

Low-valued raw materials challenge the common eligibility criteria for triaxial ceramics



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ABSTRACT

In this work, two low-valued natural raw materials were used, namely a low plasticity and high iron content clay and a powdered rock waste generated during crushing of igneous rocks to produce construction aggregates. After characterization (chemical, mineralogical, thermal and granulometric), mixtures containing up to 75 wt% of rock powder were prepared and extruded closely following industrial practice, and fired for 60 min from 900 to 1100 °C. Property development was evaluated on as-extruded, dried and fired pieces. The results obtained were interpreted based on chemical compositions, the estimates from the Al₂O₃–SiO₂–CaO phase diagram and presumed reaction kinetics. Such interpretation showed that physical characteristics dominate not only during shaping and drying, but also during low temperature firing (slow reaction kinetics), when rock additions act as inert material; at high firing temperatures, however, the rock promotes liquid phase development after first melting (fluxing effect) and thermodynamics prevail. The dominant fluxing effect results in improved fired properties, which were shown to depend almost linearly on the liquid phase content, predicted by the phase diagram and determined by the chemical composition. These results enabled the identification of behavioural composition ranges to best exploit the materials' industrial use potential and demonstrate that current paradigms in raw material evaluation and processing in traditional ceramics industries need a re-assessment.

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1. Introduction

Clay-based ceramics industry relies on the use of the so-called triaxial compositions, which are combinations of three key types of raw materials (Fig. 1): plastic, non-plastic (or inert) and fluxes. Clay minerals, which develop plasticity upon mixing with water, play the role of plastics; non-plastics include almost always silica sand or similar rock-like aggregates, which resist melting and enable the control of clays' drying shrinkage, acting as refractory filler; and fluxes are feldspars in most cases, but also feldspathoids like nepheline, which promote the formation, at the firing temperature, of the liquid phase necessary for sintering and densification and the development of the desired final product's properties [1].

Some clay-based raw materials naturally contain a fraction of inerts and fluxes, allowing direct manufacture of less sophisticated ceramic products. However, the last decades have witnessed intense innovation, both in processing methodologies and

machinery, and in products. As a result, the industrial trend became short firing cycles and superior ceramic products with lower water absorption (open porosity) and higher mechanical strength, like porcelain stoneware [2]. Therefore, a paradigm shift in the formulation of ceramic bodies and in the selection of raw materials is being observed, which called for a reappraisal of classical treatises on ceramic raw materials and manufacturing processes [3].

The increasing scarceness of conventional ceramic natural raw materials with adequate quality, and their consequent price raise, has been driving the search for alternative raw materials that might still perform the customary roles [4]. The pressure on finding replacements for conventional fluxes, less abundant in nature and more localized than clay minerals, rose earlier. Thus, the use of local alternatives (phonolite, spodumene, wollastonite) is already comparatively frequent. Recent research on igneous rocks [5], particularly silica-unsaturated basalt, showed the fluxing potential of their additions to ceramic bodies. Other igneous rocks, such as silica-supersaturated andesite and dacite [6], have been less investigated, apparently because of their higher melting range. Knowledge about their potential as fluxing ceramic raw

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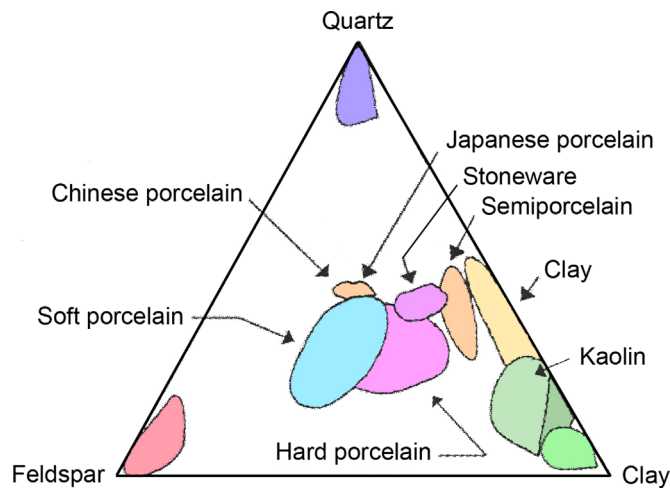


Fig. 1. Triaxial diagram for ceramic formulations (after Enrique et al. [1]).

material is practically non-existent.

On the other hand, the need to condition potentially hazardous wastes prior to landfilling, has led to the general recognition that the ceramic industry is ideal to accomplish inertization. Thus, the ceramic industry, particularly the sector dealing with construction materials, can be regarded as a potentially high consumer of wastes from other industrial sources. This also brought to light the need to re-evaluate the large quantities of minerals formerly rejected due to incomplete fulfilment of standard requirements [7–11].

As a consequence, studies of the effects of the incorporation in clay-based ceramics of a variety of unusual materials abound in the literature, aimed at partially replacing the traditional triaxial components. At a laboratory scale, however, the exact replication of the conventional industrial procedure (extrusion and slow firing) is not always easy to accomplish, and the vast majority of research papers in the literature report results obtained from small-scale dry-pressed test-pieces. Although such studies can still produce valuable results, the doubt about the industrial reproducibility of such alternative procedure usually lingers on.

Thus, today's ceramic industry still struggles to accommodate this change in the raw materials paradigm within the triaxial concept: has to make do with clay-based materials with lower plasticity or higher impurity contents, which are still labelled as "clays", has to use inert "rocks" that are far from being so, and fails to realize that fluxing character is cumulative and not exclusive of alkaline oxides [12]. In this scenario, the triaxial roles are becoming increasingly overlapped and the manufacture of ceramic products from such raw materials requires an acutely open mind, calling for a further change in traditional ceramic evaluation methodologies.

This work is aimed at bridging the processing and performance limitations of abundant low-valued natural raw materials by exploring existing synergies between chemical composition and phase equilibrium relationships. To build this case study, one low-grade clay-based material with reduced plasticity, was selected and combined with a silica-supersaturated igneous rock. The clay comes from a working quarry on a primary deposit at the Cai river valley (RS, Brazil) and could be used in red-clay ceramic products. The igneous rock comes from a local dacite dome amidst the abundant basaltic tuffs from the Serra Geral formation (RS, Brazil) and is extracted at a quarry that has been producing gravel and aggregates for the construction industry. The rock-crushing process generates a huge amount of fine powder, which, due to environmental regulations, is collected by sleeve filters but has found no use so far, and is discarded. Mixtures with tailored chemical

compositions were designed to suit specific green, dried and fired properties, which were evaluated through standard test methods and interpreted in terms of phase equilibrium diagrams.

2. Experimental

The two raw materials used in this work, clay A and rock R, were collected locally. A ~1 ton sample of clay A was dried and homogenized by quartering and lamination. The powdered rock waste (~50 kg) was used as-collected. For comparison purposes, samples of the actual rock being crushed at the time were also collected.

The raw materials were characterized in terms of mineralogical composition by X-ray diffraction (XRD, Philips/Panalytical PW 1830, Germany, using Cu K α radiation, 40 kV and 30 mA), between 0° and 70° 2 θ with 2°/min scanning rate. The identification of clay minerals was complemented by classical mineralogy XRD tests (namely, of the colloidal fraction after separation from water dispersions, after ethylene glycol immersion and after calcining at 550 °C). Chemical composition was determined by semi-quantitative X-ray fluorescence (XRF, Philips/Panalytical PW 2400, Germany, with Rh tube) on pressed pellets. Loss on ignition (LoI) was determined as weight loss upon calcination at 1000 °C for 2 h.

The thermal behaviour of the raw materials was evaluated by scanning differential calorimetry and thermogravimetry (STA 449 F3 Jupiter, Netzsch, Germany) with simultaneous analysis of evolved gases by mass spectroscopy (QMS 403 C Aëolos, Netzsch, Germany). Thermal analysis was carried out with 10 °C/min heating rate up to 1300 °C under flowing N₂ (20 ml/min).

The powders real density was determined by He pycnometry (Ultrapyc 1200e, Quantachrome, USA, 19 Psi). Three different samples of each composition (particle size < 325 mesh) were tested on 9 runs and the test result was taken as the average of the last 5 runs.

Particle size distribution (0.04–2000 μ m) was determined on very diluted deflocculated aqueous suspensions by laser scattering (LS 230, Beckman Coulter, UK) using a 750 nm wavelength laser beam (Fraunhofer model) and a Polarization Intensity Differential Scattering system (PIDS, independent of material's optical properties).

The morphology of powder particles, held on double face adhesive carbon tape on aluminium sample holders, was observed by scanning electron microscopy (SEM, Hitachi SU-70, after carbon sputtering) with electron dispersive spectroscopy (EDS, Bruker Quantax 200).

Clay+rock mixtures containing 0–75 wt% rock were prepared, moisturized (25 wt% water), homogenized and extruded under vacuum (700 mmHg, Verdés-BR051, Brazil) to produce prismatic test-pieces (20 × 15 × 100 mm³). For each mixture, extrusion moisture was calculated as the percentage weight change between as-extruded and fully dried conditions (average of the measurements of ~200 test-pieces).

The drying Bigot curve and the critical moisture content are frequently used as control parameters in industrial drying operations and were used to design the test-pieces drying schedule. Test-pieces were left to dry in the open for 24 h and then oven-dried (Tecnal TE-394/2, Brazil) in successive steps between 40 and 110 °C (22 h total drying time).

Dilatometric behaviour (DIL 402 PC/4, Netzsch, Germany) was evaluated along the extrusion direction on dried test-piece segments, with 10 °C/min heating rate up to 1150 °C in static air atmosphere (to cover the usual firing range of clay-based ceramic products).

Drying linear shrinkage was calculated from the length change of extruded test-pieces (as-extruded and after drying). For each

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