VALORISATION OF FIRED CERAMIC TILE WASTE IN THE CONSTRUCTION SECTOR

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ABSTRACT

This paper addresses the valorisation of fired red-body tile scrap as secondary raw material in the manufacturing processes of new ceramic tiles, and as a reinforcing agent in both thermoplastic and thermoset polymer matrices in order to obtain composites for use in the building industry. The text describes the operations by which waste that used to accumulate in landfills is converted into an alternative raw material that contributes to establishing a circular economy in the ceramic industry, as well as creating a mutually beneficial industrial relationship with the plastics sector. The results obtained from the various studies carried out highlight the potential of using fired red-body tile scrap as a secondary raw material in lowering both the environmental impact from the generation of such waste and the manufacturing costs in both industries.

1. INTRODUCTION

The coming decades will be marked by fundamental social and economic changes, in which business and organisations will (or should) make a fundamental contribution. In this sense business enterprises, drawing on their value proposition, structure and value chain, are expected to contribute to establishing a circular economy and achieving the goals set for sustainable development (SDGs). Both issues are closely related since, on the one hand, the SDGs constitute the roadmap to be followed, while the circular economy represents the tool with which to create a scenario of economic and social growth that respects our planet.

However, the routes towards that circular paradigm are fraught with numerous obstacles, including cultural barriers, highlighting the fact that a transition of such magnitude can only take place if accompanied by profound cultural transformation. This is because the linear model of producing and consuming is so strongly rooted in our society and is, without doubt, unsustainable. In this regard, industrial symbiosis (IS) emerges as a strategic tool in the transition towards a circular economy system, characterised by maximum efficiency in managing resources, enabling us to extract their full value while creating minimal waste, emissions and energy leakage. Such progression calls for initiatives designed to slow down, close and/or narrow the loops that materials and energy follow, such as enhancing durable design or maintenance, repair, refurbishment, reuse and recycling operations, among others.

From the particular viewpoint of business enterprises, IS can contribute to reducing costs, providing growth opportunities and consolidating a company's image in the eyes of its customers. At the same time, IS is a valuable ally in tackling common challenges, such as climate change or scarcity of resources, by mitigating the effects of production methods on the environment and promoting a more efficient and responsible use of natural resources. Basically, IS entails one company or sector using the resources of other companies or sectors, keeping resources in productive use for longer, and considering the term 'resource' in the broadest possible sense, as it encompasses by-products, waste, energy, water, logistics, capabilities, expertise, equipment and materials.

In consequence, the concept of IS basically rests on two theoretical pillars: industrial ecology (IE) and sustainability. Firstly, because it likens the functioning of industries to that of natural ecosystems, in which diverse, resilient and regenerative industrial systems are given a collective approach to achieve a competitive edge through the exchange of resources amongst its members. Secondly, because it adopts a systemic approach to more sustainable and integrated industrial development from the environmental point of view and, at the same time, brings economic benefits.

In the ceramic industry, the use of secondary raw materials derived from waste streams reduces dependence on virgin raw materials in terms of both price and availability. However, the inclusion of waste streams from other sectors is a complex issue in which multiple factors of a differing nature, be that technical, environmental or economic, come into play. From a technical point of view, in many cases adaptation efforts need to be employed to make symbiotic interaction or synergy possible.

Examples of this would be pre-treatment operations of waste streams to remove pollutants, or adapting manufacturing processes to secondary raw materials whose chemical composition is different. On the other hand, the effects of such incorporation of waste on an end product's properties also needs to be analysed, as does the environmental impact associated with using secondary raw materials and their effect on the cost/benefit balance.

Within this particular context, the RECERCO project (INNEST/2022/133), funded by the Valencian Innovation Agency (AVI), focuses on the valorisation of fired ceramic tile scrap, i.e., fragments of ceramic tiles, mainly made of red clay, which have been rejected in the sorting stage because they do not meet the quality standards required of the end product. This project aims to incorporate that waste, once duly treated, as a secondary raw material both in the manufacture of new ceramic tiles and as part of polymer matrix composites for application in the building industry. The goal is to reduce the amount of waste that is currently sent to landfill (more than 15,000 tonnes per year), minimising environmental impacts and dependence on certain virgin raw materials, such as white clay and mineral fillers like calcium carbonate or talc, while at the same time creating new products with high added value.

In this manner, waste generated by industries in the Valencia Region becomes a raw material of interest in the building sector, thus establishing IS relationships, both within the ceramic sector (intra-sectorial), and between the ceramic and plastics sectors in the field of construction (inter-sectorial).

2. CHARACTERISATION OF THE WASTE STREAM

2.1. SECTOR ANALYSIS

The manufacture of ceramic tiles in the European Union (EU) generates waste in different production process stages, total ceramic waste being estimated at around 1.4 million tonnes per year. Although a very significant part of this waste is reintroduced into the production process, there still remains a certain proportion that is not subjected to recycling operations but sent to landfill. This is due to the alterations that using them would produce in the performance and final properties of ceramic tiles. Dedicated R&D efforts are therefore required to adapt them as secondary raw materials.

In order to reduce the environmental impact stemming from the use of virgin raw materials, the ceramic industry has managed to reuse almost 100% of pre-firing clay waste and, additionally, has developed methods to recover a large part of its fired tile scrap. However, the latest developments have focused almost exclusively on fired white-body tile scrap, i.e., waste from the use of white clays. This is a result of how the sector has evolved, shifting from mostly local red-firing clays to white-firing raw materials, normally imported at greater cost. In fact, in the late 20th century, the manufacture of white-fired tiles accounted for only 15% of production, whereas in 2021 it stood at over 70%.

With regard to fired red-body tile scrap, which is the subject of study in the RECERCO project, the situation is quite different, as it comes from abundant and, at least until recently, relatively low-cost local raw materials, such as local red clays. Consequently, a large amount of this waste is landfilled or used as bulk in products with low added value, such as aggregates for concrete or backfill material for soil stabilisation. However, the supply crisis affecting white clay, which comes chiefly from Ukraine (around 75%) and is therefore a further victim of the current war, as well as the emergence of new regulations on waste and circular economy (Law 7/2022 of 8 April), highlight the need to encourage the revalorisation of fired red-body tile scrap. Among other things, this means redefining, as a valuable secondary raw material, waste that currently accumulates in landfills, both in the ceramic industry and in other sectors such as plastics, by establishing synergies between industry sectors within a framework of IS.

Spain's tile manufacturers produce 500 million square metres of ceramic tiles per year[1]. Moreover, sector data indicate that the volume of fired tile scrap generated in the sector represents about 3% of total production in m^2. On top of that, bearing in mind that red-body tile production accounts for about 30% of total output, the volume of fired red-body tile scrap generated each year by the sector can be estimated. To make that calculation, let us consider the following products and their proportion within the red-body tile category:

a) Red-body stoneware tile (30%)

b) Red-body monoporosa tile (70%)

Taking the surface density of each product, 19.7 kg/m^2 (red-body stoneware) and 16.4 kg/m^2 (red-body monoporosa), the weighted surface density for red-body products is taken to be 17.5 kg/m^2. From that, an estimated 78,750 tonnes of fired red-body tile scrap are produced annually by the Spanish ceramic wall and floor tile sector.

***Figure 1**. Fired red-body tile scrap at the ECOVILAR S.L. waste management site. Source: ITC-AICE*

[1] Spanish Ceramic Tile Manufacturers' Association - ASCER (2022).

2.2. CHEMICAL CHARACTERISATION

Within the framework of the RECERCO project, mapping was conducted of the different companies in Castellon province that manage fired red-body tile scrap from ceramic tile manufacturers. These companies collect tile fragments and crush them to a certain particle size prior to marketing and distributing them as aggregate. In this study, samples of fired red-body tile scrap were collected from different companies in order to analyse variations in their chemical composition. The samples were sorted into particle size fractions by sieving, and the resulting fractions were chemically analysed by X-ray fluorescence to observe any discrepancies in chemical composition between fractions of different particle sizes (ϕ).

Oxides (%)	$\phi > 4$ μμ	$\phi < 125$ μm
SiO_2	66.8	66.5
Al_2O_3	17.5	16.0
Fe_2O_3	3.11	2.95
CaO	4.39	5.11
MgO	1.43	1.62
Na_2O	1.86	1.84
K_2O	2.65	2.39
TiO_2	0.65	0.63
ZrO_2	0.18	0.30
BaO	0.07	0.13
PbO	<0.01	0.11
ZnO	0.13	0.29
HfO_2	<0.01	<0.01
P_2O_5	0.14	0.14
SrO	0.01	0.02
MnO	0.03	0.03
CuO	<0.01	<0.01
L.O.I.(1000ºC)	0.45	1.59

Table 1. *Average chemical composition of samples of fired red-body tile scrap fractions.*

The results of our analyses of the various samples suggest that, generally, there are no appreciable differences in chemical composition between different fractions, which highlights the chemical homogeneity of the material under study.

3. WASTE VALORISATION

3.1. PRE-TREATMENT

In IS, substitution synergies involve one company re-using the waste streams of another. Thus, once the necessary valorisation operations have been implemented, what was worthless waste can be used as raw material (secondary raw material). Based on this scheme, the fired red-body tile scrap generated in the ceramic sector needs to undergo a series of operations to alter its properties and incorporate it again into ceramic production and into the manufacturing process of reinforced plastic materials (composites). This basically consists of transforming tile fragments into a powder material that can be appropriately processed, both in manufacturing new tiles and in obtaining thermoplastic and thermoset polymer matrix composites.

The following table summarises the requirements associated with each of the applications studied in the project:

Application	Requirements
Ceramic tiles	- Particle size: 13% residue on a 63-µm sieve (wet method)
Thermoplastic composites	- Particle size: d90 1.35-2.30 µm - Hardness: similar to $CaCO_3$ (3 on Mohs scale) - Colour - L*ab colour coordinates (mixing with pigments required?) - Moisture content (pre-drying required?)
Thermoset composites	- Particle size: d90 10-15 µm

***Table 2**. Technical requirements for applications: composites and ceramics.*

Based on the above indications, the waste was processed to render it as close as possible to the limits stated above within what is technically feasible. As can be seen from the information given by the companies involved, particle size is a crucial factor in all the applications considered in the project. Specifically, the scrap was processed in a dry micronisation system for raw materials such as clays, quartz, feldspar, frits, silica and zirconium, among others, where it was possible to produce a high output of particles smaller than 10 microns with stable, low, energy consumption. The results of the study carried out with the fired red-body tile scrap are shown below:

Particle size D90/µm	Production (kg/h)	Specific energy consumption (kWh)	Quantity produced (kg)
6.9	10	1702	64
9.6	35	544	225
29.4	119	149	555

Table 3. *Results after milling the fired tile scrap.*

As expected, the smaller the intended particle size, the lower the mill's productivity and so the higher its energy consumption. With respect to the rest of the requirements, as can be seen from Table 2, the application in thermoplastics is particularly restrictive in terms of particle size and hardness. For all these reasons, a compromise has to be struck between the parameters required by the application in thermoplastics and the technical and economic viability of revalorising the waste.

3.2. APPLICATION 1: CERAMIC TILE MANUFACTURING

Incorporating fired tile scrap as secondary raw material into the ceramic tile manufacturing process calls for reformulation of the tile composition and, consequently, study of the effect that the presence of such new raw material is likely to have at each stage of the ceramic manufacturing process (pressing, drying and firing) and on the final properties of the ceramic tile bodies.

The main conclusions of this study are summarised below:

- Within the framework of the project, different percentages of fired tile scrap (0 - 40%) mixed with red clay were tested. In addition, material with two different particle sizes was used, specifically d90 29.4 and 9.6 µm.
- During the pressing stage, the compactness of the ceramic tile body (bulk density) decreases proportionally to the percentage of fired tile scrap incorporated. This loss of bulk density is greater when the finer fraction of fired tile scrap is included because the scrap does not pack as well with the clay particles. The same tendency can be seen when the mechanical strength of the body is measured.
- On the other hand, after the firing stage, the bulk density of the fired body increases as the percentage of fired tile scrap rises. This is due to the lower carbonate content in the body as a result of the clay being replaced by fired tile scrap in the ceramic composition. Carbonates slow down sintering, which causes the body to densify more at the same temperature. As expected, the highest density values are achieved with the coarser fraction of fired tile scrap, as a higher value had already been noted in the unfired body.

- In order to maximise the added amount of fired tile scrap and improve the plasticity of ceramic compositions, the addition of small percentages of various plasticisers (polysaccharides, polyacrylamides and glycols) was studied. The results obtained show clearly that small percentages of glycols (2.5% - 5%) considerably improve the bulk density of the body in the pressing stage and make it possible to work with the highest percentage of fired tile scrap (40%).

Figure 2. *Ceramic tile bodies manufactured with 40% fired tile scrap incorporated into the composition. Source: ITC-AICE*

These results enable new ceramic compositions to be designed that require lower firing temperatures and, consequently, entail lower energy consumption and CO_2 emissions into the atmosphere, as well as lower manufacturing costs, as is assessed in the second stage of the project.

3.3. APPLICATION 2: COMPOSITE MANUFACTURING

Incorporating fired ceramic scrap as a reinforcement of polymer matrices into both thermoplastic and thermoset composites requires fired tile scrap to be pre-treated in order to adjust its properties to the typical ones of the materials used for that purpose. The final properties obtained after such pre-treatment were as follows:

Particle size (D90 / µm)	Moisture content (%)	Colour (L*, a, b)	Hardness (Mohs scale)
6.9 and 9.6	0.45	(65.1, 14.4, 21.0)	6-7

Table 4. *Properties of the fired tile scrap to be used as reinforcement material in polymer matrices.*

As part of the project, the incorporation of fired tile scrap as a reinforcement agent into PVC (thermoplastic) blind slats is under study. Furthermore, new formulations of *gel coat* and *topcoat* are also being developed to maximise the incorporation of fired tile waste into the construction sector in applications such as urban furniture and (thermoset) tanks. As, in all cases, the applications are for outdoor use, the formulations need to be adapted to obtain suitable degrees of resistance to weather ageing, i.e., to temperature, UV radiation and humidity.

The results obtained to date in the formulation of PVC compositions are summarised below.

- The study focused on completely replacing (36 *phr, parts per hundred resin*) the calcium carbonate used in this type of formulation with fired tile scrap with a particle size of 6.9 µm, as close as possible to that of carbonate. As a result, the change in mineral charge did not significantly alter the rheology, degree of mixing with the resin, or extrusion performance.

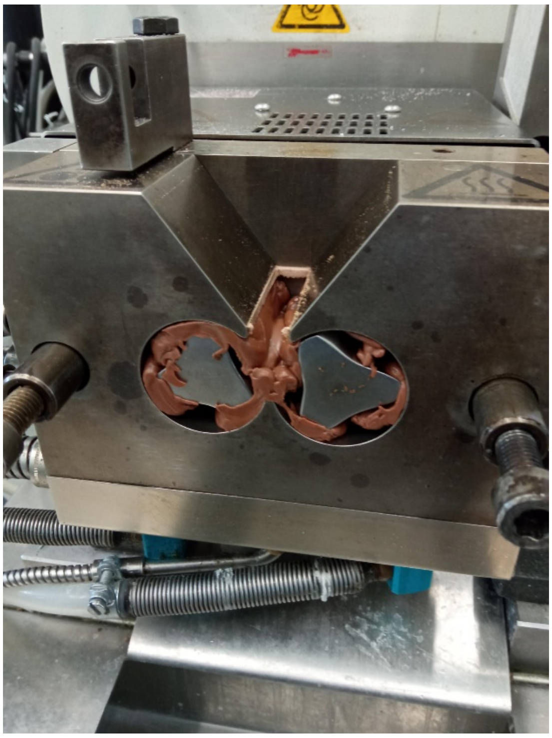

Figure 3. *Preparation of PVC compositions containing fired red-body tile scrap as mineral filler. (Source: AIMPLAS)*

- The results of the bending, hardness and tensile tests of the new PVC formulation show that its mechanical properties do not vary significantly when the mineral filler is replaced. However, lower impact resistance is observed.
- The thermal resistance of the new formulation was studied on the basis of measuring the Vicat softening temperature. The results show that the replacement of calcium carbonate by fired tile scrap does not alter this parameter.

- Finally, after subjecting the new composition to accelerated ageing in order to evaluate its suitability for outdoor applications, much slower and more gradual degradation of its mechanical properties is observed when fired tile scrap is used as mineral filler.

Figure 4. *Obtaining PVC profiles by incorporating fired red-body tile scrap as mineral filler. (Source: SAXUN - Giménez Ganga)*

In view of the above, the results obtained in PVC matrices are promising and open the door to designs of new compositions with improved properties for outdoor applications.

4. CONCLUSIONS

This paper reports the various results obtained so far from the RECERCO project have. Indeed, the study shows how fired red-body tile scrap can be used as a secondary raw material both in the manufacture of ceramic tiles and in the production of PVC blind slats. In both cases, the results are promising and indicate the technical viability of this synergy which, apart from aiding achievement of the goal of zero waste in the ceramic industry, connects our sector with the plastics industry through industrial symbiosis (IS). Thanks to this scheme, the ceramic industry eliminates waste that traditionally ended up in landfill and which can, after suitable valorisation operations, be transformed into an alternative raw material to calcium carbonate in the manufacture of reinforced PVC composites.