

Fabrication and characterization of aluminium titanate and mullite added Porcelain ceramics

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In this study, aluminium titanate (AT, Al_2TiO_5) and mullite (M, $3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$) ceramics were synthesized by reaction sintering from Al_2O_3 , SiO_2 and TiO_2 powders which consisted of homogenization by wet ball milling followed by sintering AT and M in air at 1550 °C for 2 hrs and 1400 °C for 2 hrs, respectively. AT and M in different amounts (0 and 20 wt. %) were mixed with Porcelain (P). AT and M reinforced porcelain ceramics were prepared by the powder metallurgy route. After drying, the powders were compressed to preforms of 56x12x10 mm by uniaxial pressing at 200 MPa. The green compacts were sintered at 1100-1200 °C for 1-5 hr under air. Investigations were carried out, on the effect of addition of AT and M in terms of physical properties, microstructure, XRD phases, as well as wear and mechanical properties of AT and M-reinforced porcelain ceramic composites. Phase and microstructural characterizations of the sintered materials were carried out by using X-ray diffraction technique (XRD) and scanning electron microscope (SEM). The Micro Vickers hardness testing was performed using the Shimadzu HMV-MIII hardness tester.

Key words: Aluminium Titanate, Mullite, Porcelain, characterization, Hardness.

Introduction

Porcelain is a hard, fine-grained, nonporous, and usually translucent, highly vitrified and white ceramic ware that consists essentially of kaolin, quartz, and a feldspathic rock and is fired at a high temperature. Porcelains typically have a triaxial formulation comprising about 50% clay, 25% flux and 25% filler. It is a ceramic material that can be used for technical or artistic purposes, with or without glazing, as well as semi-light transmittance. Porcelain represents one of the most complex ceramics, formulated from a mix of clay, feldspar and quartz, sintered at temperatures between 1200 °C and 1400 °C to form a glass-ceramic composite. The clay is typically comprised of kaolinite, which confers plasticity to green paste and is the precursor of mullite crystals. The fluxing agent is feldspar and the filler is quartz, which most likely leads to higher strengths of the unfired tiles. Firing bodies containing these three components exhibit a grain and bond microstructure, which consists of coarse quartz grains joined by a finer bond or matrix that contains mullite crystals and a glassy phase [1-7].

Aluminium titanate (AT, Al_2TiO_5) exhibits good thermal shock resistance and low thermal conductivity, chemical resistance in molten metals. AT ceramics

have low thermal expansion coefficient, low thermal conductivity and excellent thermal shock resistance enabling them for applications in glass, automotive and thermal processing industries. These properties and in particular the thermal shock resistance make AT a promising candidate reinforcement in ceramic systems, whereas the expansion coefficient of AT is anisotropic and shows large mismatch with matrix alumina [2,8-11].

Mullite (M, $3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$) is the unique stable intermediate crystalline phase of the binary system Al_2O_3 - SiO_2 . M is a good and low cost refractory ceramic. Its applications are very diverse and cover from refractory field to technical applications. In Amongst structural engineering materials, M ceramics have many desirable properties and very good thermo-mechanical properties such as excellent high-temperature strength and creep resistance, good thermal and chemical stability. Its thermal expansion coefficient is relatively low leading to a good thermal shock resistance [12-17].

In this study, the effects of AT and M added porcelain ceramic composites produced by the traditional ceramic production process on the physical and microstructural properties were investigated.

Experimental

The powders (Porcelain, Al_2O_3 , SiO_2 , and TiO_2) used in this study were obtained from the Eczacıbaşı

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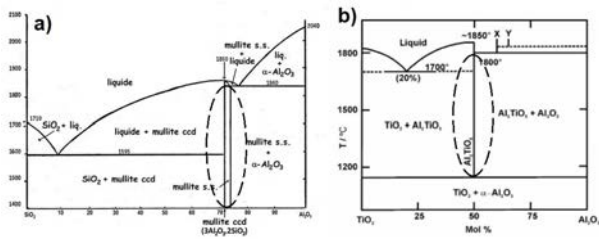


Fig. 1. Binary phase diagram of (a) $\text{Al}_2\text{O}_3\text{-SiO}_2$ and (b) $\text{Al}_2\text{O}_3\text{-TiO}_2$ system proposed by Goldberg et al. in 1968 [18-19].

(Eczacıbaşı Esan, Türkiye). In this study, AT and M added porcelain ceramic composites fabricated by the conventional ceramic production process. Mixtures were prepared by mechanical alloying method and used alumina ball mill to homogenize the blend with acetone as medium. The powders were dried in oven at 110°C for 24 hrs before mixing. AT and M ceramics were synthesized by reaction sintering from Al_2O_3 , SiO_2 and TiO_2 powders which consisted of homogenization by wet ball milling followed by sintering AT and M in air at 1550°C for 2 hrs and 1400°C for 2 hrs, respectively. AT and M in different amounts (0 and 20 wt. %) were mixed with Porcelain (P). Then, additions of AT and M in weight proportions of 0 and 20 wt.% were made to the PMAT mixture (hereafter these mixtures are denoted as P, PM, PAT and PMAT respectively). AT and M reinforced porcelain ceramics were prepared by the powder metallurgy route. The samples were wet

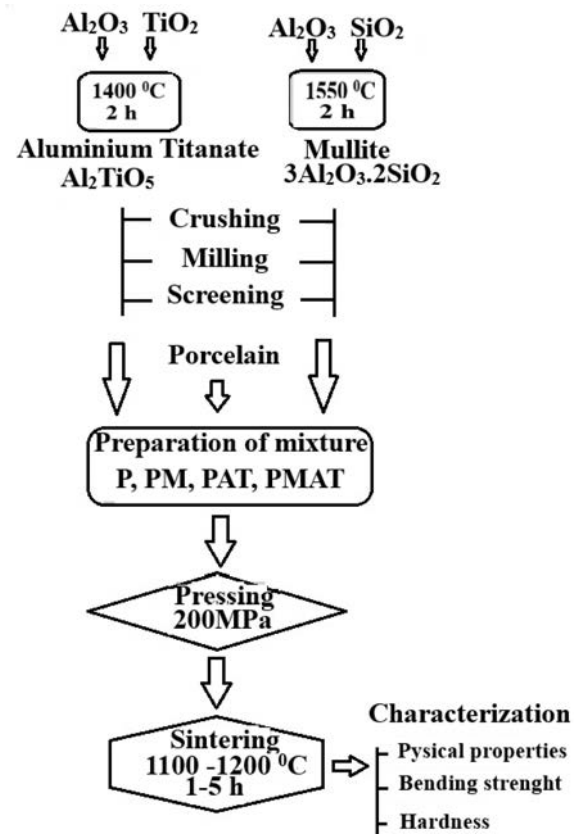


Fig. 2. A schematic illustration of the powder metallurgy route to prepare aluminium titanate and mullite added porcelain ceramic composites.

Table 1. Bulk density and porosity, relative density and water absorption values of the mullite and aluminium titanate-reinforced porcelain ceramics obtained.

	Time	Temp.	Bulk density	Relative density	water absorption	Porosity	Bending Strength
	h	$^\circ\text{C}$	gr/cm ³	%	%	%	N/mm ²
P	1	1100	2,258	90,320	5,011	9,680	53,171
PAT	1	1100	2,234	79,502	6,710	20,498	37,423
PM	1	1100	1,856	82,682	11,200	15,636	25,273
PMAT	1	1100	2,058	67,921	12,263	32,079	19,884
P	5	1100	2,363	94,520	2,187	5,480	57,254
PAT	5	1100	2,382	84,769	4,687	15,231	59,345
PM	5	1100	1,945	88,409	9,132	11,591	30,817
PMAT	5	1100	2,089	68,944	11,852	31,056	22,005
P	1	1200	2,415	96,600	1,429	3,400	65,666
PAT	1	1200	2,529	90,007	1,316	10,000	75,872
PM	1	1200	1,986	92,128	4,810	9,727	50,643
PMAT	1	1200	2,230	73,597	8,591	26,403	31,598
P	5	1200	2,448	97,920	0,293	2,080	68,583
PAT	5	1200	2,572	91,525	0,288	8,475	91,660
PM	5	1200	2,012	93,263	2,715	8,545	62,237
PMAT	5	1200	2,344	77,358	6,159	22,642	37,950

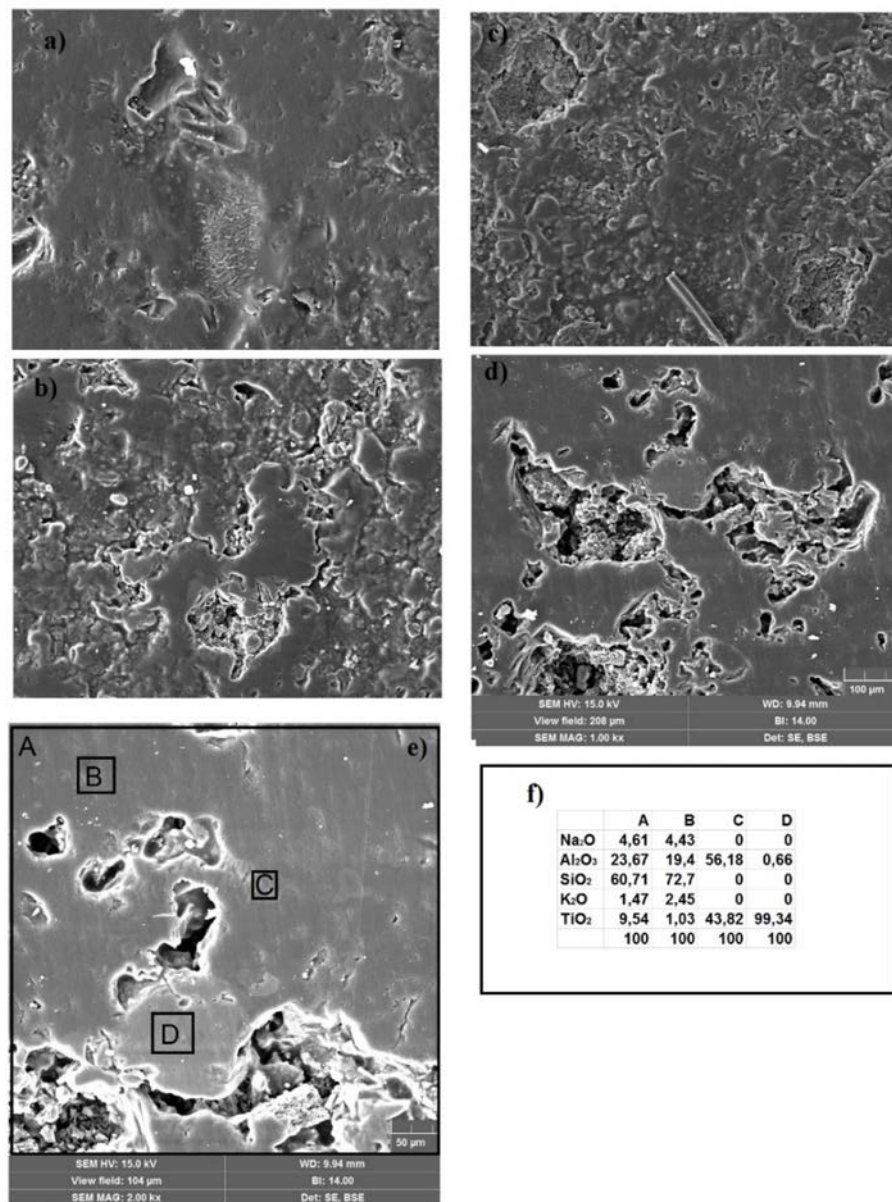


Fig. 3. SEM images of (a) porcelain (P), (b) porcelain- aluminium titanate (PAT), (c) porcelain - mullite (PM) and (d) porcelain - pluminium titanate-mullite (PMAT) ceramic composites obtained. Samples were sintered at 1200 °C for 5 hrs. The insets are the higher magnified view relative to the images. Arrows indicate incidence of microcracks formed after sintering process. (e) PMAT ceramic sample at 2000x (f) Representative result of EDS analyses performed on the PMAT sample sintered at 1200 °C for 5 hrs.

milled with alumina balls for 3 hrs and sieved. After drying, the powders were compacted to preforms of 56x12x10 mm by uniaxial pressing at 200 MPa. The green compacts were sintered at 1100-1200 °C for 1-5 hrs under air using a heating rate of 5 °C min⁻¹ in a high temperature furnace (Protherm Furnace, Turkey). Fig. 1 shows Binary phase diagram of Al₂O₃-SiO₂ and Al₂O₃-TiO₂ system proposed. Fig. 2 shows a schematic powder metallurgy fabrication flowchart of AT and M added porcelain ceramic composites.

The densities, porosity and water absorption of the sintered specimens were measured by the Archimedes method. The morphological parameters of the various

phases were characterized by using a semiautomatic image analyser, EDX and the formed phases were analysed by X-ray powder diffractometer (Rigaku, Dmax, IIIC) using Cu K α radiation. After sample sections were mechanically, the microstructural characterization of the sintered samples was carried out using scanning electron microscopy (SEM, Tescan Mira). Micro hardness (Shimadzu, HMV-MIII) was measured on the polished surface of the samples at room temperature. At least six individual tests with a peak load of 1000 g and a loading time of 20 s were performed for each set of composites.

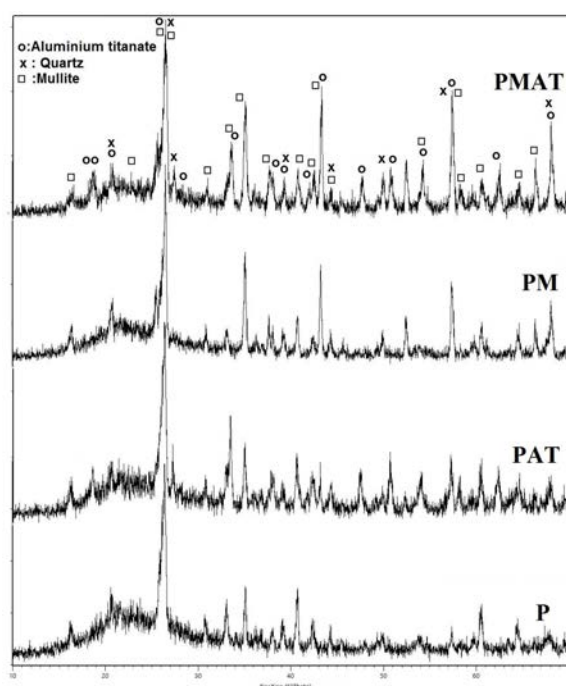


Fig. 4. XRD patterns of : porcelain (P), porcelain-aluminium titanate (PAT), porcelain - mullite (PM) and porcelain-aluminium titanate-mullite (PMAT) ceramic composites after sintering at 1200 °C for 5 hrs.

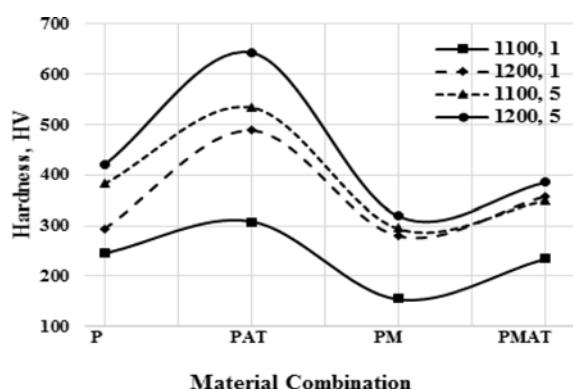


Fig. 5. Micro Vickers hardness values of the specimens.

Results and Discussion

The physical properties of AT and M-reinforced porcelain ceramics samples, sintered at 1100-1200 °C for 1-5 hrs are given in Table 1 [2, 3, 5, 7, 14].

As the temperature and sintering time increases, the density, relative density and strength are increasing, while the water absorption and porosity decreases. Relative density decreased with the addition of AT and M. Water absorption and porosity increased with the addition of AT and M. In the strength measurements, increased sintering temperature and time increased with the addition of AT added porcelain, while there was some decrease in the porcelain with M addition.

Fig. 3(a-d) are typical SEM images taken from the surfaces of AT and M -reinforced porcelain ceramic

composite samples (P, PAT, PM and PMAT). It can be seen that P appears to be denser and to have fewer pores than the samples PAT and PM. Approximately 40% of the P samples are more intense in the vitreous phase. AT and M are uniformly dispersed in the porcelain matrix. The addition of AT and M influences the grain morphology, as can be seen in the microstructures of the composites. The grain size grows, as the amount of AT and M increases. 3(e) and 3(f) shows the microstructure of PMAT sample used for phase analysis with EDS. Analysis has revealed that the matrix consists of mullite, quartz and aluminium titanate phases: the A, B and C areas demonstrated porcelain, AT and M, which consist of K, Al, Si, O and Ti [2, 3, 5, 7].

Fig. 4 shows the representative XRD patterns of the ceramic composites (namely P, PAT, PM and PMAT) which were sintered at 1200 °C for 5 hrs. XRD patterns of the resultant AT and M-reinforced porcelain ceramic composites revealed that the main phases are aluminium titanate (Al_2TiO_5 , PDF card No. 01-070-1434), mullite ($3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$, PDF card No. 00-015-0776) and quartz (SiO_2 , PDF card No. 01-078-1252). The AT and M phases became present in the pattern of the PAT and PM sample first, while an addition of 20 wt. % AT and M completed [2, 3, 5, 7, 12, 13].

Conclusions

AT and M ceramics were synthesized by reaction sintering from Al_2O_3 , SiO_2 and TiO_2 powders which consisted of homogenization by wet ball milling followed by sintering AT and M in air at 1550 °C for 2 hrs and 1400 °C for 2 hrs, respectively. AT and M added porcelain ceramic composites have been prepared by powder metallurgy process. As the sintering temperature and sintering time increases, the density, relative density and strength are increasing, while the water absorption and porosity decreases. As the temperature and sintering time increases, the density, relative density and strength are increasing, while the water absorption and porosity decreases. Relative density decreased with the addition of AT and M. Water absorption and porosity increased with the addition of AT and M. In the strength measurements, increased sintering temperature and time increased with the addition of AT added porcelain, while there was some decrease in the porcelain with M addition. It can be seen that P appears to be denser and to have fewer pores than the samples PAT and PM. Approximately 40% of the P samples are more intense in the vitreous phase. AT and M are uniformly dispersed in the porcelain matrix. The addition of AT and M influences the grain morphology, as can be seen in the microstructures of the composites. Phase analysis has revealed that the matrix consists of mullite, quartz and aluminium titanate phases. As the temperature and

sintering time increased, the hardness of the all samples increased. In the hardness measurements, increased sintering temperature and time increased with the addition of AT added porcelain, while there was some decrease in the porcelain with M addition.

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