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ELECTROCERAMICS IN SUSTAINABILITY

FOCUS ISSUE

Sustainability and decarbonisation in the ceramics sector through resource and energy efficiency

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This article addresses innovative manufacturing, circular economy, and resource and energy efficiency to reduce CO_2 emissions in the ceramics sector using the UK as an exemplar. A comprehensive action plan is proposed, that if implemented would cut emissions in line with net zero 2050 with commensurate energy and cost savings. The plan demonstrates methods to improve heat recovery, reuse of waste and enhanced product yield through digitisation and illustrates novel low-energy manufacturing methods. The use of supply chain, life cycle and technoeconomic assessment is emphasised to ensure economic, environmental and societal sustainability.

Introduction

The UK ceramic sector has a revenue of £1.6 billion p.a. with £600 million in exports [1] with ~ 17,500 people directly employed and 1000 s more supporting supply chains [1]. This economic activity is distributed throughout the UK through 150 + ceramic manufacturing sites. Ceramics is both a foundation and advanced industry [2], with applications ranging from construction and refractories to electronics and aerospace and it emits about ~ 1.2 Mt of CO_2 across all subsectors [2]. It is composed of heavy clay construction products, whitewares, refractories and technical (advanced) ceramics, Fig. 1. CO_2 sources vary from subsector to subsector but within the UK are typically ~ 67% natural gas, 17% electricity (direct and indirect) and 16% process emissions [1], Fig. 2.

Currently, H_2 is being explored as an alternative to natural gas with many heavy clay and traditional ceramic companies actively pursuing fuel switching strategies for large scale manufacturing, e.g. Forterra [3]. However, the production of green H_2 at an industrial scale sufficient to decarbonise the UK ceramics sector is not currently anticipated for > 10 years [4] and its delivery is exacerbated by the geographic location of the ceramics industry away from clusters and in dispersed sites where H_2 pipelines are not currently envisaged. Furnace electrification is a potential long-term strategy, but legacy equipment is gas burning and significant retrofitting of existing and/or purchase of new capital equipment is required for this to be an effective way to reduce emissions. Base electricity costs in the UK are also currently far higher than for natural gas per tonne of product, which has historically discouraged investment in electrification within the ceramic industry [5]. Moreover, the radiative characteristics of electric furnaces are radically different to those of natural gas, and a comprehensive redesign of the kiln/manufacturing process is likely required for production. Small batch or intermittent production using electric furnaces is commonly found and some advanced ceramics such as multilayer layer ceramic capacitor production use electrified tunnel kilns. Fuel switching from natural gas to H_2 or electricity is, however, problematic for the continuous production of heavy clay products, refractories and whitewares which constitute the largest volume of ceramics produced in the UK and worldwide.

A more holistic perspective is therefore required that tackles the resource and energy efficiency for all potential aspects of production, Fig. S1 [6], from heat recovery, reuse of waste, enhanced product yield through digitisation and the

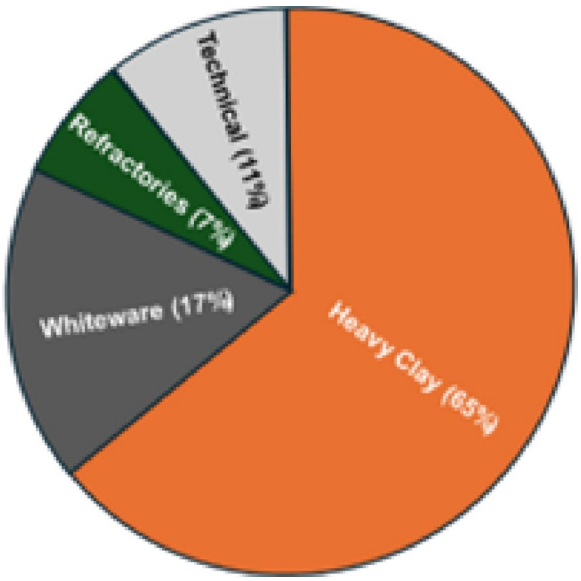

Figure 1: Schematic showing the distribution of ceramic production the UK.

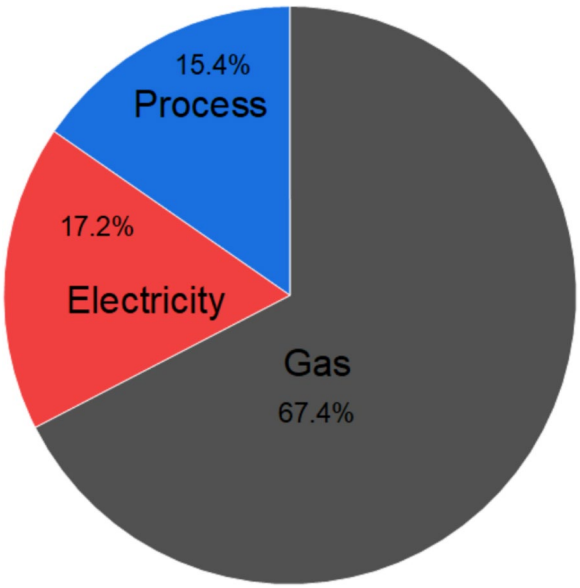

Figure 2: Breakdown of sources of CO_2 emissions in the UK ceramic sector.

introduction of novel low-energy manufacturing methods. The following sections not only address these topics in sufficient detail to offer innovations that can be directly implemented but will also emphasise the role of supply chain, life cycle and technoeconomic assessment in the roll out of any new materials/technology to ensure economic, environmental and societal sustainability.

Energy efficient manufacturing of ceramics

Heat recovery

As noted in reference [6], the recovery of excess heat by capturing kiln gases to preheat the combustion or dryer is an important way to reduce fuel consumption, reduce CO_2 and improve efficiency. The use of heat pipe heat exchangers (HPHEs) to recover waste heat from industrial exhaust streams has been investigated and applied in many sectors. HPHEs are made up of multiple independent, gravity assisted heat pipes that can be arranged to optimally match the geometry of the intended application [7]. Heat pipes are commonly arranged in either staggered or inline configurations and can be internally designed with multiple flow passes or fitted with baffles to enhance the convection heat transfer coefficient, similar to conventional heat exchangers. This improves the efficiency of full-size heat exchangers by increasing the number of passes while having little impact on maintenance. Each heat pipe is hermetically sealed and contains a selected working fluid with evaporation and condensation temperatures tailored to the operating conditions of the process. During operation, a heat pipe enables internal heat transfer between the evaporator and condenser sections through the circulation of a working fluid, with no moving parts [8]. The lower section of each heat pipe is exposed to the hot process stream, while the upper section is in contact with the cold stream. HPHEs present significant advantages over conventional heat exchangers, notably complete thermal isolation between hot and cold streams and passive heat transfer via the heat pipes. They maintain operational continuity despite individual pipe failures, allow for scheduled maintenance, exhibit higher heat transfer efficiency with reduced surface area, enable more compact and cost-effective designs, possess service lifetimes of up to 20 years and typically achieve a payback period of less than two years [9]. Three heat exchanger designs based on heat pipes have been developed for the ceramics industry (Fig. 3). The working conditions and achievements of individual designs are briefly discussed below. Within the framework of the ETEKINA project [10], an assessment of the energy intensity was conducted for a ceramic tile manufacturing facility situated in the ceramic district of Emilia-Romagna, Italy. In the ceramic manufacturing process, the primary energy sources are natural gas and electricity, with approximately 80% of total energy consumption attributed to natural gas. Thermal energy is predominantly consumed during firing (53%), followed by spray drying (35%) and drying (10%). The process of making ceramics begins in continuous mills where raw materials such as clays, sands, feldspars and kaolin are ground together with water. This is followed by the shaping of products using hydraulic presses to achieve the desired geometry. The final stages of the process involve

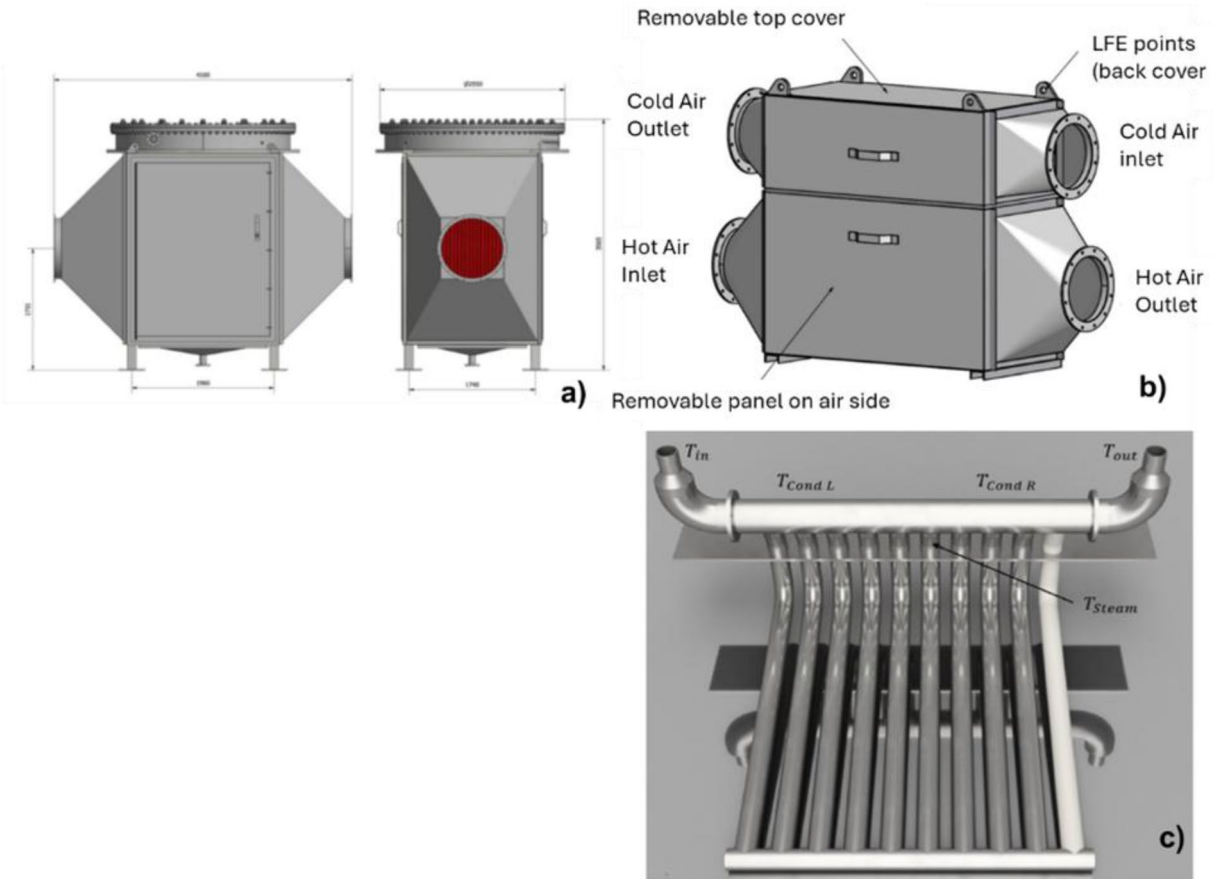

Figure 3: HPHE designs (a) ETEKINA project [7]; (b) HPHE dream project [6] and (c) radiative flat HPHE [5].

drying, decorating and firing in ceramic kilns at approximately 1250 °C. Exhaust gases exiting the kiln are directed into the HPHE at around 245 °C. Within the HPHE, the exhaust stream is cooled to approximately 155 °C before rejoining the exhaust stack. Based on an energy balance of the flue gas stream (accounting for mass flow rate, specific heat capacity and temperature reduction), this cooling represents a significant recovery of the available thermal energy. On the secondary side, water is used as the heat transfer fluid and experiences a temperature rise of approximately 50 °C, increasing from 115 °C at the inlet to 167 °C at the outlet. This recovered thermal energy, in the form of pressurised hot water at 167 °C, is transported via a closed-loop system across the factory and utilised to preheat air for the spray dryer. This integration reduces the energy required to reach the target inlet air temperature for drying. The HPHE achieves recovery of over 40% of the total available waste heat from the kiln exhaust stream, defined here as the fraction of recoverable thermal energy relative to the total thermal content of the flue gases above the stack exit temperature. This recovered energy directly offsets the thermal load otherwise supplied by natural gas, resulting in an equivalent reduction in fuel consumption at the spray dryer stage [11].

Analyses of the second HPHE design were carried out for the ceramic kiln in an actual industrial installation situated in the ceramic district of Emilia-Romagna, Italy [9]. The kiln operates with a tile production capacity of approximately 5,000 kg/h and is designed for continuous operation of around 8,700 h per year. It consists of 56 modules, each 2.1 m in length, and is divided into five main zones: preheating, firing, rapid cooling, slow cooling, and the outlet section. A theoretical approach to calculating the thermal and fluid dynamics characteristics of the heat exchanger was proposed to obtain the potential energy recovered from the ceramic process. Calculations demonstrated that up to 863 MWh of thermal energy can be recovered per year and utilised to preheat the hot air stream supplying the pre-kiln dryer. Consequently, the annual fuel savings from the dryer burners amount to approximately 110,600 Sm^3, corresponding to a reduction of 164 t CO_2 per year and fuel cost savings exceeding €22,000 annually. These results indicate that the implementation of heat pipe-based heat recovery in ceramic processes is advantageous in

terms of energy efficiency, environmental performance and economic viability.

The novel radiative HPHE recovers heat and improves the cooling profile of the tiles. It is composed of ten parallel pipes, each 28 mm in diameter and 2 mm thick. At the lower end, the pipes are joined by a 28 mm connecting manifold and integrated with a 50 mm condenser section [8]. The condenser operates as a shell-and-tube exchanger and contains nine tubes with a diameter of 10 mm. The length of the evaporator section is 430 mm with an inclination of 5°. The theoretical predictions of heat transfer in the kiln show excellent agreement with the experimental results. At higher heater temperatures (400–500 °C), the system achieved significantly higher rates of heat recovery, with the total amount of heat transferred remaining approximately constant for different flow rates, and the main change being in the temperature difference between the inlet and outlet of the cooling system. Thus, it was demonstrated that the radiative heat pipe successfully captured heat via radiation and natural convection in a closed kiln environment, with a heat recovery capacity reaching approximately 4 kW. The phenomenon of Geyser boiling was also observed, which caused a temporary increase in the temperature of the heat pipes during system heating. The average efficiency of the heat pipes was approximately 42% at 500 °C and 26% at 400 °C, assuming a constant power supply from the heater. Depending on the temperature of the heaters, the heat pipe was able to extract between 400 and 3100 W of thermal energy from a heat source with an area of 0.261 m^2. At a heater temperature of 200 °C, the heat transfer rate was primarily driven by natural convection, whereas radiation became the dominant mechanism at higher temperatures. These results indicate that the use of heat pipes in a ceramic furnace is most effective at higher operating temperatures.

Applications of MW heating in the ceramic industry

Microwave processing, due to its volumetric heating capability, utilises shorter processing times (and in some cases lower processing temperatures) and thus is more energy efficient with lower environmental impact. Research and development in the application of microwaves for processing ceramics is on-going in subsectors ranging from tiles, dinner plates, sanitary ware to specialty materials such as bio-ceramics and electro-ceramics. This section briefly overviews some of the well-established, industrial applications of microwaves for traditional ceramics, mainly, in drying of tiles and foams and calcination of limestone.

Fundamentals of microwave heating

The microwave region of electromagnetic (EM) radiation lies in wavelengths in the range of 1 mm to 1 m [12]. When microwaves irradiate a dielectric material, the charged dipole in the material responds to the EM electric field component which in turn results in internal volumetric heating. As this phenomenon occurs at the molecular/ionic level, provided the material itself is homogeneous heating occurs uniformly and rapidly [12]. The power absorbed per unit volume (P) during microwave heating is given by

$$P = 2\pi f \varepsilon_r' \varepsilon_0 |E, H|^2 \tan\delta,$$

where E is the electric field across the volume, f is the frequency of the microwave, ε_r' is the relative permittivity, ε_0 is the permittivity of free space, H is the magnetic field component and tan δ is the dielectric loss factor. ε_r determines the ability of microwave absorption of a material and tan δ dictates the material's ability to convert the absorbed energy into heat. The commonly used frequencies for industrial applications are 916 MHz, 2.45 and 9.8 GHz. Although microwaves are utilised to process many oxide and non-oxide ceramics, at the industrial scale their established usage is mainly associated with the drying of tiles and ceramic foams.

Drying of ceramic tiles

The fabrication procedure for a ceramic involves batching, mixing and grinding, forming (wet or dry), drying, glazing and firing, Fig. S1. Drying forms a major part of the total energy consumption and if there is residual water in the material, there is risk of damage to the tiles due to handling or during further processing. Efficient removal of water is therefore required. Drying using convective or radiative heating can result in stress distributions within the ceramic due to non-uniform heating from the surface inwards. To prevent cracking therefore, conventional drying is performed over many tens of hours [13, 14] and is usually carried out on green ceramic tiles in natural gas-fuelled vertical dryers. Microwave heating provides an eco-friendly technology, and it has been explored for drying ceramic tiles, following examples developed by the food industry.

As water efficiently absorbs adsorbs microwave radiation, its use to dry, especially thicker tiles, has been explored, heating evenly throughout. Pressure, which increases with temperature, transfers the water molecule to the surface, where the pressure is lower. Depending on the material and the water content, the net shape of the tile can be regulated more precisely, to achieve controlled, yet rapid evaporation/removal of water. The stress difference between the surface and the core is negligible, and optimum usage of power results in little or no deformation.

Durgut [14] investigated the drying of ceramic tiles using microwaves, reporting how the microwave power and the rate of drying affected the surface temperature, moisture and bending strength of dried ceramic tiles. Kaolin-based floor and wall tiles with 6% moisture content were used with dimensions of 33 × 33 cm and 25 × 40 cm, respectively. The samples were dried individually in a horizontal pilot-based microwave dryer using various microwave power levels (20, 35 and 50 kW) and belt

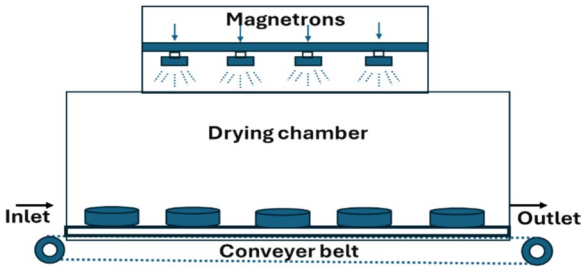

Figure 4: Schematic of the horizontal pilot-based microwave.

speeds (0.5, 1.0. 1.5 and 2 m/min). The schematic of the microwave dryer is shown in Fig. 4. Minimum and maximum surface temperatures of 51/143 °C and 50/138 °C were observed for the floor and wall tiles, respectively. The moisture removal increased with increase in microwave power and decreased with belt speed. The moisture values were reduced from 6 to 0.1% at 35 kW power with a 0.5 m/min belt speed for floor tiles with 50 kW power required for wall tiles at 0.5 m/min belt speed. Moisture removal was found to be more efficient in the centre of the tile compared to the corners (due to the inside-out gradient during microwave heating and the associated thermal loss) in contrast to conventional drying. Maximum bending strength values for floor and wall tiles were 29.2 and 31.2 kg/cm^2, respectively, which decreased with increase in belt speed with more time needed to dry the tiles fully to improve the properties.

The results indicated 1000 and 837.5% estimated increases in drying capacity, 52.4 and 42.6% of energy savings, and 50.0 and 42.1% of environmental gain could be realised with microwaves compared to natural gas heating for drying floor and wall tiles, respectively. This demonstrated the efficacy of the microwave methodology as a more energy efficient, enhanced throughput, environmentally friendly process than the natural gas-fuelled vertical dryers for drying ceramic tiles.

In addition, many refractory, porcelain and pottery ceramic components have been processed using microwave-assisted methodology. Table S1 shows the consumption of energy involved in the processing of these ceramics using conventional and microwave technologies [15, 16]. The energy efficiency of the microwave process is quite evident.

Drying of ceramic foam filters

Ceramic foam filters (CFFs) are used in foundry/aerospace industry for filtering molten metals and Ni-superalloys. These are open cell polymer foams impregnated with ceramic slurry. Conventional drying of these ceramic foams typically takes 12 h, limiting the overall manufacturing throughput as well as taking up a large industrial floor space (~ 100 million CFFs per year by one UK manufacturing company alone). However, ROMILL, a Czech-based microwave equipment manufacturing company, in collaboration with the leading ceramic foam producer LANIK,

Figure 5: Microwave conveyer belt drying system for drying ceramic foams [6].

has successfully utilised microwave heating technology to significantly reduce the foam drying time to just 40 min. A powerful 12-m microwave dryer (Fig. 5) with openings at both ends enables continuous production and dries the entire array of filter products [17]—thus leading to time/energy savings.

Microwave-assisted technology for limestone calcination

In cement, steel and glass industries, the most energy-intensive process is limestone calcination. Ceralink [18] along with an industrial lime partner and a university collaborator constructed a 22 kg, 27 ft^3 MAT kiln (Fig. S2). Limestone was placed in alumina kiln furniture and heated to 1200 °C with 5 °C/min ramp rate at 20 kW microwave power. Energy and mass loss data were measured. The MAT pilot scale (22 kg) results indicated that using a 2 h soaking period resulted in ~ 25% reduction in energy consumption when compared to the conventional heating experiment. Ceralink and their collaborators [18] reported that this could represent in US lime production (19.8 million tonnes lime per year [19]) a saving of $29 million and ~ 2.14 m MWh.

As indicated above, microwave-assisted drying has achieved higher Technology Readiness Levels (TRLs) and hence is much closer to industrial adoption; however, the use of microwaves for high-temperature sintering/calcination still requires considerations on field uniformity, penetration depth, scale-up equipment costs and thorough Life Cycle and Techno Economic Analysis (LCA/TEA), prior to widespread industrial take-up.

Low-energy/temperature sintering methods

One of the major limitations and issues with traditional sintering is the high temperature required to achieve full densification. This leads to high energy requirements and costs, grain coarsening when potentially a small grain size is desired to optimise physical properties and severe limitations in the types of materials that can be co-sintered with ceramics. There are several techniques which industrial ceramic production

has in its 'toolbox' to lower processing temperatures. This includes the use of sintering aids, or nano-powders, or different sintering techniques such as hot pressing and spark plasma sintering (SPS). However, all these techniques rarely lower the sintering temperature below 900–1000 °C, often require a large capital outlay on equipment (hot pressing, SPS) and in some cases affect the desired material properties. Although a full discussion comparing and contrasting the strengths and weaknesses of various low-energy/temperature techniques is beyond the scope of the review, cold sintering is discussed in detail due to its relatively low capital outlay (at least at the lab scale) and ultra-low sintering temperature, typically < 300 °C, that promotes compatibility with e.g. polymers [20, 21].

The cold sintering process (CSP)

The cold sintering process (CSP) is a recently discovered densification technique [20, 21] that builds on previous work on hydrothermal hot pressing [22], allowing for the full densification of ceramic materials in many cases at < 300 °C. In CSP, the material to be sintered is combined with a small amount (several vol%) of a 'transient solvent' and hot pressed at 25–300 °C for ~ 0.5–1 h, leading to full densification.

There are many interacting mechanisms in CSP and it is still a field of intense study, but it is broadly agreed that the dominant densification mechanism is pressure solution creep (PSC) [23], a process commonly found in nature (such as in the formation of sedimentary rock), well known to geologists since it was first recognised in the early 1860. The wider sintering process is thought to occur over three stages. In the first stage, particles go under initial compaction, high energy surfaces such as sharp edges are dissolved and capillary action allows for particle sliding and rearrangement leading to a higher packing density. As the temperature increases, PSC is the primary mechanism in the second stage, Fig. 6. High-pressure zones at the closest liquid–solid interface points between grains lead to particle dissolution which creates a high chemical potential gradient compared to low-pressure zones, leading to particle mass transport to areas of low-chemical potential. Dissolved material then precipitates and recrystallises at low-energy surfaces leading to densification and grain growth, following the terrace-ledge-kink model [20]. In the final stage, the liquid-solvent phase is removed from the material, leaving only the desired densified phases.

Advantages of CSP

To date, a large number of materials have been successfully densified using CSP [24], with the energy requirement of densification showing a reduction as high as 95% in some cases [25]. One of the unparallelled advantages of CSP is the ability to produce fully densified ceramic composites with materials (e.g. polymer–ceramics) that would be impossible with any other sintering techniques [26]. An early example of this includes the manufacture of a PTFE-ZnO varistor device [27]. In the study, ZnO combined with 10–40vol% PTFE was cold

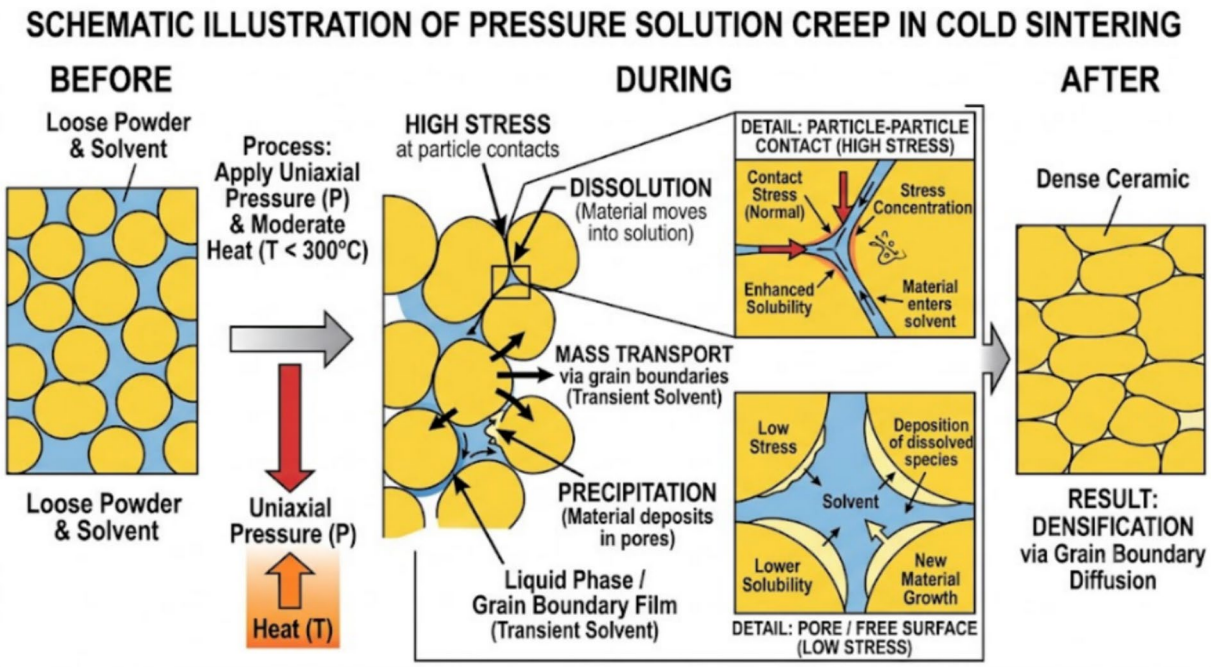

Figure 6: High-pressure zones at the closest liquid–solid interface points between grains leads to particle dissolution in the solution–precipitation creep process, after [23].

sintered at 285 °C. The best varistor performance was found at compositions of 5–10 vol%, beyond which zones of unsintered ZnO were observed, caused by the segregation by the polymer phase. While the varistor performance was not up to commercial standards, it was an early demonstration of the ability to co-sinter ceramics with polymers to produce a functional ceramic device.

Other demonstrations of ceramic-polymer composites have included the ability to directly sinter a transparent layer of ZnO to the surface of the temperature resistant thermoset polymer Kapton®, producing a multilayer ceramic composite [28]. In this work, nano-ZnO was tape-cast onto the surface of Kapton® sheet; after which the binder phase was removed and the ZnO was cold sintered between layers of Kapton®, producing the first demonstration of a transparent ceramic directly sintered onto a polymer.

Some materials, however, have proven more difficult to cold sinter such as α-Al_2O_3, due to its high chemical resistance, so focus has been on using CSP as a pre-sintering step to help lower the final sintering temperatures without the need for sintering aids or nano-powders. So far, work has involved making use of its precursor phases such as γ-alumina [29], a form of functionalised boehmite (AlO-(OH)) called carboxylate–alumoxane [30], inspired by work with the material in the late 90's [31] and most recently making use of hydrated ρ-alumina [32].

The first published work [29] combined 20–50 wt% γ-alumina with α-alumina and cold sintered the material at 300 °C and 300 MPa with glacial acetic acid. This led to densification of the γ-alumina, which was followed by a final heat treatment step at 800–1350 °C to produce dense α-alumina. The best results were achieved in 20 wt% γ-alumina samples, heat treated at 1350 °C, producing > 98% dense material with Vickers hardness and a Young's modulus comparable to conventional alumina sintered at 1500–1700 °C. This work demonstrated that cold sintering could indeed be utilised to lower the densification temperature of α-alumina and produce high-quality material. However, the high cold sintering pressure and the use of glacial acetic acid at high temperatures would pose a significant barrier to scale up.

Later studies adopted a different approach [30] using carboxylate–alumoxane ($[Al(O)_x(OH)_y(OOCR)]_z$), a highly soluble, functionalised form of the mineral boehmite (γ-AlO(OH)). In this work, boehmite was mixed with various ratios of either micron- or meso-sized α-alumina powder. This was then cold sintered with 2 M acetic acid at 125–300 °C, before final heat treatment at 1450 °C to produce densified single-phase α-alumina. Densities of 95% single-phase α-alumina were achieved at 1450 °C in 70% boehmite material with hardness values equivalent of material sintered at 1650 °C. This work showed several improvements on the previous works as while it required a higher final sintering temperature, much lower cold sintering temperatures were needed coupled with a more benign solvent.

More recent work [32] utilised hydrated ρ-Alumina with distilled water followed by hot pressing at 450 °C and 100–250 MPa to directly synthesis a single-phase, 42–34% porosity, α-alumina body with compressive strength of 89.4–140.2 MPa, stronger than many porous α-alumina materials produced by other methods. While high densities are not achieved in this work, it is the first that demonstrated that direct transformation to α-alumina with strong grain/particle bonding at comparatively low sintering temperatures.

Limitations of CSP

Cold sintering has several capabilities unmatched by any other low temperature/energy sintering technique. However, there are many current challenges that must be overcome before scale-up can be considered feasible [33, 34]. Major hurdles include the high-pressure requirements for several materials, the limited geometry due to the use of uniaxial pressing and the restricted batch size. Hence, CSP's ability to sinter at such low temperatures will never, in isolation, be enough to compete with the already well-established economies of scale within the current ceramic manufacturing industry. For commercialisation of CSP to be feasible, any manufacturing process needs to take advantage of its ability to produce materials not previously possible, such as polymer–ceramic composites, or being effectively used as an assistive step to other sintering techniques, either as a pre-step as discussed in this paper, or even incorporated into well-established techniques such as spark plasma sintering [35] or flash sintering [36].

Overall, cold sintering is a fascinating technique that has great promise, but its future most likely lies in unique, specialised ceramic manufacturing, or as an assistive process. Unless the research community comes to terms with this reality, it is unlikely to progress beyond lab-scale demonstrations. Many claim significant reductions in CO_2 emissions using cold sintering along with other low-energy/temperature sintering techniques. While for a given system this may be correct, it is extremely unlikely that it will be used in the manufacture of whitewares and heavy clay products due to the pressures required for large parts and its inherent batch process character. Given that heavy clay products and whitewares constitute 80% of the volume production of ceramics in the UK and around the world using almost exclusively natural gas fired kilns, its effect on the reduction of CO_2 emissions for the industry as a whole is questionable.

Mechanochemical synthesis

In the 20th century there was the emergence of advanced ceramics starting with high purity and dense alumina. This was

followed by transition metal carbides [37] boron carbide, silicon carbide [38] and silicon nitride [39] materials for applications in extreme environments and conditions and by the development of ceramic matrix composites [40] reinforced with particulate and fibre phases, including nanoscale phases [41]. More recently, there has been the emergence of layered compounds, such as $M_{n+1}AX_n$ (MAX) phases [42] and transition metal dichalcogenides.

Beyond oxides, carbides and nitrides, there are a few applications such as sensors and infra-red windows for advanced ceramics with other anions such as alkaline-earth fluorides (CaF_2, SrF_2, BaF_2), alkali halides (NaCl, KCl, KBr) and chalcogenide ceramics [43]. In recent years, there has been increasing interest in sulphide materials because of their favourable electronic band structure for photovoltaics, catalytic, thermoelectric and battery applications [44]. However, sulphides, with their weaker anionic bonding, are less stable, moisture sensitive and have poor mechanical properties compared to oxides. They have the advantage that they are based on inexpensive, earth abundant and environmentally friendly elements. They can potentially be synthesised at large scale by mechanochemistry, which has advantages when using low melting point and volatile elements [45, 46]. The use of mechanochemistry provides a lower energy scalable route to synthesise the precursor powders for sintering for a range of sintering methods such as cold compaction and pressure-less sintering, hot pressing and SPS, compared with traditional milling/mixing and high-temperature calcination. In particular, there has been greater emphasis on sulphide thermoelectrics, such as those based on zinc blende and wurtzite structures, including $Cu_{12}Sb_4S_{13}$. The common mechanochemical synthesis processing route for these materials involves reactive ball milling of pure metal, sulphur and sulphide precursors. The product is mostly a highly reactive, amorphous target phase with a small amount of precursor impurity. The subsequent sintering of these precursors produces a dense, highly crystalline and single-phase ceramic as illustrated in Fig. 7(a) [47]. The proportion of energy consumed in ceramic production by ball milling is dependent on the required composition and purity, and batch size. It is estimated to be in the range of 20–25%, with furnace processes constituting the main proportion of energy consumed [48]. Of the energy consumed by ball milling, it was estimated that 91% of the energy was lost as heat, with only 9% actually contributing to material processing [49].

Advanced ceramics tend to have relatively simple compositions, often of high purity or consisting of solid solutions of no more than two/three cations, with the addition of small quantities of dopants (< 5 mol%). The presence of more than one distinct cation sites adds some structural complexity. Inspired by research on high entropy metal alloys [50, 51], there is increasing interest in high entropy, multi-element

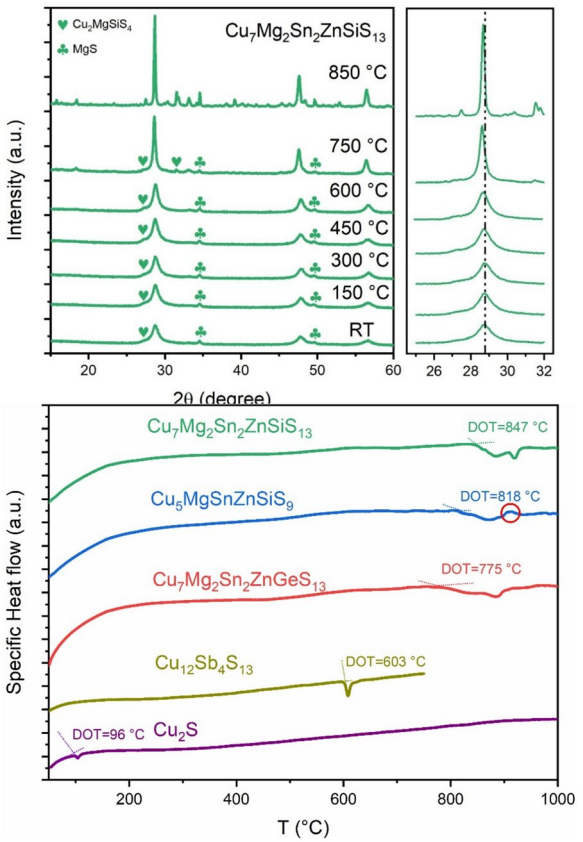

Figure 7: (a) Synthesis of zinc blende structured sulphide thermoelectrics by mechanochemistry (RT), followed by annealing. (b) Oxidation onset temperature (OOT) of low and high entropy sulphides [47].

ceramic materials, containing five or more elements [52, 53] with long-range structural but short-range chemical order. This leads to several characteristic features of high entropy materials, namely, thermodynamic stabilisation, lattice distortion, slow diffusion and synergistic effects. This emerging field opens up new compositional space, stabilised by increased entropy, in which interesting and useful materials might be discovered. The combination of different elements down to the atomic level produces synergistic effects, which takes properties beyond those predicted by the rule-of-mixtures, e.g. mechanical properties, electronic and thermal transport. The increased stability of high entropy materials compared to their precursor compounds was recently demonstrated for thermoelectric sulphides, where the oxidation onset temperature of zinc blende-based compounds was increased from 600 to > 800 °C, Fig. 7(b) [47]. A consequence of the random, disordered solid solutions is lower diffusion mass and thermal conductivity, which has implications on their synthesis and sintering behaviour, including smaller grain size [54]. The use of multiple components makes their synthesis more demanding, longer milling and calcining/annealing times to achieve uniform distribution of the elements and to avoid the

formation of second phases. While chemical synthesis routes can achieve high chemical homogeneity, they are less amenable to scaling up. Currently, high entropy ceramics are synthesised in laboratories using relatively expensive precursors, which are usually mixed, ball milled and calcined. There is an opportunity here, by the choice of less pure precursors along the supply chain, to considerably reduce the effort, cost and carbon footprint of ceramics, while at the same time benefiting from the good properties of high entropy ceramics. While mechanochemical synthesis does not offer a viable route for the fabrication of traditional ceramics, it opens up a cost-effective design space for the manufacture of novel advanced ceramics for high value industries such as electronics and aerospace and reduces energy use by minimising high-temperature calcination.

Resource efficiency in ceramic manufacturing

Digital transformation in the ceramics industry

Recent advances in Industry 4.0 and Industry 5.0 paradigms, combined with artificial intelligence (AI) and digitalisation, have introduced transformative opportunities to enhance sustainability and competitiveness. These benefits are often realised indirectly through enhanced product quality, process optimisation, increased operational efficiency and defect reduction.

This section reviews recent applications of AI and digital twin technologies in ceramic materials processing, highlighting their potential to drive process improvements, enhance product performance and support more sustainable manufacturing practices.

Quality and sustainability indicators of traditional ceramic manufacturing

Recent studies, summarised in Table 1, have highlighted how industrial data-driven tools and cyber-physical systems can enhance both environmental and economic performance in ceramics tiles manufacturing, supporting a transition towards circular economy. Raffaeli et al. [55] described a four-year project implementing digital twins, virtual commissioning, and AI-based inspection systems in ceramic tile manufacturing. A dedicated pilot production line was established to evaluate these technologies, showing improvements in several sustainability key performance indicators (KPIs), including a 20% increase in production capability, a 10% reduction in waste, and a 12% improvement in overall equipment effectiveness. Similarly, Contini et al. [56] proposed a set of digital KPIs and introduce a "Digital Twin of Sustainability" concept to enable real-time decision-making, highlighting the role of big data analytics and cyber-physical systems in monitoring economic, environmental and social performance indicators.

More recent work applied supervised AI models, including deep neural networks (DNN) and support vector machines (SVM), to predict product quality and detect defects in continuous ceramic manufacturing [57]. The study used data acquired from a pilot ceramic tile production process, incorporating parameters from digital printers and vertical dryers alongside quality information recorded by the quality control department. However, the study included only a small dataset and was able to classify only defective tiles but not the type of defect. Finally, a hybrid modelling framework that integrates fundamental physical principles—specifically kinematic

TABLE 1: AI approaches in traditional ceramics manufacturing.

References	Approach	Application area	Key outcomes/findings	Data source
Raffaeli et al. (2024) [55]	Digital twins, virtual commissioning, AI-based inspection systems	Ceramic tile manufacturing (pilot production line)	Demonstrated feasibility of integrated cyber-physical systems. Key Performance Indicators (KPIs): • 20% increased efficiency and production capability; • 10% waste reduction; • 12% improvement in overall equipment effectiveness (OEE)	Pilot production line data
Contini et al. (2023) [56]	Digital KPIs, 'digital twin of sustainability', big data analytics, cyber-physical monitoring	Sustainability assessment and real-time decision-making	Enabled monitoring of economic, environmental, and social KPIs; Supported circular economy strategies through real-time insights	Industry-collected multi-domain performance data
Ozcan & Sengul (2025) [57]	Supervised ML (DNN, SVM), multisensor fusion, AI-based defect detection	Continuous ceramic tile production (quality prediction and defect detection)	Improved defect detection; enhanced production throughput; reduced waste via predictive analytics	Sensor data of printer & dryer parameters, QC department records
Sousa et al. (2020) [58]	Hybrid physics informed with ANN modelling (kinematic equations neural networks)	Polishing of stoneware tiles (gloss distribution prediction)	Higher accuracy than physics-only models; improved process control and quality assurance	Experimental polishing and gloss measurement data

equations—with ANN-based data-driven modelling has been developed to predict gloss distribution during the polishing of stoneware tiles, enabling more precise process control and improved quality assurance in polishing operations [58].

While these studies demonstrate the potential of AI tools in improving both quality and efficiency of traditional ceramic manufacturing, results are limited to pilot studies. Scaling up AI deployments in traditional ceramics manufacturing requires solving several challenges: (i) the seamless integration of heterogeneous data sources from sensors, machines and visual systems while addressing issues such as missing, noisy or insufficient labels; (ii) building and maintaining high fidelity digital twins that require real-time data synchronisation, and substantial computational resources; and (iii) overcoming limited digital skills, high initial investment costs and the difficulty of scaling pilot solutions to full industrial environment.

Enhancement in quality and performance of advanced/technical ceramics

In advanced ceramics, AI tools have been used to support the material design, processing and quality optimisation to enable more accurate property prediction, reduce reliance on trial-and-error experimentation and enhance manufacturing precision (Table 2).

Furushima et al. [59] employed a multilayer AI framework to predict the thermal conductivity (TC) of silicon nitride ceramics prior to fabrication, using powder processing parameters and estimated relative density (RD) as key inputs for advanced technical ceramic design (Fig. 8). Random Forest regression was used for both the RD and TC prediction models. The RD model was trained on a dataset of 1,206 samples and incorporated seven explanatory variables describing the properties of the base powders, sintering additives, organic pore formers and heat treatment conditions (including nitriding and/or sintering). The resulting AI models enabled highly accurate TC prediction before manufacturing, accelerating the development cycle for Si_3N_4 ceramics. Data from published literature were used to train the models. Zong et al. (2024) [60] proposed a high-accuracy ML framework based on XGBoost to predict both thermal conductivity and bending strength of aluminium nitride (AlN) ceramics. Their model incorporated raw material properties, sintering additives and processing parameters, with SHAP analysis used to identify and interpret influential features such as oxygen content and sintering temperature. Experimental validation confirmed the model's reliability, achieving AlN ceramics with thermal conductivity up to 195.63 $W\ m^{-1}\ K^{-1}$ and bending strength of 371.60 MPa using conditions optimised by the model. Their dataset combined information extracted from patent databases and published literature.

In ceramic forming processes, Qiao et al. [61] proposed a conceptual framework that combines finite element method (FEM) and machine learning (ML) techniques to optimise the injection moulding process for manufacturing large-aperture silicon carbide (SiC) ceramic mirror. The method predicts stress and displacement during gel-casting of SiC ceramics. FEM generates temperature and stress field data, which is used to train ML models (GRNN, BPNN, ELM). Several methods were compared and the Extreme Learning Machine (ELM) model achieved the best predictive performance, enabling process parameter optimisation (temperature, pressure, loading speed) for improved product quality. In another process-oriented study, CAD modelling was combined with reinforcement learning (RL) to optimise key ceramic manufacturing parameters such as raw material ratios, moulding

TABLE 2: AI in materials development and processing of advanced ceramics.

References	Approach	Key findings	Data source
Furushima et al. (2024) [59]	Prediction of thermal conductivity prior to fabrication using multilayer AI (RF models for RD and TC)	Accelerates Si_3N_4 material development; reduces experimental workload	Published literature (1,206 samples)
Zong et al. (2024) [60]	Prediction of thermal conductivity and bending strength of aluminium nitride ceramics and identification of key processing factors	Provides interpretable ML tool for optimizing thermal and mechanical performance. Accelerates materials development	Patent databases, published literature, validated with experimental data
Qiao et al. (2022) [61]	Prediction of stress and displacement during gel-casting of SiC ceramics. Uses a conceptual framework that combines finite element method (FEM) and ML techniques	Extreme learning machine model was used to optimise SiC mirror fabrication and reduce defects and improves process	FEM simulation-generated data
Ouyang & Li (2024) [62]	Optimisation of raw material ratios, moulding pressure and sintering temperature using computer-aided design (CAD) modelling and reinforcement learning agents	Reduces time and cost; improves product strength, hardness, and appearance	Virtual CAD-based simulation data
Xu et al. (2025) [63]	Convolutional neural networks accurately predicted ceramic core deformation during sintering	Improves dimensional accuracy for aerospace ceramic components	Finite element thermo-viscoplastic simulations
Ghosh et al. (2025) [64]	ML models predicted taper angle and optimised laser drilling conditions	Improves drilling precision; reduces microcracks and experimental cost	Experimental laser drilling dataset (71 samples)

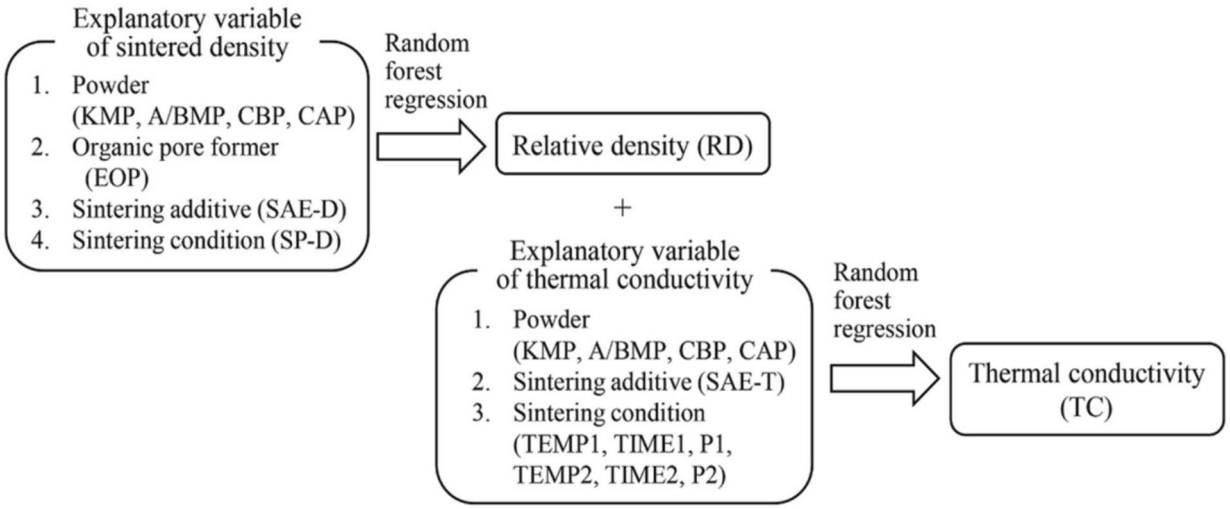

Figure 8: Conceptual diagram of the multilayer AI framework proposed by Furushima et al., 2024 to predict thermal conductivity of silicon nitride ceramics [59].

pressure and sintering temperature [62]. A simulation platform evaluated candidate parameter combinations, allowing RL agents to autonomously learn strategies that maximise ceramic performance attributes including strength, hardness and surface quality. While this approach has the potential to reduce experimental cost and time compared to conventional iterative tuning, the study relies on simplifying assumptions and lacks industrially relevant validation.

In a recent work, a CNN-based deep learning approach to predict deformation of ceramic cores during sintering was developed [63]. The model uses process data and geometry features obtained from simulations (finite element thermo-elasto-viscoplastic model for ceramic core sintering) to forecast dimensional changes, reducing trial-and-error and improving precision in aerospace ceramic components manufacturing. A finite element thermo-elastic-viscoelastic model for ceramic core sintering was employed to generate the training data. The model was experimentally validated, showing the ability to accurately predicting sintering deformation. Ghosh et al. [64] investigated AI-based predictive modelling for laser drilling of alumina ceramics. Machine learning (ML) models (Random Forest, Decision Tree, SVR) were used to predict taper angle and optimise parameters such as pulse energy, frequency, and speed, reducing reliance on costly experimental trials and improving drilling quality by predicting defects and optimal conditions. The proposed approach was experimentally validated on a small data sample.

Overall, these studies highlight the growing impact of AI and machine learning in improving the design, processing and quality control of advanced ceramics. By enabling more accurate prediction of material properties and optimising complex manufacturing parameters, AI-based methods can reduce development time and enhance product performance. However, as shown in Table 2, most of the studies are based on experimental, literature and simulation data, lacking validation and deployment in broader manufacturing environments.

Additive manufacturing of ceramics

In the area of ceramic additive manufacturing, recent studies have focused on integrating ML and closed-loop control to enhance printing stability and part quality. Zhou et al. [65] proposed a closed-loop control system using machine learning and computer vision to monitor and optimise extrusion parameters (printing speed, screw speed, nozzle diameter, height). Ensemble learning models were trained using data from experiments and used to predict deposition line width and classify line quality. The method achieves uniform deposition lines and reduces over-/under-extrusion defects. Similarly, Zhang et al. [66] introduced machine learning-based closed-loop control for ceramic robocasting. Artificial neural network model predicted extrusion length from real-time load cell data, enabling adaptive control without complex rheological modelling. The research showed improved uniformity of printed patterns and reduced defects compared to open-loop systems. Despite these advances, these studies are constrained by limitations such as small dataset sizes, which restricts the generalisability and scalability of the proposed approaches.

Recycled waste and carbon mitigation in traditional ceramics

Reduced process emissions in tile manufacturing

While reported values may vary due to changes in thickness, tile type and fuel used, a rough estimate shows that the global tile industry is producing about 100 million metric tonnes of

CO_2 annually [67] with 15–20% attributed to process emissions [68]. The subsequent section will address mitigation of process emissions via systematic integration of recycled content and replacement of calcium carbonate within body formulations to eliminate emissions from its decomposition.

Integrating recycled content in tile formulation

Ceramic tile recipes can vary significantly depending on the type of tile (floor tiles, wall tiles, etc.) and colour of body (red, white). However, almost all ceramic tile bodies rely on a triaxial blend of clays, fluxes and fillers [69]. Clays provide plasticity allowing them to be shaped and provide the necessary green strength. Fluxing agents lower the melting point of the mixture. Feldspars are usually used for floor tiles as melt to form a liquid glass phase (vitrification) that binds the other particles together, reducing porosity and increasing strength. Wall tiles on the other hand usually use carbonates such as calcium carbonate. Fillers are non-plastic materials used to provide structural stability, control shrinkage and tailor the thermal expansion coefficient for a proper glaze fit. Silica sand is commonly used as filler material [70].

A significant body of research focuses on the incorporation of waste glass into ceramic bodies. Waste glass acts as a strong flux, promoting liquid phase sintering at lower temperatures, which directly translates to reduced fuel consumption [71] [72]. It is shown in many studies that the inclusion of waste glass can lower sintering temperature by up to 200 °C. For example, one study demonstrated that adding 10% recycled glass reduced the firing temperature by 100 °C, which resulted in lowering CO_2 emissions by up to 19% [73].

In addition, other materials such as fly ash [74], granite sludge [75] and metallurgical slags [76] have been shown to successfully replace virgin materials in ceramic tile formulations while keeping or enhancing the properties. Boron mining waste has also been identified as an effective flux allowing for lower firing temperatures and reducing the need for virgin materials [77]. Other studies have explored the possibility of replacing minerals with eggshell waste [78]. While it will not reduce the CO_2 emissions during calcination, it valorises food waste. Research shows that substituting limestone with eggshell waste can increase flexural strength by 19% without negatively impacting other technical properties [79]. While all the materials mentioned above are external waste, there is another concept called "zero-waste" manufacturing which involves recycling internal ceramic waste in the tile formulation. The waste produced by the tile industry can be broadly classified into fired (broken fired tiles) and unfired waste. The fired waste is often referred to as 'pitchers' in the UK and chamotte in Europe. In this article we will use the word pitcher for fired waste.

Research using mathematical modelling has successfully developed tiles composed entirely of tile waste (sludge, raw waste, and pitcher) that meet ISO standards [80]. However, a more practical approach is to reuse the internal waste in replacing some of the virgin materials. Unfired waste has not gone through any irreversible chemical or mineralogical transformation and is already milled to a suitable particle size. Unfired waste is therefore reintroduced at the beginning of manufacturing process, a common practice in industries across the UK and Europe with approximately 95% reused [81].

In contrast, fired waste consists of materials that have undergone the sintering process but were rejected due to defects, breakage or quality control failures during sorting. Unlike unfired scrap, these materials have undergone irreversible mineralogical changes, making them hard and inert [80]. Fired scraps can account for approximately 41% of the total waste generated by the ceramic tile industry [81]. One of the main challenges is to grind the fired waste back to powder, consuming energy but making it suitable for use as a non-plastic structural component in the triaxial blend.

Case study: Integration of fired waste in ceramic tile formulations

A standard ceramic wall tile recipe is composed of four primary raw materials: ball clay (~ 50%), China clay (~ 13%), silica sand (~ 20%) and limestone (~ 12%) in addition to green clay scraps (~ 5%). A systematic study was conducted to determine the effect of pitcher (fired waste) additions at concentrations of 0, 5, 10, 15 and 20% in the recipe. The respective samples are represented by 0, 5, 10, 15 and 20P. The incorporation of pitchers did not substitute a specific material but instead, replaced part of each ingredient according to their respective percentages. All the raw materials used were collected from the industrial grade supply for Johnson Tiles UK. The ball clay, a blend of clays (CMO 764) and China clay labelled as GL2 were supplied by Imerys. Silica sand was sourced from Bathgate, and limestone was provided by Longcliffe. Pitchers were collected from the crushing machine used to crush the industry's own fired waste. To compensate for laboratory milling limitations, the pitchers were pre-screened through a 1 mm mesh. Similarly, ball clay was processed in an attrition mill to ensure sufficient fineness.

Experimental batches were prepared in 1 kg units according to each recipe. The milling process was conducted in ceramic jars using alumina balls as media. Each batch was processed with 475 g of deionised water and 3 g of sodium polyacrylate dispersant (Poliflux) to achieve a target slip density of 1.75 kg/l. The total milling duration was 190 min, with an intermediate inspection after 20 min. Any material sticking to the jars was cleared at this stage. Post-milling, the resulting

slurry was discharged through a 120 μm sieve. The rheological properties of the slurry were evaluated by measuring viscosity and density. Residue analysis was performed using a 63 μm mesh to verify the effectiveness of the milling cycle. The refined slip was tray dried in an oven at 80 °C and then subsequently crushed into powder. To prepare the material for pressing, the resulting powder was granulated by adding 6% moisture by mass and passed twice through a 1 mm sieve to ensure a uniform particle size distribution.

Two distinct geometries were produced using a hydraulic press: 4 × 2 inch samples for green strength analysis and 6 × 2 inch samples for fired property evaluation. The firing cycle was conducted in a laboratory kiln (with comparative trials performed in an industrial roller kiln). Samples were heated to a peak temperature of 1100 °C with zero dwell time to simulate fast-firing industrial conditions. The samples were furnace cooled and then key properties such as loss on ignition, linear firing shrinkage, water absorption, coefficient of thermal expansion and modulus of rupture were measured. Loss on ignition (LOI) was determined by measuring the difference between the dried sample weight and the fired weight. Linear shrinkage was calculated from the difference in length between the dried and fired samples. For water absorption (WAB) testing, dried tiles were placed in boiling water for two hours. Following this, the heat was removed, and the tiles were left in the water for an additional four hours. The tiles were then taken out. Surface moisture was carefully removed using a chamois leather before recording the weight of the soaked tiles. The modulus of rupture (MOR) was measured using a flexural testing machine where a load was applied to the tile slab until breaking occurred.

Due to the absence of a dedicated pilot facility, the development of ceramic tile recipes proceeded directly to industrial scale trials. Appreciating the fact that there will be challenges in scaling up from 1 to 1000 kg, changes were introduced incrementally through small, gradual adjustments. To achieve the required mechanical strength, the content of ball clay was progressively increased in parallel to increases in pitcher, replacing the highly energy-intensive China clay. This strategic substitution was aimed at simultaneously reducing production costs, lowering carbon emissions and meeting the required standards for ceramic tiles. Due to the numerous process adjustments made, this report will focus only on the final successful recipe to be represented by FR from here onwards. This final formulation FR (ball clay 49%, China clay 9%, limestone 11%, silica sand 16% and pitchers 15%) produced high-quality ceramic tiles at an industrial scale, encompassing various formats from small 10 × 10 cm squares to large 30 × 60 cm rectangles, and compatible with a range of glossy and matt glazes.

Ceramic tiles are subject to several technical specifications. Among these, the mechanical strength of both the green and fired bodies is of primary importance. Green strength is essential for safely transporting the tile bodies to the kiln. For safe transportation to the kiln, the body needs to have an MOR of about 3 N/mm^2 with the fired strength necessary for the final application. According to ISO EN14411, the average modulus of rupture should be no less than 15 N/mm^2. For single fired ceramic tiles, the required mechanical strength must be achieved with negligible shrinkage. Excessive shrinkage makes co-firing of glaze and body in single step impossible. A further minimum requirement for wall tiles is to have a water absorption of between 10 and 20% [82]. Table S2 summarises the data for the key properties measured in the fabricated samples.

The experimental data indicate that the increasing pitcher content leads to a measurable reduction in the MOR for both green and fired ceramic bodies. A detailed analysis reveals that the degradation of mechanical properties is not severe within the tested range. The introduction of 20 wt% pitchers resulted in a decrease in green strength of approximately 10%; however, at 15 wt% substitution level, this reduction was limited to only 6%. A similar pattern was observed in the fired samples, where the 20 wt% pitcher addition caused an 18% reduction in strength, compared to an 11% reduction at the 15 wt% level.

This decline in mechanical performance can be attributed to the substitution of plastic clay components which are fundamental for binding and structural integrity with non-plastic, inert fired material [83]. Consistent with the reduction in mechanical strength, water absorption values increased by approximately 5%, while linear contraction increased by ~ 2%. Although the addition of pitchers shows a negative correlation with these physical properties, the magnitude of deterioration remains moderate especially up to 15 wt%. In addition to material changes, mechanical properties can also be enhanced through process optimisation, such as improved homogenisation or adjusting the particle size distribution of the milled pitcher [84]. Industrial ball milling and spray drying could produce improved homogenisation and better particle size distribution compared to the manual granulation in the lab. Incorporating fired waste at 15 wt% into the forming recipe enables the systematic reincorporation of production rejects back into the process, thereby significantly reducing net waste output. Crushing a tonne of rejected ceramic pieces was estimated to cost £4.04, based on January 2024 energy prices, substantially lower than that of any virgin raw material. For instance, the cheapest raw material in the mix at that time was sand, which was sourced at approximately £25 per tonne. Initial experiments with raw materials indicated that increasing the ball clay content positively impacted mechanical properties. Consequently, to further enhance the mechanical performance, a portion of the high-cost, energy-intensive China clay was substituted with ball clay. The final recipe, after this adjustment, was tested on production lines, and the results are presented in Table S3.

The new recipe is estimated to have a process emissions carbon footprint of 6.12 kg/tonne, which represents a reduction of approximately 1 kg/tonne compared to non-pitcher recipes. Although 1 kg/tonne may seem insignificant, its impact is substantial when considering the millions of tonnes of material produced annually.

Of the 6.12 kg of carbon produced per tonne, 4.84 kg/tonne is directly attributable to the decomposition of calcium carbonate [85]. A substantial and more effective reduction could be achieved by substituting the limestone with a non-carbonate source. Calcium carbonate remains the industry's material of choice due to its easy availability and low cost. Moreover, its function is more than a simple flux. It provides structural support, enhances mechanical strength via the formation of crystalline phases such as anorthite, gehlenite and wollastonite and keeps the body porous by off-gassing CO_2 [86]. A potential replacement, however, is talc.

Talc as a potential replacement for $CaCO_3$ in ceramic tiles

Talc is a monoclinic hydrated magnesium silicate ($Mg_3(Si_2O_5)_2(OH)_2$), theoretically composed of 31.8% MgO, 63.4% SiO_2 and 4.8% H_2O. Unlike calcium carbonate, which forms calcium silicate phases, talc introduces magnesium into the ceramic matrix. While historically less common than carbonates due to cost or availability, technical literature identifies talc as a viable and advantageous substitute for carbonates in white body ceramic wall tiles. The substitution of calcium carbonate with talc fundamentally alters the mineralogical reactions during firing. In standard carbonate-based bodies, calcium oxide reacts to form phases like gehlenite and wollastonite. In talc-based bodies fired below 1050 °C, the talc does not react extensively with other components but re-combines in the solid state [87].

Upon firing, talc undergoes dehydroxylation between 900 and 1000 °C. Subsequently, it transforms into clinoenstatite and then protoenstatite ($MgSiO_3$), which can provide the mechanical strength of ceramic tiles [88]. Similarly, it can also fulfil the role of a flux. A specific mixture of 15% talc and 85% feldspar is noted to create a eutectic of remarkable interest for the ceramic industry, significantly lowering the melting point of the glassy phase [89]. In summary, talc represents a potential alternative to partially or completely replace calcium carbonate for specific applications. In wall tiles, talc may offer a meaningful reduction in process CO_2 emissions, subject to formulation level validation of firing window compatibility, water absorption, glaze fit, whiteness and raw material availability.

In conclusion, the decarbonisation of the ceramic tile industry must be approached through a clear classification of emission sources. Approximately 15–20% of total emissions are process related, primarily from the thermal decomposition of carbonates. The use of pitchers and possibly talc in tile formulations does not require kiln changing technology or green fuel and is a viable short-term method to reduce CO_2 emissions within the ceramic sector, though both approaches should be subject to life cycle assessment and technoeconomic analysis prior to industrial scale adoption.

Supply chain, life cycle assessment (LCA) and technoeconomic analysis (TEA) of ceramic technology

Hybrid LCA (HLCA) and TEA are well-established methods to assess environmental and economic sustainability. Unlike process-based LCA, HLCA quantifies direct and indirect impacts across the supply chain and the wider economic sector, removing truncation errors in process-based LCA, hence providing a holistic environmental sustainability assessment. TEA, on the other hand, quantifies the technology efficiency and economic viability, ensuring levelised cost (LCOE) and payback period are optimised. Combining both HLCA and TEA as a framework minimises the risk of trade-off between environmental, operational and economic performance, hence optimising the desired system's outcome. It is vital to perform both HLCA and TEA at the design stage of any new materials and technologies, where emerging supply chains, options, adjustments and scenarios are modelled, to identify the most sustainable manufacturing route before scaling up. Early intervention at the design stage reveals the most resilient low carbon supply chain, avoids significant emissions and optimises resource and energy efficiency. This approach, pioneered by Koh, Reaney and their respective groups, has been demonstrated in ceramics and is vital to establish relative CO_2 emissions, supply chain sovereignty and material security. The following illustrates several case studies that have used HLCA and TEA to compare substituent materials within advanced ceramic applications.

Case study: Lead-based zirconate titanate versus lead-free potassium sodium niobate piezoelectric ceramics.

Emphasis has been placed on one of the most likely piezoelectric materials, potassium sodium niobate (KNN), as a lead-free replacement for PZT [90]. KNN has been speculated to have better environmental credentials and is considered as a 'greener' than PZT. However, a comparative environmental impact assessment of the life cycle phases of KNN versus PZT piezoelectric materials found otherwise. A methodologically robust life cycle supply chain assessment based on an integrated hybrid life cycle framework is undertaken within the context of the two piezoelectric materials. Results show that the presence of niobium in KNN constitutes a far greater impact across all the

16 categories considered in comparison with PZT. The increased environmental impact of KNN occurs in the early stages of the LCA due to raw material extraction and processing. As a result, the environmental damage has already occurred before its use in piezoelectric applications during which it does not constitute any threat [90]. As such, the use of the term 'environmentally friendly' for the description of KNN should be avoided. Cost benefit analysis of substituting PZT with KNN also indicates that the initial cost of conversion to KNN is greater, especially for energy usage during production. This environmental assessment has allowed us to define and address environmental health and safety as well as sustainability issues that are essential for the future development of these materials. Overall, this work demonstrates insightful findings that can be garnered through the application of life cycle assessment and supply chain management to a strategic engineering question which allows industries and policy makers to make informed decisions regarding the environmental consequences of substitute materials, designs, fabrication processes and usage [90].

Case study: Multilayer ceramic versus Ta electrolytic capacitors

Using an integrated hybrid life cycle assessment framework, this study identified, quantified, ranked and compared the environmental impacts of the multilayer ceramic capacitors (MLCCs) and tantalum electrolytic capacitors (TECs) supply chains [91], and found while the industry-led transition from TECs to MLCCs offers both an operational and functional edge, it is also an environmentally intelligent move. Three recovery methods––incineration; hydrometallurgy and pyrometallurgy––are considered in the overall impact assessment. Electrical energy consumption during fabrication alongside the use of nickel paste are the major environmental hotspot for MLCCs. The high proportion of Ta in TECs results in an overall greater environmental impact in comparison with MLCCs, due to intensive extraction, processing and purification requirements of tantalum. Of the three recovery methods, the hydrometallurgy process offers the least environmental impact for both MLCCs and TECs [91]. Overall, the current work shows that intervention options that can further drive down the environmental impacts of MLCCs are also proposed such as a reduction in the reliance of MLCCs on rare earth elements and Cu external electrodes in some designs and material recovery.

Case study: Solid oxide fuel cells new material structures

Solid oxide fuel cells (SOFCs) have been touted to play a role in achieving a reduction in global GHG emissions, offering numerous advantages including higher efficiencies and reduced emissions, over other conventional methods of energy generation [92]. The increasing recognition and emphasis on fuel cells as a representative power generation system of the future has raised concerns over their environmental profile. Extensive research regarding the environmental profile of current structures of SOFCs can be found in the literature, but none consider the use of new materials to achieve lower environmental impacts. This research fills the gap and presents a comparison of the environmental profile of three SOFC structures: a commercially available structure, and two intermediate temperature structures, one using Erbia-stabilised bismuth oxide electrolytes and a proposed structure using strontium-doped sodium bismuth titanate electrolytes. Using a functional unit of kg/100 kW of power output for each of the SOFC structures (excluding the interconnects), within a hybrid life cycle analysis framework, the environmental hotspots across the supply chains of each SOFC type are identified, quantified and ranked [92]. The results show the use of these novel material combinations leads to a reduction in embodied materials and toxicological impact but higher electrical energy consumption during fabrication, in comparison to commercial SOFCs. The findings support the move to reduce the operating temperatures of SOFCs using these novel material architectures, which leads to an overall reduction in environmental impact due to the lower operational energy requirement of the chosen material constituents.

Case study: Solid-state versus conventional Li-ion batteries

This study compares the environmental impacts of a lithium-ion battery (LiB), utilising a lithium iron phosphate cathode, with a solid-state battery (SSB) based on a $Li_{6.4}La_3Zr_{1.4}Ta_{0.6}O_{12}$ garnet-structured electrolyte [93]. It uses a HLCA, according to two functional units, delivery of 50 MJ of electrical energy and kg of battery, to expand the system boundary. The results of the process LCA indicate that the environmental impact of LiBs is lower than SSBs across most environmental impact categories. Conversely, the input–output upstream global warming potential (GWP) of the LiBs, calculated by HLCA, is higher than that of the SSBs [93]. Sensitivity analysis shows that the SSB cycle life must increase from 100 to 2800 to achieve a GWP impact lower than that of LiBs and therefore outperform LiBs in this environmental impact category. The study, therefore, demonstrates that research into SSBs must be accelerated to achieve a functional and safe battery technology with a reduced impact on the environment.

These case studies demonstrate the strategic role of HLCA and TEA in ceramic technologies. They not only highlight the advantages of substituent versus the incumbent ceramics but also indicate unsuspected environmental hotspots. Such techniques are now routinely applied in advanced ceramic

technologies but are rarely applied to assess new processes and materials in the traditional ceramics industry. The authors strongly recommend that such methods are used more widely to assess CO_2 emissions, environmental hotspots and cost effectiveness of new materials technologies before their initiation in ceramic production.

Conclusions

The ceramic sector is one of the most difficult sectors to decarbonise within the so-called 'heavy' or 'foundation' industries. For volume production, the ceramic sector relies almost exclusively on the use of natural gas kilns, often continuous, with only a few technical ceramics such as MLCCs fired in electric tunnel furnaces. Some batch production of technical and specialist ceramics also takes place using large electric muffle furnaces. Fuel switching away from natural gas to H_2 or electrification is problematic for high volume ceramic production. Greater understanding is required of the heat transfer characteristics associated with natural gas vs H_2 vs electrification and a consensus needs to be reached as to which subsectors require the heat transfer characteristics of a flame and which, by default are more suited for electrification. Effectively, to progress in a sensible direction towards decarbonisation, the ceramics industry needs to address the fundamental question '*where does it need a flame*'.

Once each subsector is aware of the heat transfer requirements for efficient production, corporate strategies can then be explored to reduce carbon emissions in the longer term. Where a flame is required, fuel switching to H_2 and biofuels are options, provided these can be sourced at a competitive price and supply chains are available and reliable. For electrification, further effort is required to design suitable large continuous furnaces that have the potential for volume production. However, most efforts have focussed on developing H_2 furnaces or retrofitting H_2 to natural gas furnaces. The focus on H_2 has impeded technological improvements in large scale electric furnaces and this needs to be urgently addressed. Irrespective, H_2/biofuel/electrification are not technologies currently available on sufficient scale to affect the ceramic sector. Instead, the sector needs to focus immediately on existing technologies that mitigate carbon emissions, enhance energy efficiency and overall reduce the embodied carbon within their products. These immediate measures if enacted appropriately can lead to ~ 30% reduction in carbon emissions even before steps are taken to move away from fossil fuel-based production.

These technologies are badged collectively as resource and energy efficiency (REE). There are numerous REE technologies available a different technology readiness levels (TRLs), many of which are discussed in this contribution. Some (cold sintering/mechanosynthesis) are clearly insufficiently advanced (low TRL) and/or are better suited to small scale batch production and consequently not likely to present a solution for e.g. the manufacture of heavy clay products and whitewares. They may find their niche in small scale batch production of advanced ceramics and composites. However, reuse of fired waste and reduced process emissions are all viable technologies that can be introduced into the ceramic sector and can rapidly reduce their carbon footprint. The barrier here is undertaking the required research in house on the product base to ensure that properties and aesthetics are maintained. A further issue is that the core aspects of many of these technologies are outside the traditional skillset within the ceramics industry. Digital twinning and advanced monitoring and control systems are uncommon in ceramic factories that have operated in largely the same way for decades, but these technologies have been tested in other industries and are available. Heat recovery and use in e.g. spray drying is an 'off the shelf' technology that has relatively short payback periods, but capital costs are high for an industry at the margin of profitably and experiencing heavy competition from India and China whose net zero targets are a decade later than those in Europe.

If the European and UK ceramic sectors are to remain profitable while utilising net zero practices, two clear strategies need to be adopted. First, companies need to further embrace REE implemented in the short term, potentially derisked financially through government initiatives and second, greater clarity is required on the long-term decarbonisation route, whether a flame is essential or whether production, given the right investment, can switch to electric powered furnaces. The latter scenario requires an acknowledgment that wholesale electricity prices need to be competitive and that the required power infrastructure can be made available where required.

Author contributions

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