Technological Trends in the Use of Natural Marble Residue in the Cement Portland Production from Prospecting in Patent Documents

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Abstract
The civil construction industry affects the environment mainly due to the high generation of waste and extraction of natural raw materials. However, this industry is also responsible for absorbing a large part of the waste, not only generated by itself, but also those generated in other production cycles. These residues are often reused as an alternative raw material, after some treatment. The use of ornamental stone cutting waste has already been studied by many authors, and it is a technically viable alternative in cement matrix products for civil construction. Thus, this technological prospection work analyzed the technological trends regarding the use of marble waste in cement production, based on the registration of patents in recent years. For this, a search was carried out in the Brazilian Nacional Institute of Intellectual Property (INPI) and Dewert Innovation Index (DII) databases, in April 2021, with no period limitation. The main documents were found in the DII database, and the results showed that among the countries that filed a patent on this subject, China is the country with the highest number of filings, with the majority of these filings occurring between 2014 and 2020, suggesting a reflection of the current interest in ways to give an adequate destination to ornamental stone waste.
Key-words: Clinker, Portland Cement, Marble Waste, Patent Indicators.

1. Introduction

Civil construction is one of the largest consumers of waste from other industries, contributing to reduce the negative effects of the disposal of these materials into the environment. The diversity of raw materials that can be used in civil construction allows a large number of studies to be developed on the subject (MATTA et al., 2013; REHMAN, AHMAD and RASHID, 2020). In fact, research on the incorporation of ornamental stone residues in concrete, mortar, and ceramics is constantly carried out due to the large volume and impact of waste generated by the ornamental stone industry.

In a study conducted by Alyamaç, Ghafari and Ince (2017), in which a data survey on the topic was developed, it was presented that 70% of extracted marble blocks are transformed into waste. Rodrigues, Brito and Sardinha (2015) indicate that this waste reaches between 80% and 90% of the total volume of the block, showing the need for an adequate destination for this material. Therefore, in addition to the remaining blocks, there is also the generation of abrasive sludge, which, after drying, releases a fine inorganic powder, increasing global environmental liabilities (SOUZA, PINHEIRO and HOLANDA, 2013).

The fine residue, if properly treated and combined with compatible chemical composition, could be used as raw material by the cement industry (MARTÍNEZ-MARTÍNEZ et al., 2019), replacing limestone. If feasible, this action would assist in decreasing limestone consumption, which corresponds to about 90% of the raw material for cement production, whose extraction impacts on topography, soil, and, sometimes, on basins and ecosystems (NATIONAL CONFEDERATION OF INDUSTRY, 2017).

Some studies address the incorporation of marble dust residues in cement as an alternative to replacing part of the limestone (raw material) or as an addition, proving the potential for its use. However, there is no significant volume of patent filing on the subject, which may indicate a lack of interest in the sector or difficulty in obtaining a patent for this type of product. Thus, the objective of this work was to evaluate the technological trends in this field from the search for patent filings in recent years on the subject, making it possible, therefore, to verify the countries most engaged in this topic and the greatest interests in it.
2. Methodology

For the technological prospecting, the patent documents related to the production of cement with the use of marble residues available in the Brazilian National Institute of Intellectual Property (INPI) and Dewert Innovation Index (DII) databases were analyzed, in April 2021, with no period limitation. These databases were selected due to their accessibility and reliability of available data. In addition, as these are national (INPI) and international (DII) databases, the search would cover a greater number of filing countries.

The first search (01-INPI) was at the INPI database using the theme “clinker with marble residue” in the title field. As no patent documents on the topic were found, the search was extended for “cement with marble” in the abstract field (02-INPI), thus obtaining one document.

In the DII, the first search (01-DII) used the theme ALLD=(Clinker) AND ALLD=(Marble ADJ Waste), with which a single patent document was identified. Expanding the use of marble for cement production (02-DII) with the theme ALLD=(Cement) AND ALLD= (Marble ADJ Waste), 66 patent documents were found, one of which was already obtained in search 01-DII. Finally, considering the theme ALLD=(Cement) AND ALLD=(Marble) AND ALLD=(Waste) AND ALLD=(Clinker) (03-DII), the listed patent documents reduced to 24. As with 02-DII, the patent document from search 01-DII was also pointed out in search 03-DII.

Given the low volume of patents found on the subject in the chosen databases, it was decided not to use specific software for data processing, but to evaluate the abstracts of each document. To maintain the focus of this research, the document found in the 02-INPI survey was not considered for dealing with a subject related to civil construction, the production of tiles, but outside the scope of this study. The same happened with 30 others that concerned only the use of artificial marble. However, patent documents relating to products whose composition contained some artificial marble residues and cement or clinker were evaluated, including those whose end uses differ from Portland cement production.

3. Results and Discussions

From the results obtained in patent research, it was possible to identify the main depositing countries, the number of deposits over the years and even the trend of sub-themes regarding the use of marble waste in cement production, as shown in the Figure 1.
Of the remaining 59 patent documents, a concentration of deposits by China was noticed, with 50 records in the last 10 years, as can be seen in Figure 2. The performance of this country suggests a strong interest in the search for environmentally correct destinations for marble waste, which is justified by the 280 million tons of remnants produced in 2015 (CHINA STONE MATERIAL ASSOCIATION, 2016 apud Li et al., 2019; HEBHOUB et al., 2011).

Source: The authors (2021)
Despite the high number of papers published on the subject by Saudi Arabia, Egypt, Portugal and Brazil, countries that, like China, have a large amount of remnants of natural marble, there is no registration of patents from these countries. India and Turkey are also references in the publication of papers, however, they have little participation in filing patents, having registered 2 and 1 documents, respectively.

The registration of the first patent filing found on the use of natural marble waste in cement production dates back to 1982, made by a Canadian research institute. In 1989, there is a second registration, this time by the former Soviet Union, and in 2001 a third, from Japan. Only in 2014, the number of patents starts to grow, with most of the filings taking place after 2016, afterward a fall in 2015, as shown in Figure 3.

The increase in the number of patent registrations after 2014 may be a reflection of the interest in ways to properly dispose of industrial solid waste, in line with international environmental treaties, the latest being Rio+20, of 2012, and the Paris Agreement, of 2015. To comply with the precepts of this last agreement, for example, the Technological ROADMAP for Cement was developed, with the objective is helping to reduce the emission of carbon dioxide by the cement industry (NATIONAL CEMENT INDUSTRY UNION and BRAZILIAN CEMENT ASSOCIATION PORTLAND, 2019). One of the proposals is to use natural marble waste as a substitute for clinker into cement production. Therefore, an increasing number of patents on the subject is expected.

Figure 3 - Annual evolution of patent filing using natural marble waste

Source: The authors (2021)
As Figure 1 emphasizes, although this research has focused on the use of natural marble waste in Portland cement production, there were several patents of the use of this remnant in civil construction for the production of other materials, such as ceramic and ecological bricks. Despite this, Figure 4 shows that it is still possible to observe a research trend on the production of cement matrix materials with the use of marble waste: clinker (10 deposits), cement (8 deposits), mortar (7 deposits), and concrete (9 deposits), totaling 57.6% of the number of filings from 1982 to April 2021.

Regarding clinker production, all patent documents indicated the use of marble residue as a partial or total substitute for limestone. However, the residue amount in clinker production varied, in mass, from 5%, when used with granite powder, to 90%, in pellets without limestone. In six deposits, of the eight in which the marble residue was used in the production of cement, marble waste was in its powdered and pure form, as an addition, its participation varying, in mass, between 5% and 85%. The other two documents also pointed out the use of the material as an addition in cement production, but one of the documents indicated the need to premix 20 to 30% of the material with epoxy resin residue, while in the other 10% to 30% of marble residue was combined with sulfuric acid.

Figure 4 - Number of patent filings related to the final use of natural marble waste

According to Sutcu et al. (2015), Buyuksagis, Uygunoglu and Tatar (2017), Martínez-Martínez (2019), Benjeddou et al. (2020) and Ghirardi (2020), the large number of patents on the use of natural marble waste in clinker and cement production can also be justified by the fact that this residue has characteristics very similar to those of limestone, one of the main raw materials of cementitious matrix products. Like limestone, the marble waste is mostly used in its powdered...
form, as shown in Figure 5. This preference probably is also due to the large availability of marble dust, generated by the ornamental stone industry.

Figure 5 - Number of patent filings related to the use of natural marble waste by its form

Industrial interest in the subject was also evident from the analysis of patent filings. As can be seen in Figure 6, 57.6% of the documents were filed by industries, which reinforces the search for the proper destination for natural marble waste. Next, there are universities with 22% of deposits. It is important to highlight that it is possible that in some of the deposits made by universities there was a partnership for development of innovation projects with industries.

Figure 6 - Number of patent filings by type of applicant

Source: The authors (2021)
4. Conclusion

Given the data obtained from the analysis of patent documents filed since 1982 on the use of natural marble waste in the production of Portland cement, it is possible to conclude that there is great interest from China in the subject, with a focus on the use of powdered waste into the production of clinker, concrete, cement, and mortar, respectively. Considering the type of depositor, there is a highlight on the part of industry and universities and an increase in filings from 2014 onwards.

Despite Brazil's role in research on the use of natural marble waste in civil construction, which can be confirmed by the volume of papers published on the subject, there is still no patent filed from the country. It is understood that the reason for this discrepancy may be due to the time required for patent registrations in the country, low investments, or lack of interest from the civil construction sector.

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