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Waste Materials in Construction: Putting Theory into Practice
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STATE OF THE ART OF GYPSUM RECOVERY FOR A SPANISH POWER PLANT

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1. - PREAMBLE

The problems of pollution of water, soil and atmosphere are increasingly considered, by the planning and development policies of most of the states of the world. Nowadays, the technology and management of the environment present a great growth, and receive important amounts of public fund investments from the European Union. In the past, most environmental policies have had a corrective character (who contaminates must pay). Afterwards, the trend was modified and legislation aimed to preventing and avoiding the damage; was the Environmental Impact Assessment system. The different laws limiting atmospheric emissions and the EIA Directive are respectively under these two points of view. The "**Fifth Action Program in Environment Matter of the European Union**" actually is based on the correction, prevention and on the better participation and planing.

Atmospherical pollution was one of the first environmental problems approached by the European Community, when in the 1970s Council Directive (*70/220/EEC*) on "**Approximation between the State members related to the measures to take anti-pollution of the air by gases originating from vehicle motors**" was approved followed by that regulating the vehicle emissions (*72/306; 74/290/EEC*).

Fuel power plants are probably the most thermo-efficient system to convert combustion energy. The size and design of a combustion power plant are created to obtain the maximum rentability of the chemical reactions. Other thermic machines like motor cars, etc., are designed to produce a maximum rate of H.P./Engine-weight. An special case occurs when the objectives are the utilization of a specific fuel: industrial or agricultural wastes, or a particular fuel with low calorific power such as lignites.

After the petroleum crisis in the 70's (between 1970s/74 the price of the oil barrel multiplied by six) many coal power plants were constructed. Thus, many power plants to use low power and high sulphured coals were built. Environmental protection was not then considered as a priority criteria; only several decades later the consequences offered new criteria.

Nowadays technologies to correct pollution are available, and the social body has accepted the relevance of some new environmental protection values. Thus, "**clean production**" should be presumed more expensive than the "**dirty-one**". In any situation, the decision of the "**cleanliness**" degree that will be demanded to any technology, will always be a political decision, in response to some social values.

2. - OBJECTIVE OF THE PRESENT STUDY

This study developed after the requirement from Technical Management of "**CENTRAL TERMICA DE ANDORRA**", belonging to *ENDESA (Spanish Public Enterprise)*. The program titled "**Project Teruel**" represents important investments in the factory, with the purpose of reducing the emissions, especially of sulphur oxides, coming from the combustion of coals with high sulfur contents.

The construction of the new desulphuration plant has already begun. The project anticipates a reduction of current emissions by 90%. The technology consists in the utilization of 700.000 tons of limestone ($CaCO_3$) per year with the production of approximately 1.000.000 tons of gypsum ($CaSO_4.2H_2O$).

As it happens with other residues produced in large bulks, the plasters generated in the desulphuration process cause a management problem; however, they also possess a potential profitability if an adequate market for them is found.

3. - ANTECEDENTS AND HISTORY

Enormous investments are required to reduce the sulfur emissions produced from coal power plants. In the United Kingdom, only one company has done the necessary investments to obtain a satisfactory reduction in the emissions of sulphur dioxide; this was accomplished when the factory was a public enterprise. In Spain only a public company ENDESA has decided for cleaner combustion after a hard public opinion struggle against the consequences of air pollution. All this suggests that funds are more easily obtained when political factors are presents.

In the United States, the protection of the environment has been promoted exclusively through the pressure of the social movements in the sixties. The market and the cost of energy has not stopped in the United States the enaction of laws for reducing the pollutant emissions. Title IV of the "**Amendment to the Clean Air Act**" of 1990 has imposed new limitations to the emission of SO_2 and NO_x from fuel power plants, which must be adapted to new fuels or procedures (*Locker, 93; South & Bailey, 95; Barsotti & Kalyonku, 95*).

In Japan, desulphuration gypsum has been employed as an alternative material to the traditional chemical plaster since the 70's (*Nagata, 1995*). The Japanese anti-pollution laws and measures against the emission of sulphur oxides date of 1968; the first desulphuration plants are running since 1972. Other desulphuration facilities were installed between 1972-1977 in 18 central thermal and in 18 metallurgical industries, in addition to in other 13 industries.

In Canada (*Luckevich, 1993*) desulfogypsum are an alternative to mineral plaster began to be applied in the 80's. The cited author thinks that "**... in North America .. there is much to learn from the European and Asian experience insofar as the by-products utilization**".

In Europe, during the last years, there has been an important increase in the non-polluting legislation , within in the European Union and in the OECD (*Franz Wirsching, Rolf Hüller & Rainer OlejnikJ, 1994*) . The Residues Directive (*91/156/EEC, of 18 March 1991*) contains a catalogue with by-products that are defined as: "**substances or objects described in the Annex I and that are referred to waste materials**". In the OECD Directive (*Control of Transborder Residue Movements intended for Recovery: C(92)39*) desulphogypsum is considered as a material included in the " **green list**" (non-dangerous). German legislation, which included initially plasters Fuel Gas Desulphuration (FGD) as residues, now considers it as a "product".

In the Netherlands, lignite power plants equipped with decontamination systems have introduced in the market important quantities of gypsum plaster (400.000 tons, *Moonen, 1991*). As consequence of mineral resources shortage, the Dutch administration favors the research and development of technologies that provide construction materials as recovery products (*Winden, Zwan, Zeilmaker 1991*).

The experience with desulphogypsum plasters is not extended to all the countries of the European Union. Experimental work with ashes and plasters in Italy, (*Gera, Mancini, Mecchia, Sarrocco & Scheneider 1991*) appeals to the use of imported desulphogypsum plasters because there are not power plants in this country with systems of desulphuration.

In Sweden (*Timm, 1991*) 52 million tons of wastes are produced annually. 20 million are recovered,either as energetic material or as by-products. Within those, 500.000 tons are combustion wastes, including plasters. Environmental protection laws do not consider including the products of decontamination of gaseous emissions as dangerous materials, but the "**Swedish Environmental Protection Agency**" has developed environmental protection procedures, setting very strict limits, mainly to reduce the emissions that had caused the destruction of thousands of small lakes.

4. - QUALITY OF THE MATERIAL

Wirsching (93, 95) defines the plaster *FGD* as:

> **"The plaster produced in plants of desulphuration (gypsum FGD or desulphogypsum) is calcium sulfate dihydrate very pure, wet, crystalline, finely granulated. It is produced specifically through processes of desulphuration in those which, through injection of calcium carbonate to the scrubbers, is therein after submitted to oxidation processes and dehydrated. The plaster FGD is produced in the fuel power plant under criteria of quality for that material".**

4.1. Generation of the gypsum *FGD*.

The process of desulphuration is based on the following reactions:

$$SO_2 + CaCO_3 + \frac{1}{2}H_2O \rightarrow CaSO_3.\frac{1}{2}H_2O + CO_2$$

Of this reaction results that each 64 grams of SO_2 will consume 100 grams of calcium carbonate $CaCO_3$, but will issue to the atmosphere 44 grams of carbon dioxide CO_2. The calcium sulfite hemihydrated is submitted to forced oxidation in all the modern systems to produce calcium sulfate dihydrate:

$$CaSO_3.\frac{1}{2}H_2O + SO_2 + H_2O \rightarrow Ca(HSO_3)_2 + \frac{1}{2}H_2O$$

$$Ca(HSO_3)_2 + O_2 + 2H_2O \rightarrow CaSO_4.2H_2O + H_2SO_4$$

$$CaCO_3 + H_2SO_4 + H_2O \rightarrow CaSO_4 2H_2O + CO_2$$

That globally:

$$2CaSO_3.\frac{1}{2}H_2O + O_2 + 3H_2O \rightarrow 2CaSO_4.2H_2O$$

In the first step, the insoluble calcium sulfite reacts with more sulphur dioxide producing calcium bisulfite (soluble); this reacts spontaneously with the atmospherical oxygen, producing calcium sulfate dihydrate and also is produced more sulfate, by direct reaction of calcium carbonate with the sulfuric acid, that is generated in the oxidation reaction.

4.2. Properties

The "**Decision of the Council of the OECD over the Transborder Control of Residues intended for Recovery Operations**" *(C(92)39)* establishes some lists (green, amber and red). In principle, the gypsum FGD were included in the green list, of where it were eliminated to instances of "**EUROGYPSUM**", that requested that it would be recognized its character of "**product**". This request was based on the following considerations:

The gypsum FGD, as a matter outweighs secondary, it is used by European industry of the plaster.

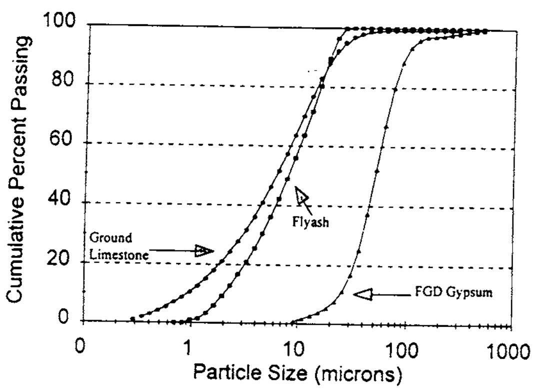
Its economic value is comparable to the natural plaster and its price is conditioned by transportation freight

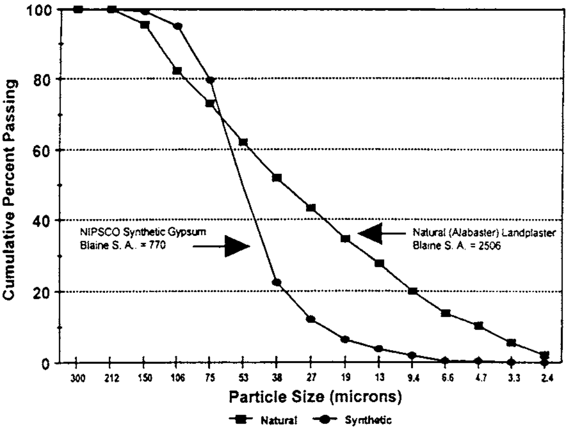
The gypsum FGD is transported by highway, railway or load ships without need of special cautions .

In the German legislation (*Federal German Emission Protection Law)* only are considered as residues, the materials that they can not be reutilized and they must be rejected. In the United Kingdom, France, Belgium, Holland, Denmark and Austria the gypsum FGD has the character of "**product**".

4.3. Granulometric distribution

The grain of the natural or artificial gypsum present important differences. The *figures 1 and 2* (*Glasscock, 93*) allow to compare the different size distributions of the natural and artificial plasters. The first is generated by grinding of mineral chunks (*3" to 6"*) and the second by the directly formed crystals during the desulphuration process into the scrubber. These last have been formed in an stirred environment, where the presence of numerous condensation nucleus generate grains with very uniform shape and size.





4.4. Normative

The parameters that indicate the quality of the desulphogypsum, because its utilization as by-product in European industry of the plaster are withdrawals in following Table I (*Wirsching, 1993*). The "**United States Gypsum Company**" (*USG*) has a long experience in the characterization and the marketing of the american plasters; It establishes a series of characteristics for the artificial plasters,(*Henkels, Gaynor, 1995*). *Ojanpera and Cabbage (1993)* (see Table II) offer a comparison between natural plasters and desulphogypsum in the one which are emphasized important advantages of these last with respect to several of the characteristic parameters.

Table I. Quality criteria for Gypsum FGD

Parameter	% weight
Water content	< 10
$CaSO_4.2H_2O$	> 95
Soluble MgO	< 0.1
Cl^-	< 0.01
Na_2O	< 0.06
SO_2	< 0.25
pH	5-9
Color	white
Smell	neutral
Toxicity	no-toxic

Table II. Quality of the Gypsum FGD and natural

Propriety	Gypsum FGD	Natural Gypsum
Free water %	6-10	0.2-3
Combinated water %	19-21	19-21
Ca Mg Carbonates %	0-5	0.15-1.6
$CaSO_4.nH_2O$ %	91-99.8	97-98.5
SiO_2 ppm	1700-7000	5000-6500
Al_2O_3 ppm	1000-5000	<5000
Fe_2O_3 ppm	140-1300	600-4200
MgO ppm	100-800	5000-14000
Na + K ppm	300-1400	200-500
SO_2 ppm	10-700	0
Chloride ppm	10-100	70-80

5. WORLD WIDE PRODUCTION (Unpredictable trends)

Different evaluations have been done to estimate the plasters total of desulphogypsum currently produced and reutilized in each one of the different countries, and the world over. Any estimation would be a provisional figure, as consequence of the figures of consumption is conditioned by the following circumstances:

1. The petroleum crisis and its spectacular rise in costs in the seventies was the cause of the implementation of the utilization of sources of fossil fuels, that they had been rejected by its poor energetic properties (low power) and environmental (high contents in sulphur).

2. The prices readjustment of the liquid fuel has not made to reduce energy demands originating from the coal. Nuclear energy has been submitted to critical on the part of the public opinion, in such a way that the construction projects of new nuclear plants has been reduced in the last years.

3. The Rio de Janeiro United Nations Conference of 1992 established a series of commitments, about the limitations in the pollutant emissions to the atmosphere, above all centered in the emissions of CO_2, hydrocarbons and substances related with "green house effect" and in the depletion of the ozone layer.

4. Without may have been object of international procedures of that range, many countries in those which acts the free market have developed a vast application of desulphuration systems that in most cases produce a momentary "**excess**" of gypsum of good quality in the by-products market.

5. The technology of clean combustion is an elaborated system that, as of high initial investments allows to reach materials of uniform quality.

6. REUSE OF GYPSUM FROM DESULPHURATION PROCESS
6.1. Gypsum consumption

For better understand the tendencies on the production and reutilization of desulphogypsum we can refer the situation in EEUU and Japan. The table III shows the production, imports and consume of gypsum in EEUU. *Barsoty & Kalyoncu (1995)* estimate proximately 20.000.000 tons the production of FGD in EEUU in 1993, before the enaction of IV amendment in the Clean Air Act.

Table III: Production and reuse of Combustion Products in EEUU

	Fly Ash	**Bottom Ash**	**Slag Furnace**	**GypsumF GD**
Production ton	43,4	12,8	5,6	18,4
Use ton	9,5	3,8	3,1	1,0
%	22	30	55	6

In Japan desulphogypsum is the most important secondary material used in construction *(Makansi & Ellison, 1993)* (Fig. II). Because they are not mineral exploitation and the absence of land to be used like damping area, the policy of wastes is in this country especially strict. The consume gypsum demand has been estimated in 9,3 millions of tons *(Nagata, 1995)*. A total 25 % are gypsum from power plants desulphuration what is recovered in 100%. The 43 % is imported from Tahiland and Mexico.

6.2. Applications

Very diverse applications are mentioned in the technical literature. three different type of refereed applications can be distinguished:

a) Applications with higher added value. In this reutilization the gypsum is transformed in building materials (pre-fabrication) to be applied like a finished product
Board walls, Blocks for construction; Shape-molded elements; Artificial reefs.

b) Use as prime material. The gypsum can be used, directly or after a semifinished transformation in a material to be applied in building or civil works

Mortars; Artificial gravel; Additive or cement filler for concrete; Special cement; Self-leveling floors material; Concrete for pipelines; Mortar for mining;

c) Use in a big amount of quantities, to be applied in civil engineering and environmental purposes. These are the applications were an high volume of secondary materials are used.

Material of mines landfill; Residues stabilization ; Material for highway bases; Soil stabilization

7. - COMMERCIAL ASPECTS

In other estimation *Makansi & Ellison (1993)* evaluate in 27 million of tons of plaster used per year in EEUU. A part is imported from Canada and Mexico (between 30% and 40%). In Canada, the annual consumption is about 3.5 million of per year tons. In 1986 the *FOB* price of the natural mine gypsum was of approximately 8 $/ton (according to the Mineral Industry Survey of the Bureau of Mines, EEUU 1977). In consequence of this low price it is difficult the introduction of artificial gypsum produced in very expensive installations.

Until the enaction of the "**IV amendment to the Clean Air Act**" in EEUU, the preoccupation for obtaining desulphogypsum in a commercial quality was constituting only a marginal phenomenon (*Locker, 1993*). Now, the things have changed in this country; the companies consuming the by-product demand specifications of quality very rigorous, the impurities presence, color, etc. will have to be absolutely guaranteed in the product. The price and the difficulty of the transportation is the factor more unfavorable.

In Europe the principal consumers countries of synthetic gypsum are Germany, Holland and Austria (*Makansi & Ellison, 1990*). The clean combustion technology, originated from Germany and Japan has been developed, until obtaining plasters that fulfill the specifications demanded by the consumers. Nevertheless, the adjustment of the European markets, to the substitution of the mineral plasters by the by-products, it is very difficult and it has obligated to improve the quality.

In Spain, the natural gypsum market consumes more than 2.5 millions tons every year in a disperse little manufacture industries. The future introduction of more than one millions tons of gypsum FGD will represent a problematic management.

8. FINAL CONSIDERATIONS

We can analyze, some of the common characteristics defining this situation.

a) The industries consuming gypsum are not new installations; they are decades on a traditional activity, consuming a cheap prime material. Nevertheless, in several countries the activity is based on imported minerals (EEUU, Japan, Holland), although the plaster constitutes a relatively small part of the production costs (*Makansi & Ellison, 1990*); (*Henkels, Gaynor & Garceau, 1993*); (*Kuntze, 1993*); (*Nagata, 1995*).

b) Frequently, the location of transformation gypsum industry are where the gypsum mines are abundant and inexpensive of extracting. The products that are manufactured in that industry are not expensive, the most of them devoted to construction. The plaster products can deal affected for uncontinuous demands (campaigns) (*Locker, 95*).

c) The reason for assuming the clean combustion is to protect the environment, taken as a **political decision.** The initiative has begun in the countries, in those which the free competence is an economic and ideological paradigm. The market occurred for those by-products has been appeared, after to have generated it in a non return way. We can interpret the situation as a stages succession:

1) To clean the atmosphere produces pollution by solid residue; management of these is viable and known, before of the beginning of its appearance

2) To sell the by-product can produce benefits

3) To put in the market the by-product could have indirect environmental advantages, as the avoiding of the opening of new mines, energetic saving, etc. (*Wirsching, 1993*); (*Wirsching, 1995*); (*South & Bailey, 1995*); (*Armour, 1995*); (*Jensen, Freimut & Wetegrove, 1995*)

d) Industry consuming gypsum does not accept unconditionally, the presence of a new product, not even to "**zero cost**". The improvement of quality of the produced plasters has gone increasing, throughout the years in which it has been developed the technique, and yet today it continues improving. The exigency of quality for the product, composition and regularity as supply are an exigible condition by all the gypsum consumers. The acceptance of the alternative materials, as matters outweigh secondary by industry is not only an economic problem, but cultural (*Harness & Shang, 1993*); (*Barsotti & Kalyoncu, 1995*); (*Takshina, Ukawa & Kimura, 1995*).

e) On important problem for the management of synthetic gypsum is the "**diversity of scale**" between producer and consumers. For the gypsum producing companies, the adaptation to some requirements of a "**small consumer**" constitute an important restriction. As consequence of this diversity of productions and requirements, they have come out on the market intermediary companies (*Feeney & Novak, 1993*); (*Makansi & Ellison, 1990*); (*Jurkowitsch & Hüller, 1995*); (*Lani, College & Babu, 1995*); (*Welliver, Roth & Colijin, 1995*); (*Larrimore, 1995*).

f) The production of the by-product in the power plant is uniform and will be produced every day, with or without market. The various markets existence in volume, pace of consumption and geographical dispersion would compel, in any event, to have a system stocks storage, that deadens the fluctuations of demand. The capacity of the deposits of those stocks will have to anticipate, the situation of "**not reuse**" for unlimited periods of time (*Armour, 1995*).

9. CONCLUSIONS

As a conclusion: The gypsum FGD production generates a big amount of the by-product. The reuse of this material does not present technical problems in the most of his applications. Nevertheless the activity consuming the greatest volume of materials, **the civil engineering**, in public works, appears like the most shy consumer. A short number of the papers consulted describe experiences consuming a big volume of material in banks, highway bases and works of this character. In this case it is tended furthermore to propose, the mixtures employment of other by-products, also generated in the power plant, as bottom and fly ashes. In most of these experiences are presented promising results, but always is necessary the technological research at appropriate scale (*Golden & Saylak, 1993*); (*Clarke & Smith 1991 and Clarke 1992, 1993*); (*Bhat & Rogers, 1995*).

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