USE OF A NEW BORATE RAW MATERIAL FOR GLAZE FORMULATION

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ABSTRACT

The Rio Tinto Minerals company has developed a new borate (E-4972), which can be used in glaze formulation (patent pending WO 2007/148101).

This new borate, synthesised by low-temperature calcination, fundamentally contributes five oxides: silicon oxide (SiO_2), aluminium oxide (Al_2O_3), boron oxide (B_2O_3), calcium oxide (CaO), and sodium oxide (Na2O), its content in B_2O_3 being between 10 and 11% by weight. It is largely amorphous, and quartz is the major crystalline phase present.

The characteristics of this new borate, such as its low solubility and ability readily to form glassy phase, enable it to be used as a raw material in glaze compositions. Its suitability for glaze formulation has been the result of several years' research in collaboration with the Instituto de Tecnología Cerámica.

In this paper, the feasibility has been studied of fabricating ceramic glazes by using a new synthetic borate raw material that contributes boron to the glaze composition without this needing to be done in fritted form. It has been possible

to obtain fired glazes with similar technical and aesthetics characteristics to those obtained from industrial glaze compositions that contain typical frits in their compositions, thus enabling glazes to be formulated by using the new synthetic boron raw material.

It has been verified that use of the new boron raw material E-4972 enables glazes with different characteristics to be obtained, without a frit being needed as a raw material. The main objective in the development of this new borate raw material has been to provide frit and glaze manufacturers with an instrument that enables technical, economic, and environmental benefits to be obtained by making it possible, partly or entirely, to replace the conventional frits used in certain glaze formulations.

The results obtained show that this new raw material (E-4972) is particularly appropriate for use in producing glazes with low gloss at high temperature.

1. INTRODUCTION

The coating of ceramic bodies with a glaze layer is one of the most widespread decorating techniques in ceramic tile manufacture. This glaze layer waterproofs the body and provides the tile fair face with particular aesthetic characteristics (gloss, colour, and opacity) and technical characteristics (hardness, resistance to the chemical attack, to abrasion, or to scratching, among others) for the intended use.

The final glaze coating is obtained by the application and subsequent heat treatment of a glaze composition that is applied on to an engobe layer, which are both consolidated on a ceramic body. The glaze composition is made up of a series of inorganic raw materials. It contains silica as fundamental constituent (glass former), as well as other elements that act as fluxes (alkalis, alkaline earths, boron, zinc, etc.), as opacifiers (zirconium, titanium, etc.), and as colorants (iron, chromium, cobalt, manganese, etc.). Depending on the type of product, the firing temperature, and the effects and properties pursued in the finished product, a wide variety of glazes are formulated.

In other ceramic processes (artistic porcelain, sanitary ware), exclusively crystalline, natural or synthetic, raw materials are used in the glaze formulations, which provide the necessary oxides. In contrast, in the ceramic floor and wall tile manufacturing process, raw materials of a glassy nature (frits) are used, which are prepared from the same crystalline materials previously subjected to heat treatment at high temperature (about 1500ºC). Ceramic glazes are made up of frits, between 20 and 95% by weight of the composition, and crystalline raw materials.

Frits are typically more insoluble in water than the crystalline raw materials used to fabricate them. Frits also have a lower melting temperature and a wider

melting range; when they are used as glaze composition ingredients, they favour the obtainment of uniform final glaze coatings and the reduction of glaze surface defects.[1],[2],[3].

Boron is a common compositional constituent in glaze compositions, and is contributed as a frit, since frit solubility is lower than that displayed by other raw materials that contribute this element. In fact, the high solubility displayed by natural as well as synthetic boron-containing raw materials prevents appropriate rheological characteristics from being maintained in the glaze suspensions that might potentially be used.

2. BACKGROUND AND OBJECTIVE OF THE STUDY

For some years, the high competition in the international ceramic tile market and the presence of low cost products, essentially from Asian countries, have forced the ceramic floor and wall tile sector to set itself increasingly ambitious objectives with regard to end-product properties, cost cutting, energy saving, and minimisation of the environmental impact.

In this context, it has been sought to develop an alternative material for ceramic glaze formulations, which would contribute to overall energy saving, since it could be made at lower temperatures, while at the same time not modifying current ceramic tile manufacturing process or impairing the technical and aesthetic characteristics of tile glazes. For this reason, this material, in addition to being insoluble in water, must sinter between 800 and 1100ºC and have linear coefficients of thermal expansion (α) similar to or slightly lower than those customarily displayed by ceramic bodies (60-75 10^{-7}ºC^{-1}), as well as a sealing temperature between 950 and 1150ºC, depending on the type of end product, in order to keep defects from appearing in the final glaze coatings.

References may be found in the literature to certain studies in which it has been attempted to formulate and to prepare glaze compositions solely by using crystalline raw materials. However, in none of the cases could the formulated glazes be used without changing the industrial firing cycles. In certain cases, although these cycles were rapid, it was necessary to use maximum temperatures above 1200ºC[4], whereas in others, though the maximum temperatures used were between 1180 and 1200ºC, very long firing cycles were needed[5]. Other studies have evaluated the possibility of preparing glaze compositions without frits, with fluxing agents such as mining industry wastes or traditional borate raw materials, but the resulting glazes were either strongly coloured (12-15% Fe_2O_3)[6], and therefore of little ceramic interest, or were very fluxing, with sealing temperatures below 1100ºC,[7],[8] in addition to exhibiting a matt[9],[10] or coloured appearance[11]. As a result, it may be considered that, in practice to date, there is no evidence of industrial use of fritless glazes in the manufacture of glazed ceramic tiles.

Moreover, in relation to the addition of borate raw materials to glaze formulations, though one finds so-called low-solubility or 'insoluble' borates on the market, these display such solubility that that they cannot be used in preparing glaze slips because, even though their solubility is low, low cation solubility causes serious rheological problems.

The present paper describes the results of the study conducted with a synthetic borate developed by Rio Tinto Minerals (E-4972), involving a partly vitrified material whose properties match those indicated above, in order to determine its feasibility for use as an alternative ingredient to conventional frits in ceramic glaze compositions.

For this purpose, the new borate raw material was first characterised and compared with a frit of similar chemical composition. This was done by solubility tests and thermal tests. Glazes for use in the manufacture of earthenware wall tiles, vitrified floor tiles, and porcelain tiles were formulated. Processing feasibility was evaluated, as well as the resulting appearance and technical properties of the final glaze coatings, which were compared with those obtained in coatings resulting from glaze formulations with similar compositions using conventional raw materials (frits and other raw materials).

3. MATERIALS AND METHODS

3.1. Materials.

3.1.1. New borate raw material (E-4972).

A new borate raw material (E-4972) (patent pending WO 2007/148101 A1 'Glaze compositions') has been developed by BORAX (Rio Tinto Minerals).

The five oxides that this raw material mainly contributes, SiO_2, Al_2O_3, B_2O_3, CaO, and Na_2O, are usual frit and glaze constituents, as are also its minor constituents or impurities and the quantities in which they appear. It is a material with a high amorphous component, in which quartz is the major crystalline phase. Table 1 shows the chemical composition, table 2 presents the mineralogical composition, and table 3 details the most noteworthy thermal properties of the borate mentioned from a ceramic viewpoint.

Figure 1. E-4972.

The thermal expansion and fusibility of the new borate raw material may be considered to resemble those of the frits customarily used in glaze compositions. Its dilatometric softening temperature is about 750ºC, thus making it more fluxing than other raw materials, though not as much as some boron compounds, and it begins to sinter at about 800ºC.

SiO_2	Al_2O_3	B_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	$LOI_{(900ºC)}$
59.0±0.9(*)	18.8±0.2	10.5±0.3	0.30±0.06	5.0±0.1	0.11±0.04	4.9±0.2	0.41±0.04	0.4±0.1

(*)Variation range of the different borate batches.
Table 1. Oxide composition (% by weight) of the new borate raw material (E-4972)

Quartz (SiO_2)	Anorthite ($CaAl_2Si_2O_8$)	Wollastonite ($CaSiO_3$)	Mullite ($Si_3Al_6O_{15}$)	Cristobalite (SiO_2)	Amorphous phase 65±4
21±2(1)	5±1	3±1	6±1	<1±1	

Table 2. Constituent crystalline structures (% by weight) of the new borate raw material.

Fusion test		Dilatometric analysis	
Characterist temperatures	(ºC)	Coefficients of linear expansion	10^{-7} ºC^{-1}
Shrinkage start (T_{IC}) Shrinkage end (T_{FC}) Softening (T_R) Sphere (T_E) Semi-sphere ($T_{1/2}$) Fusion (T_F)	750±10(1) 995±10 1130±10 1240±20 1390±20 1420±30	α (50–300) α (300–500)	62±2 65±2
		Diatometric softening temperature	(ºC)
		T_{RD}	750±20

Table 3. Thermal properties (thermal expansion and fusibility) of the new borate raw material.

3.1.2. Other raw materials.

To conduct the study, other raw materials, such as boric acid, kaolins, carbonates, silicates, and feldspars that are customarily used in frit and ceramic glaze formulations, were also used.

3.1.3. Industrial reference glazes.

Three industrial glazes used for coating earthenware wall tiles (EMA), vitrified floor tiles (EMG), and porcelain tiles (EMP) were selected. All yield matt glazes, which is one of the most common finishes. The composition of these glazes is given in table 4.

Glaze	SiO_2	Al_2O_3	B_2O_3	CaO	MgO	Na_2O	K_2O	BaO	ZnO	ZrO_2	LOI
EMA	44.1	16.0	2.0	6.9	0.9	3.3	1.7	<0.1	9.0	5.2	3.9
EMG/EMP	43.8	24.0	2.2	13.8	0.5	3.3	1.7	<0.1	2.9	4.8	2.1

Table 4. Chemical composition (percentage by weight) of the references glazes.

3.2. Methods.

3.2.1. Determination of solubility.

A method that simulates glaze preparation conditions was used to determine solubility. The method consists of three phases: suspension preparation, separation of the liquid fraction, and determination of the cation content in the liquid fraction.

The suspensions were prepared by wet milling the raw materials mixture (test material and kaolin (6%)), using distilled water, in a fast laboratory mill with alumina balls. In preparation, sodium tripolyphosphate (TPP) and sodium hexametaphosphate (HMP) were used as deflocculants, and sodium carboxymethylcellulose (CMC) was used as a binder. To determine solubility, extractions were performed from the suspensions after preparation. In each extraction, the liquid fraction was separated by centrifugation and subsequent vacuum filtration using a cellulose nitrate filter of 0.2 μm pore size.

After the liquid fraction had been filtered, calcium, magnesium, potassium, silicon, and boron were determined by ICP-OES.

The solubility of the new boron raw material (E-4972) was compared with the solubility of a ceramic frit of similar composition, as well as its solubility in a glaze composition. For this purpose, different glaze suspensions were prepared whose solid fraction composition is given in Table 5.

Raw material	C 1	C2	C3	C4
Frit	94	--	31	--
New boron raw material (E-4972)	--	90	--	30
Quartz		4		
Corundum	--	--	13	11
Zircon			7	7
Nepheline			27	23
Wollastonite	--	--	20	21
Kaolin	6	6	6	6

Table 5. Composition (% by weight) of the solid fraction of the mixtures used for the solubility tests.

3.3. Characterisation of the glaze compositions.

The glazes were characterised as follows:

- Fusion test using the hot stage microscope, in which the following characteristic temperatures were determined: T_{IC} (shrinkage start), T_{FC} (shrinkage end), T_R (softening), T_E (sphere), $T_{1/2}$ (semi-sphere), and T_F (fusion).
- Dilatometric analysis, determining the linear coefficients of expansion, $\alpha_{50\text{-}300}$ and $\alpha_{300\text{-}500}$, and the dilatometric softening temperature (T_{RD}).

- Sealing temperature, or temperature range in which the glaze becomes completely impermeable. It is important that this should not be too low in order to avoid surface defects (pinholing).
- Obtainment and characterisation of glazed test pieces. Colour, gloss, and chemical resistance to strong acids (hydrochloric and lactic) and bases (potassium hydroxide) (UNE-EN ISO 10545-13:1998) were determined in the glazed test pieces obtained at different temperatures (1100ºC for the earthenware tile glaze, 1140ºC for the vitrified floor tile glaze, and 1180ºC for the porcelain tile glaze). The mechanical properties, Vickers hardness (EN 843-4: 2005), Berkovich microhardness (CEN/TS 1071-7), modulus of elasticity (Young) (CEN/TS 1071-7), toughness (ASTM C 131:1996), and abrasion resistance (UNE EN ISO 10545-7: 1999), were determined in the glazes intended for the manufacture of vitrified floor tiles and porcelain tiles. The microstructure of the resulting glaze coatings was characterised by scanning electron microscopy (SEM) and X-ray diffraction (XRD).
- Pilot plant tests. Glazes were prepared with the borate in discontinuous alumina ball mills, referenced 'Pilot', and each glaze was subsequently applied by bell waterfall on to the body matching its characteristics.

4. RESULTS AND DISCUSSION

4.1. Solubility of the new raw material.

Table 6 presents the results, in solubilised mg L^{-1} of different cations using two types of deflocculants (TPP and HMP).

	B	Ca	Mg	K	Si
CI-TPP	370 ± 26	200 ± 21	16 ± 6	17 ± 4	326 ± 50
C2-TPP	579 ± 21	275 ± 33	19 ± 2	21 ± 6	305 ± 45
C1-HMP	259 ± 33	173 ± 18	3 ± 2	13 ± 3	214 ± 58
C2-HMP	547 ± 31	245 ± 29	8± 2	23 ± 6	300 ± 45
C3-TPP	138± 11	297± 38	11± 3	210± 11	447± 42
C4-TPP	237± 25	323± 41	14± 4	130± 10	492± 45
C3-HMP	97± 9	288± 26	9± 2	155± 10	480± 34
C4-HMP	242± 18	295± 25	11± 2	120± 9	512± 45

Table 6. Intrinsic solubility (mg L^{-1}) of the frit and of the new borate raw material using TPP and HMP as deflocculants.

As may be observed, the quantity of boron solubilised by the suspension prepared with the new borate is similar, though slightly higher than that solubilised by the standard composition with a frit and lower than that corresponding to other borates that might be used in ceramic glaze preparation [12].

Of the solubilised cations, Ca^{+2} [13] has the greatest influence on the rheological behaviour of the suspensions, followed by Mg^{+2}, even in relatively low concentrations (10^{-2} M≈400 mg Ca^{+2} L^{-1} ≈ 250 mg Mg^{+2} L^{-1}). However, the presence of K^{+} and B^{+3} only affects this rheological behaviour when concentrations of the order of 10^{-1} M (≈3900 mg $K^{+}L^{-1}$ ≈1100 mg $B^{+3}L^{-1}$) are reached. In fact, in practice, K^{+} may be considered hardly to alter the rheological properties of the suspensions [14].

In view of the data presented in the tables, it is clearly observed that boron is quite a soluble element compared with other cations, owing to its high charge/mass ratio, though it does not exceed a concentration of 1100 mgB.L^{-1}, above which it has been demonstrated[14] that the rheological properties of glaze suspensions are affected. Boron is thus solubilised by the new borate raw material to a much smaller extent than by other borate raw materials, whether synthetic or natural [12], though it does so to a slightly greater extent than the frit, which makes its use feasible in ceramic glaze preparation.

4.2. Glaze formulation with the new boron raw material.

Glaze compositions were prepared for the earthenware tile (EMA), vitrified floor tile (EMG), and porcelain tile (EMP). The glaze compositions all produced matt glazes, this being the most common floor tile finish. In the case of the glaze for the earthenware wall tile, a matt appearance was also chosen, though this is not as usual, owing to the difficulty of obtaining glossy finishes with an appropriate texture [15].

The characterisation of the three ceramic glazes (table 7) shows that glaze compositions can be prepared for different types of ceramic tiles, without it being necessary to use typical frits as raw materials. These glazes display similar characteristics to those of traditional glazes, the borate content being larger than 20% by weight in the test glazes.

		EMA-F	EMA-B	EMG-F	EMG-B	EMP-F	EMP-B
Glaze composition (% by weight)	Frit	64	--	25	--	25	--
	E-4972	--	32	--	24	--	24
	Quartz	--	--	4	4	4	4
	Wollastonite	--	32	19	19	19	19
	Sodium feldsp.	30	28	--	7	--	7
	Nepheline	--	--	25	17	25	17
	Zinc oxide	--	--	2	4	2	4
	Corundum	--	--	12	10	12	10
	Zircon	--	--	7	7	7	7
	Kaolin	8	8	6	8	6	8
Fusion test (ºC)	T_{IC}	860	895	910	900	910	900
	T_{FC}	1020	1090	1105	1090	1105	1090
	T_{R}	1115	1110	1125	1140	1125	1140
	T_{E}	1130	1145	1150	1190	1150	1190
	$T_{1/2}$	1170	1185	1170	1225	1170	1225
	T_{F}	1205	1225	1225	1260	1225	1260

Dilatometric thermal expansion	$\alpha_{50\text{-}300}\cdot 10^{-7}$ ºC^{-1} $\alpha_{300\text{-}500}\cdot 10^{-7}$ ºC^{-1} T_{RD} (ºC)	65 74 792	71 71 1047	72 72 1050	68 71 975	71 72 1009	66 67 997
Sealing temperature	Range (ºC)	960-990	1020-1040	1060-1070	1040-1050	1060-1070	1040-1050
Colour (engobed pieces)	L* a* b* Ib Ia	92.1 -0.48 5.11 74.2 10.0	90.7 -0.79 6.27 69.9 12.0	89.6 -0.82 2.39 79.8 4.4	90.2 -0.55 2.05 81.5 3.9	90.4 -0.68 3.68 78.2 7.0	91.9 -0.43 3.28 79.9 6.4
Gloss	$(Gloss)_{T(ºC)}$	$(45)_{1100}$	$(30)_{1100}$	$(8)_{1400}$	$(4)_{1140}$	$(9)_{1180}$	$(6)_{1180}$
Chemical resistance (1)	HCl(2) Lactic acid(3) NaOH (2)	GHB GHA GHA	GHB GHA GHA	GHB(v) GHB GHA	GHB GHB GHA	GHB(v) GHB GHA	GHB GHB GHA
Mechanical properties	Vickers hardness Berkovich microhardness (GPa) Modulus of elasticity (GPa) Toughness (MPa•m$^{1/2}$) Abrasion resistance (weight loss at 6000 rev (mg))			5.4±0.1 9.3±0.8 85 0.9±0.1 25±5	5.6±0.2 9.5±0.8 101 1.1±0.1 10±6	5.5±0.3 11.2±0.8 114 1.1±0.1 24±2	5.3±0,3 9.3±0,8 77 1.0±0.1 8±5

(1) GHA no visible effects GHB visible effects when pencil lines are drawn and GHB (v) definite change in appearance (2) (18% v/v) (3) (0.1 kg L^{-1}).

Table 7. Characterisation of the glaze compositions obtained using frits (-F) and the new boron raw material E-4972 (-B).

The technical and aesthetic characteristics of the glazes for vitrified floor tiles and porcelain tiles, as well as those of glossy opaque and matt glazes for earthenware wall tiles, obtained from glazes formulated with the new borate raw material, are all fully comparable to those of their commercial counterparts obtained from frit-containing glaze compositions. In the glazes for porcelain and stoneware tiles, abrasion resistance and resistance to chemical attack were even improved.

4.3. Function of the new boron raw material in the glaze composition.

When a thermal fusion test was performed of a glaze made up only of raw materials (MP) and of other glazes to which the new boron raw material (MP-B) or a frit (MP-F) was added, an earlier shrinkage start was observed in both cases, due to glassy-phase formation in these glazes during the first sintering stages (figure 2). In the case of the frits, however, shrinkage start occurred at slightly lower temperatures, though shrinkage end coincided with that of the glaze formulated with the new boron raw material. This is because the frit fusion range is a little wider.

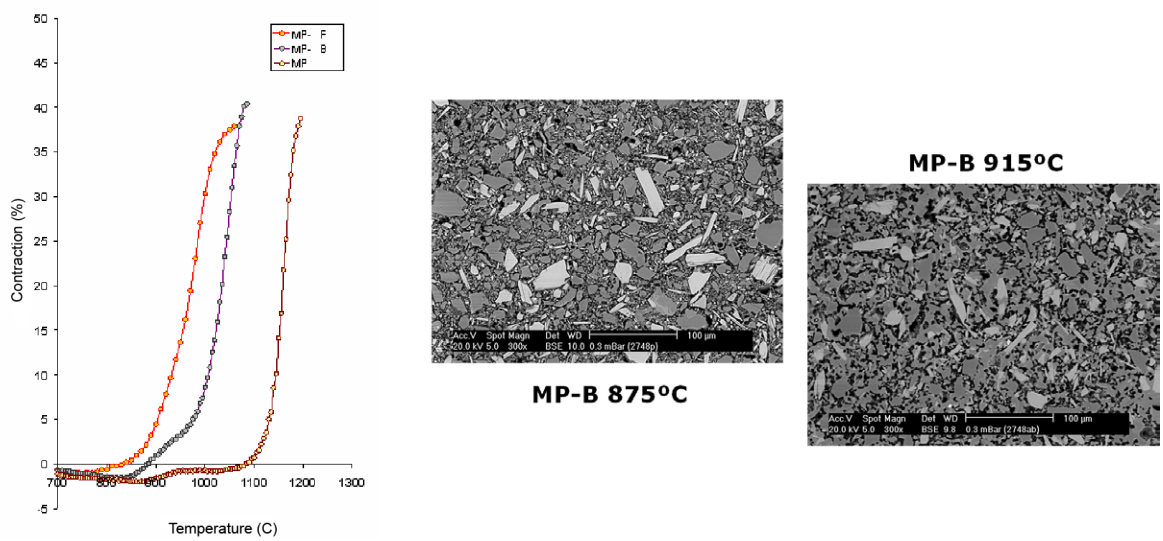

Figure 2. Shrinkage curves of glazes MP, MP-B, and MP-F and micrographs of the glaze prepared with raw materials and the new boron raw material during the first sintering stages.

The glazes obtained from the borate were microstructurally characterised. For this purpose, the crystalline structures present were identified by X-ray diffraction, and glaze cross-sections were observed, photographed, and analysed with a scanning electron microscope, coupled to an energy-dispersive X-ray microanalysis (EDAX) unit. Figures 3 to 5 show micrographs of the glaze cross-sections, in which the different identified crystalline structures have been labelled.

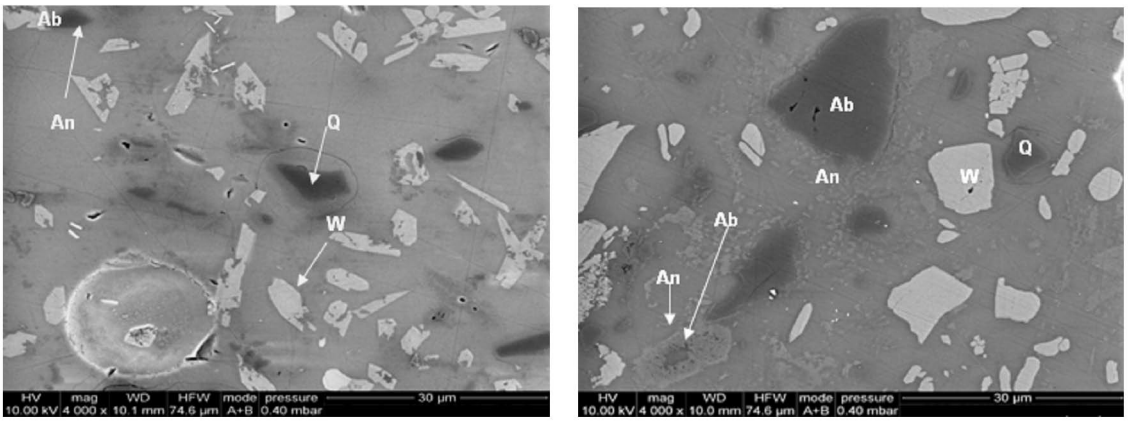

Figure 3. Cross-sections of glazes EMA-F and EMA-B. Identified crystalline structures: Q= Quartz (SiO_2), W= Wollastonite ($CaSiO_3$), An= Anorthite ($CaAl_2Si_2O_8$), and Ab= Albite ($NaAlSi_3O_8$).

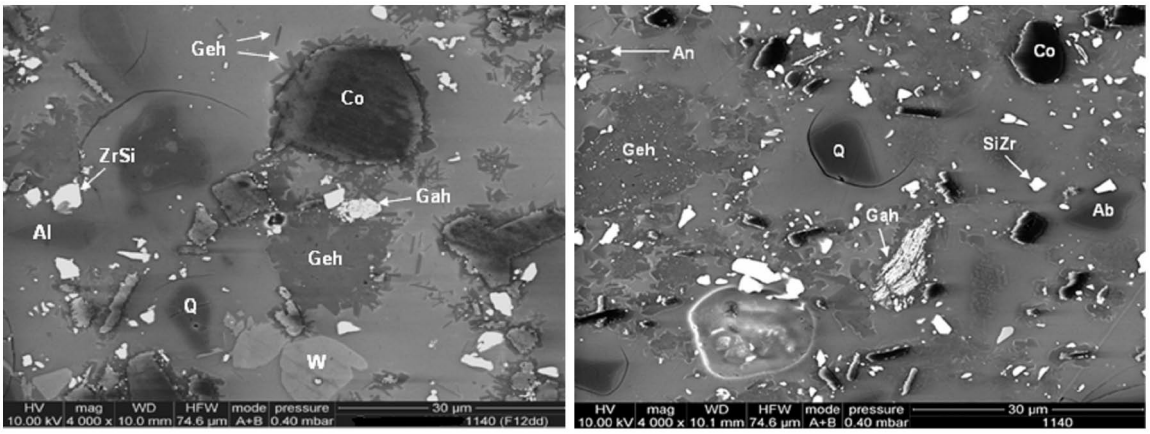

Figure 4. Cross-sections of glazes EMG-F and EMG-V. Identified crystalline structures: Q= Quartz (SiO_2), Co= Corundum (Al_2O_3), ZrSi= Zircon ($ZrSiO_4$), and Geh= Gehlenite ($Ca_2Al_2SiO_7$) (10 kV) (4000x).

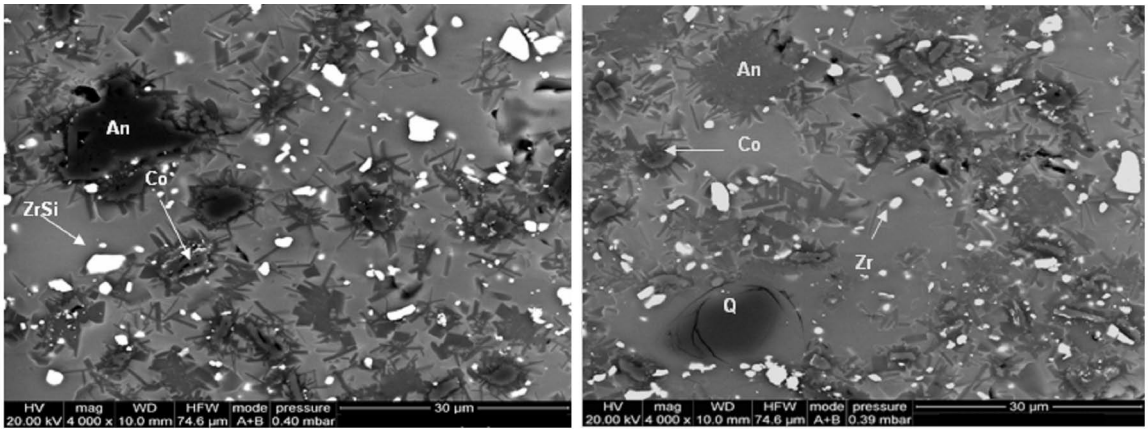

Figure 5. Cross-sections of glaze PG. Identified crystalline structures: ZrSi= Zircon ($ZrSiO_4$), Co= Corundum (Al_2O_3), Geh= Gehlenite ($Ca_2Al_2SiO_7$), and Gah= Gahnite ($ZnAl_2O_4$) (4000x) (20kV).

This characterisation demonstrated that the glaze microstructures were similar and that the main function of the borate was glassy-phase formation, favouring, just like the frits, the formation during firing of crystalline structures that contain some borate elements (Si, Al, and Ca): Anorthite ($CaAl_2Si_2O_8$) and Gehlenite ($Ca_2Al_2SiO_7$). In addition, in these glazes, borate is the source of the identified quartz particles.

4.4. Trials in a pilot plant.

The facility used for wet milling was a pilot plant ball mill (figure 5), with a useful diameter of 50cm and length of 53cm, involving a useful volume of 104 dm3. The mill operates at a rotational speed of 47 r.p.m. The milling media used were alumina balls. Glaze compositions EMA-B, EMG-B, and EMP-B were prepared at a solids content of 72%, with the addition of 0.3% deflocculant (TPP), 0.3% adhesive (CMC), and 28% water. When the milling step had ended, the material was discharged and sieved in a vibratory screen with a mesh aperture of 500 µm. The glaze was applied in a pilot-scale glazing line (figure 5).

The resulting test pieces were fired in industrial kilns at the temperature required by the body. The pieces were fired at two companies (Pilot-1 and Pilot-2); the thermal cycles used were those that the company set when firing was to be carried out. The chromatic coordinates of the glazes after their industrial firing are shown in table 8.

Figure 6. View of the mill used for the trials, and bell waterfall used in the trials before and during the glazing process.

Reference	Tª(ºC)	L*	a*	b*	WI	YI	Gloss
EMG	1140	89.6	-0.82	2.39	79.8	4.4	8
EMG-B-Pilot-1	1140	89.6	-0.58	2.75	78.7	5.3	5
EMG-B-Pilot-2	1140	89.0	-0.85	3.03	77.2	5.7	9
EMP	1180	90.4	-0.68	3.68	78.2	7.0	9
EMP-B-Pilot-1	1180	91.2	-0.49	4.02	76.9	7.8	8

Table 8. Chromatic coordinates. Whiteness index (WI), yellowness index (YI), and gloss of the glazes prepared on a pilot plant scale, formulated with the new boron raw material, and of the reference glazes.

No problems appeared during this firing: the pieces obtained had a good surface appearance and displayed no significant curvatures.

These results demonstrate that the glazes obtained using the new borate raw material produced no problems during the tile firing process and provided glaze finishes with similar characteristics to those obtained with conventional frits.

5. CONCLUSIONS

The results obtained in this study allow the following conclusions to be drawn:

- The feasibility of the use of a new synthetic borate (E-4972) (patent pending 2007/148101 WO A1) as a raw material in formulating glazes has been demonstrated.

- The quantity of boron solubilised by the new raw material analysed in this study is low, and is similar to, albeit a little higher than that of a conventional ceramic frit.
- This new borate readily forms glassy phase and can be used as a fluxing raw material in ceramic glazes. This has been verified by the development, with the same compositions, of fritless glazes that can be used as coatings for wall and floor tiles, with good results from both an aesthetic and a technical viewpoint.
- It is feasible to replace the entire frit with mixtures of raw materials and the new synthetic borate in order to produce commercial glazes, in particular, matt glazes.
- The technical and aesthetic characteristics of the glazes for vitrified floor tiles and porcelain tiles, as well as those of the glossy opaque and matt glazes for earthenware wall tiles, obtained from glazes formulated with the new borate raw material, are fully comparable to those of their commercial counterparts obtained from frit-containing glazes.

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