cum. Sulfur - %
- 1.35	35.88	7.29	0.88	0.88
1.35 - 1.45	33.69	9.06	1.01	0.95
1.45 - 1.60	3.77	29.37	3.20	1.06
1.60- 1.80	5.05	44.11	4.14	1.26
1.80-2.00	2.86	52.66	7.64	1.48
2.00 - 2.20	1.61	53.03	6.83	1.59
+ 2.20	17.14	76.06	8.81	2.83
	<b>100.00</b>	<b>24.40</b>	<b>2.83</b>	

To fulfill the required quality of a maximum of 0.95 % sulfur content in the saleable coal product with the highest economical yield, the middlings between 1.45 and 2.00 g/cm<sup>3</sup> including the clean coal part of 1.35 to 1.45 g/cm<sup>3</sup> has to be desulfurised by mechanical treatment. The next figure shows a simplified flowsheet as an example for the liberation and separation of pyrite through treatment of this material.

The selected process comprises again of different separation steps for three particle size ranges. The

first step of the raw coal treatment of size 100 - 0 mm is the classification by screening at 40 mm. The coarse raw coal, size 100-40 mm contains the major part of the subsequently produced rejects. In a deshaling plant, consisting of a dense medium bath separator, rejects and coarse pyrite will be separated. The coal and middlings with pyrite inclusions will be crushed to < 40 mm by impact crushing. These crushed material and the original raw coal size 40 - 0 mm runs to desliming screening for classification at 2 mm.



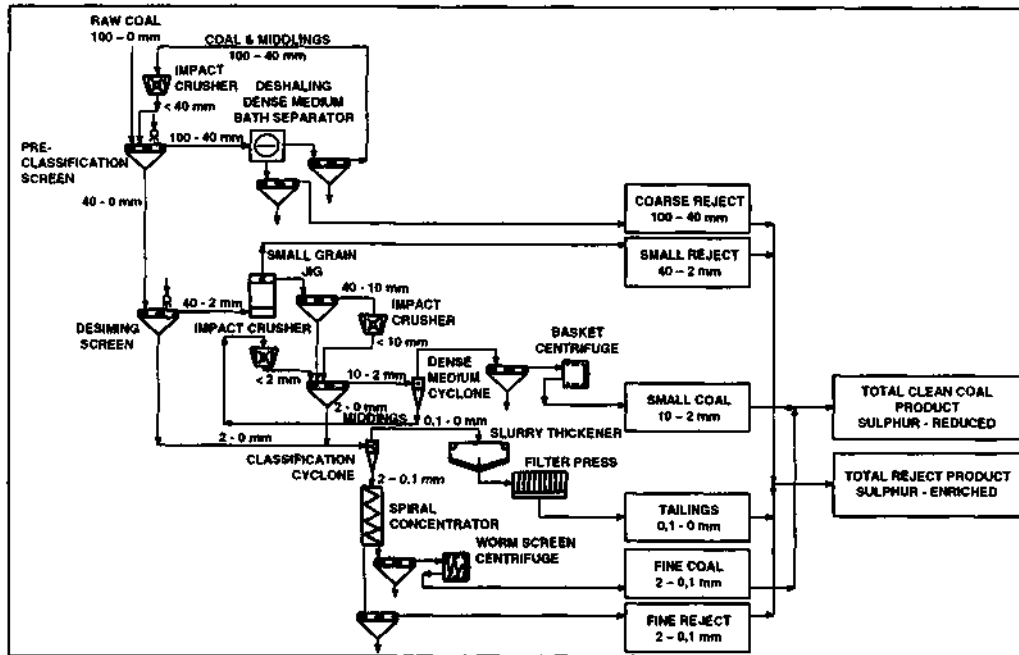


Figure 14. Example 2, Liberalisation & separation of pyrite through middlings & partly coal treatment

Size 40-2 mm will be washed in a two-product jig, for separation of reject. The jig overflow, coal and middlings with pyrite inclusions will be sreedewatered and subsequently crushed to < 10 mm by impact crushing. These crushed material 10-0 mm runs to desliming screening for classification at 2 mm.

Due to the fact, that the coal contains a lot of near-gravity material the grain size range of 10-2 mm will be washed in two-product dense medium cyclones, for separation of low-sulfur clean coal with a density < 1.35 g/cm<sup>3</sup>. This material is part of the saleable coal product. Cyclone underflows are middlings and clean coal with densities higher than 1.15 g/cm<sup>3</sup>, containing inclusions of pyrite. The underflows will be crushed as well to < 2 mm by impact crushing. These crushed material and the original raw coal size 2-0 mm will be deslimed at the same time at 0.1 mm in classification cyclones. The grain size fraction 2-0.1 mm will be washed on spiral concentrators. Products of the spiral separators are desulphered clean coal and sulfur-enriched reject.

Size 0.1 mm is high in ash content and will be rejected after thickening and press-filtration

At the end of the process all the produced clean coal sizes together will be in a saleable quality of the required sulfur content of lower than 0.95 %.

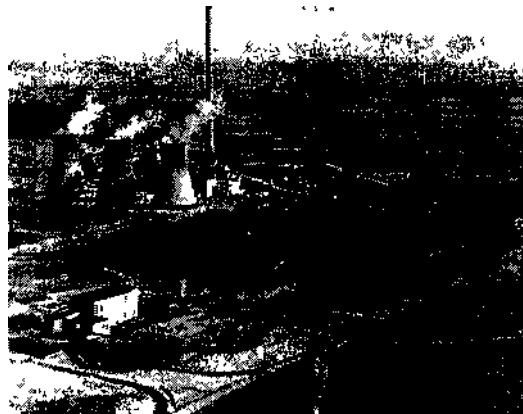


Figure 15. Preparation plant for sulfur reduction

Figure 15 shows a preparation plant for sulfur reduction of Lignite in Spain. This plant was erected

lignite has been build and operated for the Energy-Suppei ENDESA in Spain. The results of all the test and investigations have formed the basis for the design and construction of several industrial scale preparation plants for the mechanical desulfuration



Figure 8. Pilot plant for the desulfurisation of lignite

On Figure 8 the pilot plant which was designed and operated solely for sulfur reduction of extremely high-sulfurous lignites in Spain is shown. The preparation plant is in the right part of the photo and in the background you see a coal-fired power station. This adjacency gave the opportunity to test the coal products of the pilot plant in the power plant and to test their impact on the combustion process.

Our engineers were not only involved and played a significant role in the design, development and construction of those plants, but also during the performance of the individual test series. The know-how and experience gained in these projects today contribute very much to construction planning and remodelling designs of preparation plants aimed at reducing sulfur contents.

#### 4 RESULTS OF SULFUR REDUCTION IN COAL PREPARATION PLANTS

A few preliminary remarks:

The reduction of sulfur content by means of mechanical processing can only be realised by eliminating pyrite. The degree of pyrite separation which is possible depends on:

- the particle size and density distribution of the pyrite and the degree of intergrowth

of raw coal in Germany, Spain and Poland. Specially in the newer plants in Poland, more than 90 % of the pyrite contained in the raw material could have been removed.

- the occurrence of pyrite which is usually macrocrystalline (epigenetic) or microcrystalline (syngenetic)
- the crushing equipment applied
- the separation density and efficiency of the separating equipment.

The processing costs rise more than proportionally with decreasing particle size because of

- increasing financial costs
- increasing yield losses
- additional expenditure for dewatering and thermal drying, if necessary

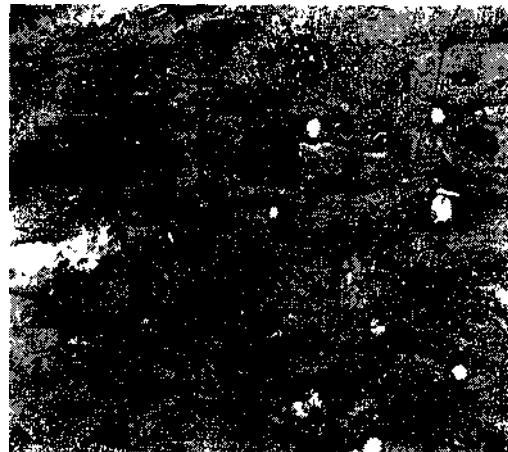


Figure 9. Pyrite inclusion of washed pure coal before size reduction

What is important for the degree of liberation of pyrite is the sulfur distribution in the different density ranges before and after size reduction of the coal. Have a look at this example:

next to a power plant (capacity 3 x 350 MW), as the raw Lignite come from different mines with different production capacities.

Last but not least it should be mentioned that the separated pyrite can be upgraded mechanically and serve as an important secondary raw material for the production of sulfuric acid. According to Chinese experts there is a need for this product in the Chinese industry.

Apart from the mechanical sulfur reduction, the processing in preparation plants lead to a reduction

in ash content by separating the waste rock, which originally is the aim of such a plant. This results in a reduced energy consumption for the transport of coals to the power station and in a higher combustion efficiency, which all in all reduces carbon dioxide emissions as well. Statements about the quantity cannot be made until we know something about the coal qualities used in the power stations and about the distances between preparation plant and power station.

