TUNISIAN SMECTITIC CLAYS AS RAW MATERIALS FOR THE PRODUCTION OF LIGHTWEIGHT AGGREGATES

Emma FAKHFAKH [1], **Islem CHAARI** [1], **Mounir MEDHIOUB** [2], **Fernando ROCHA** [3]*, **Celso GOMES** [3], **Joao Correira MARQUES** [4], **Joao COROADO** [5], **Alberto LOPEZ-GALINDO** [6], **Fethi KOOLI** [7], **Foued ZARGOUNI** [8] **and Fakher JAMOUSSI** [1]*

[1] *Laboratoire de Géoressources INRST BP 95, 2050 Hamam-Lif, Tunisia,*
[2] *Département des Sciences de la Terre, Faculté des Sciences de Sfax, Tunisia*
[3] *Univ. Aveiro, Campo de Santiago, 3810 Aveiro, Portugal*
[4] *Centro tecnologico da Cerâmica e do vidro, Rua Coronel Veiga Sima, 3000 Coimbra*
[5] *MIA-Centro de Investigaçao "Minerais Industriais e Argilas" da Fundaçao para a Ciência e a Tecnologia (FCT), Instituto Politécnico de Tomar*
[6] *IACT.CSIC-Univ. Granada. Avda. Fuentenueva, 18002-Granada, Spain*
[7] *ICES, 1Pesek Road, Jurong Island 62 7833, Singapore*
[8] *Département de Géologie. Faculté des Sciences de Tunis. 1060 Belvédère, Tunisia*
Corresponding author's e-mail: Fakher.Jamoussi@inrst.rnrt.tn, frocha@geo.ua.pt

ABSTRACT
Smectitic clays from 12 deposits located in the Meridional Atlas domain (Tunisia) are explored for manufacturing of expanded aggregates. Two types of firing processes (slow and quick firing) were carried out, and properties of the bloated products were examined. Used raw materials are mainly composed of smectite, kaolinite and illite phases with quartz, calcite, dolomite and feldspars as impurities. They contain high amounts of fluxes (Fe_2O_3, CaO, MgO, Na_2O and K_2O) with CaO and Fe_2O_3 dominating others. Quick firing or "flash heat" must be preceded by two hours drying at 350°C in order to avoid any aggregate explosion which seems to be related to the fineness of a raw material. The clay fineness appears to be the cause of ball explosion. Clay aggregates were heated at the maximum bloating temperature (1170 °C) during 5 min. and then characterized through a determination of the apparent density, the mechanical resistance corresponding to the rupture under point compression, and water absorption measured after 72 hours of immersion in distilled water. Obtained results are encouraging and may open new perspectives for the utilization of some investigated smectitic clays in civil construction works.

KEYWORDS: smectite, light weight aggregate, firing processes, apparent density, Meridional Atlas domain, Tunisia

1. INTRODUCTION

Lightweight aggregates (LWA) are defined as either natural or artificial materials with certain granular, porous and light characteristics. These materials are produced from different natural sources such as volcanic rocks (pumice, pozzolan, volcanic tuffs…), sedimentary rocks (diatomites…), sedimentary to metamorphic rocks (clays, slates…) or from industrial by-products such as glass, ash, etc. (Harris et al., 1962, Cougny, 1990). LWA have gained a lot of interest due to their good thermal and acoustic insulation features along with a good fire resistance. In fact, these materials are used as an essential component of numerous construction products. They are used in the production of a wide range of building blocks, lightweight aggregate concretes, structural concrete as well as ballast for rail-road or are associated with bituminous materials for road coating.

The combination of a porous character of LWA materials with their high water adsorption makes them promising and excellent candidates for the hydro phonic agriculture because of being sterile, neutral, light and long-lasting. In this study, smectitic clays from twelve deposits were tested and examined as a raw material for the synthesis of expanded aggregates using different methods of quick or slow firing processes. The physical properties of the bloated materials are also reported.

2. MATERIALS

Researched smectitic clays came from twelve deposits located in the Meridional Atlas domain (Fig. 1). The samples were collected from different locations. AYD1, AYD2 clays from Jebel Aidoudi (El Hamma region) and Jebel Berda (JBD1) are of Coniacian to Santonian age and belong to Aleg formation (Burrolet, 1956). The samples from Oued Tefal (TFL1, Jebel Stah), Jebel Hamadi (HMD1) and Jebel Orbata (KB) are of Campanian to Maastrichtian age and belong to the middle member of Abiod formation (Jamoussi, 2001). The clays from Jebel Sehib (SHB1), Jebel Chamsi (CHM1) and from
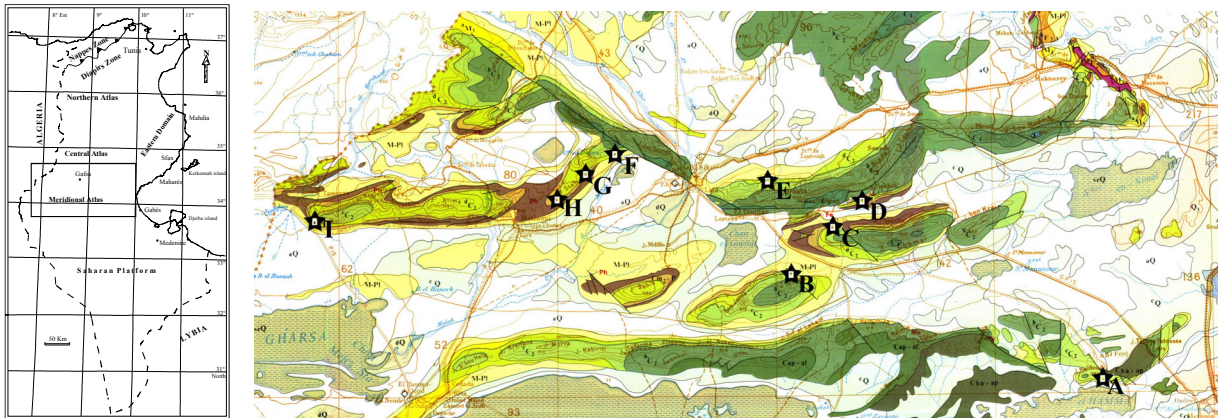
A: J. Aïdoudi (AYD1) (AYD2)	**E:** J. Orbata (KB)	**I:** Oum Kckeb (OMK1)
B: J. Berda (BRD1)	**F:** J. Sehib(SHB1)	**J:** Kef Eddour (KFED1)
C: J. Chamsi (CHM1)	**G:** OuedTefal (TFL1)	**K:** Chbika (CHB1)
D: J. Hamadi (HMD1)	**H:** Secteur 100 (SC100)	

Fig. 1   Localisation map of the smectitic clay deposits

Chebika (CHB1), Oum Khechab (OMK1) and Secteur of 100 localities are of Maastrichtian-Paleocene age and belong to El Haria formation. Finally, those collected in Kef El Dour region (KFED1) are of Miocene age and belong to Beglia formation.

LWA materials were treated as follows: small balls (1.2 to 1.5 cm in diameter) prepared from wet clay were air-dried at room temperature for several days, then heated at 150 °C for at least 24 hrs and finally treated at 350 °C for 2 hrs. These successive pretreatments are necessary to avoid explosion of the small balls during the final firing process. The dried balls were heated in different ways.

1-Slow firing process, where the dried balls were treated at 350 °C and continuously heated till 1170 °C.

2-Quick firing or "flash heat" (mostly used process in the production of industrial LWA). The previously dried balls at 350 °C were introduced into the furnace previously heated to the bloating temperature (1170 °C) for 5 min. The fired balls were then quenched in air for many minutes. Used furnace is of type Thermolyne 46200.

3.   METHODS

Mineralogical analysis of the bulk samples was carried out by X-ray diffraction (XRD, with CuKα radiation, Philips X' Pert). Oriented aggregates were treated with ethylene glycol and heated to 500 °C for 2 hours. The chemical composition was determined by ICP-ES after LiBO$_2$ fusion. The loss on ignition was considered as the weight percent difference between sample heated at 100 °C and 1000 °C. A grain size distribution was performed by an X-ray particle size analyser (Sedigraph, Micromeritics 5100). The total organic carbon was estimated by the analyzer "Carmograph Whostoff 8".

Apparent density of the obtained LWAs (expressed in g.cm^{-3}) was measured by using the method of sand dislocation. The mechanical resistance corresponding to the rupture under point compression was expressed in Nm^{-2}, and water absorption was estimated after 72 hours of immersion in distilled water and expressed in weight percentage.

4.   RESULTS AND DISCUSSION

4.1.   CHARACTERISTICS OF RAW MATERIALS

X-ray diffraction indicated that used raw materials have similar clay compositions, with different percentages of individual components: smectite (35 [KFED1] - 81 %), kaolinite (0 - 51 %) and illite (0 - 13 %). The non-clay minerals are represented by quartz, calcite, dolomite and feldspars (Table 1). Detected high content of fluxing elements is in the range of 11.42 % [KFED1] to 28.49 % being mainly represented by CaO (0.21 % to 20.18 % [SHB1]) and Fe$_2$O$_3$ (4.31 % [SHB1] to 7.72 %) depending on the origin of samples. The SiO$_2$ and Al$_2$O$_3$ contents were also estimated with amounts in the range of 32.1 % [SHB1] to 53.29 % [KFED1] or 11.7 % [SHB1] to 21.01 %, respectively (Table 2). Organic components in the clays were detected only as traces (0.21 % - 0.71 %) (Table 3). As noted a reasonable loss on ignition was an important factor for these clays (13.1 % to 26.5 %. On the basis of the grain size analysis, we have confirmed that our raw materials were extremely fine with an important portion of clay fraction (below 2 μm) of 58 % to 93 % (Fig. 2).

4.2.   FIRING PROCEDURE

As noticed, the dried balls exploded during their placement into the furnace already heated to 1170 °C

Table 1 Mineralogical composition of the studied clays (%) (S = smectite, I = illite, K = kaolinite, I/S = interstratified illite-smectite, Q = quartz, Ca = calcite, F = feldspars, D = dolomite)

Localities	SAMPLES	S	I	I/S	K	Q	F	Ca	D
Aïdoudi	AYD1	74	3	-	9	6	2	6	-
	AYD2	81	5	-	9	4	1	-	-
Berda	BRD1	66	8	-	9	6	2	6	3
Stah (O.Tfal)	TFL1	74	5	-	-	5	-	13	3
Hamadi	HMD1	77	1	-	2	4	-	13	3
Orbata	KB	55	13	-	14	6	2	10	-
Chbika	CHB1	64	6	-	16	5	2	5	2
Sehib	SHB1	44	-	-	5	3	tr	43	5
Chamsi	CHM1	67	-	-	14	5	1	12	1
Oum Khechab	OMK1	42	4	9	38	5	1	1	Tr
Secteur 100	SC100	40	-	-	51	3	1	1	4
Kef el Dour	KFED1	35	6	-	27	25	7	-	-

Table 2 Chemical compositions of the studied clays (%)

Samples	SiO_2	Al_2O_3	Fe_2O_3	MgO	CaO	Na_2O	K_2O	TiO_2	P_2O_5	MnO	Cr_2O_3	LOI	Total	Fluxing	Si/F[a]
AYD1	47.74	18.59	7.39	1.98	3.85	1.94	1.58	0.92	0.27	0.03	0.026	15.3	99.62	16.74	2.85
AYD2	51.35	19.15	7.72	2.19	0.57	1.72	1.41	0.94	0.27	0.02	0.021	14.3	99.66	13.61	3.77
BRD1	46.14	16.92	7.14	2.42	6.77	0.71	1.51	0.82	0.37	0.02	0.024	16.9	99.74	18.55	2.49
TFL1	41.33	11.75	4.6	3.08	12.13	2.24	1.16	0.6	0.44	0.02	0.016	22.4	99.77	23.21	1.78
HMD1	43.99	12.18	5.15	2.73	12.04	1.28	0.93	0.59	0.52	0.02	0.02	20.2	99.65	22.13	1.99
KB	50.92	16.29	6.46	2.27	7.62	0.96	1.29	0.85	0.40	0.02	-	13.0	100.08	18.6	2.74
CHB1	44.68	16.9	6.73	3.19	6.47	0.98	1.77	0.86	0.45	0.02	0.021	17.6	99.67	19.14	2.33
SHB1	32.1	11.7	4.31	2.97	20.18	0.3	0.73	0.53	0.7	0.02	0.036	26.5	100.08	28.49	1.13
CHM1	41.65	16.08	5.8	1.78	10.55	1.82	1.46	0.82	0.18	0.01	0.015	19.5	99.66	21.41	1.94
OMK1	49.1	21.01	7.08	1.99	0.44	2.29	1.89	0.89	0.31	0.01	0.022	14.6	99.63	13.69	3.59
SC100	46.25	20.76	6.9	2.93	2.67	1.16	1.3	0.91	0.28	0.02	0.025	16.7	99.90	14.96	3.09
KFED1	53.29	20.72	5.69	2.45	0.21	1.24	1.83	1.04	0.13	0.02	0.015	13.1	99.73	11.42	4.67

[a] SiO_2/ fluxing (Fe_2O_3, CaO, MgO, Na_2O and K_2O)

Table 3 Content of the Total Organic Carbon of the studied clays (%)

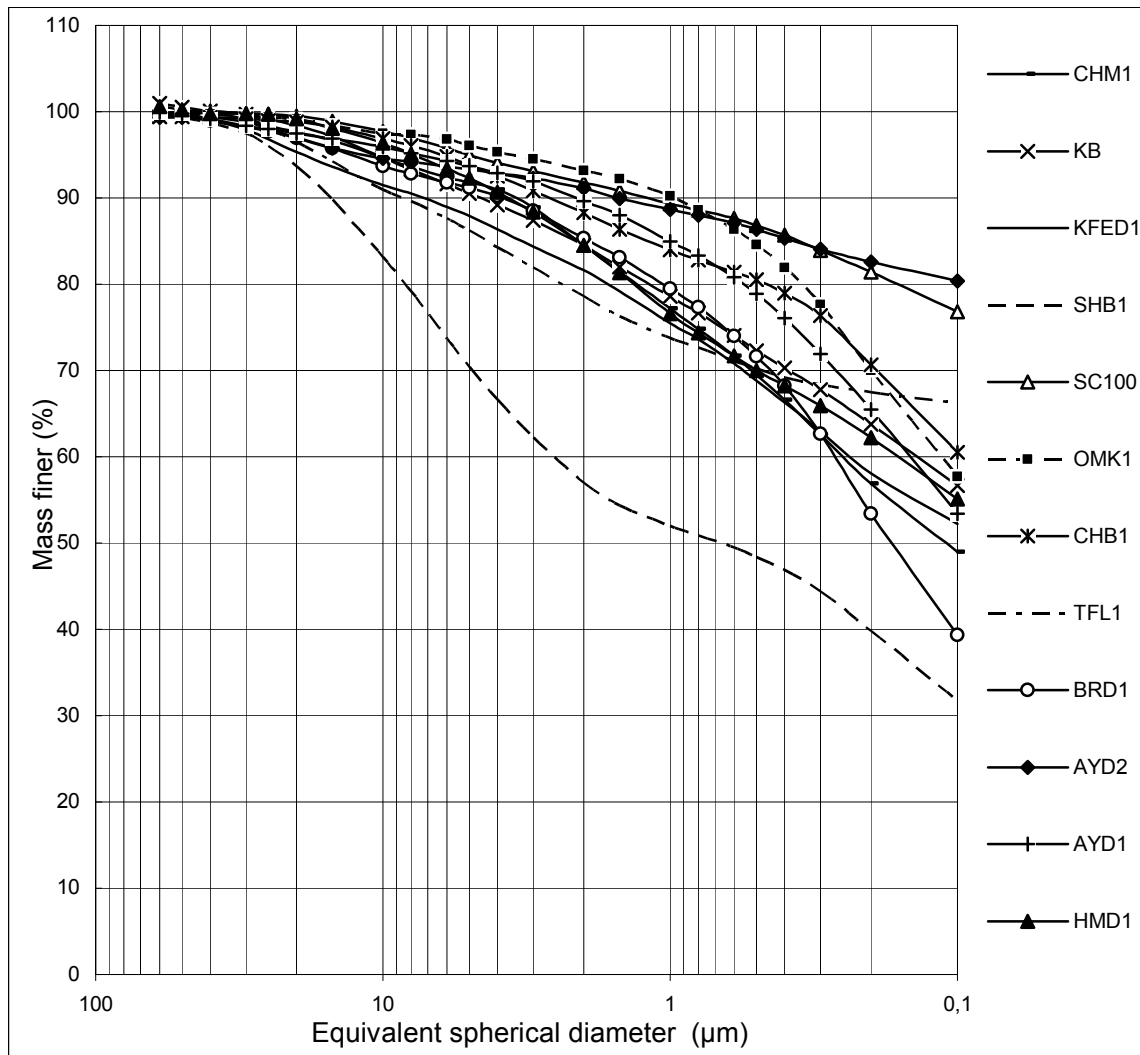| Samples	AYD1	AYD2	BRD1	TFL1	HMD1	KB	CHB1	SHB1	CHM1	OMK1	SC100	KFED1
TOC	0.21	0.30	0.68	0.26	0.71	0.43	0.40	0.54	0.21	0.21	0.40	0.52

Fig. 2   Grain size distribution curves of the studied clays

in case of all the starting clays, except SHB1, where splitting up were observed. To avoid the explosion of balls, numerous pretreatments at different temperatures before firing were envisaged and tested, up to 600 °C for half an hour. Additional drying at 350 °C for 2 hours allowed the expansion of clays, and enhanced the quality of the fired balls from OMK1 and KB clays. Meanwhile, no improvement was observed for the other clays, with an explosion and/or fusion of the obtained balls.

On the other hand, the slow firing was an alternative way to avoid explosion, however, it affected the bloating phenomena. Indeed, the obtained fired balls were not exploded, however, they had less swollen volume compared to those being fired quickly, and sometimes, other balls shrinkage in the volume was also observed for. The best result was obtained from AYD1 clay, which proved a good shape of the bloated balls and favorable swollen properties.

Dahab and Champetier (1984) reported that the quick firing is advantageous due to complete releasing of gases during the softening of the clay in the pyroplastic state. Meanwhile, during the slow firing process, the reactions involved in the release of gases were not completed before the clay vitrification (Riley, 1951) and swelling phenomena occurred after the temperature of vitrification, when the gases were completely released.

The explosion process is not well understood. It could be assigned to the presence of carbonate in high amounts (Riley, 1951; Cougny, 1990) or to the presence of some components which are completely dehydrating at high temperatures (Blank, 1976). However, in our case, the explosion appears to be related to the particle size of the raw materials. An extreme fineness of the starting clays (Fig. 2) did not allow easily the complete drying of the balls preventing a smooth release of the gases at high temperatures.
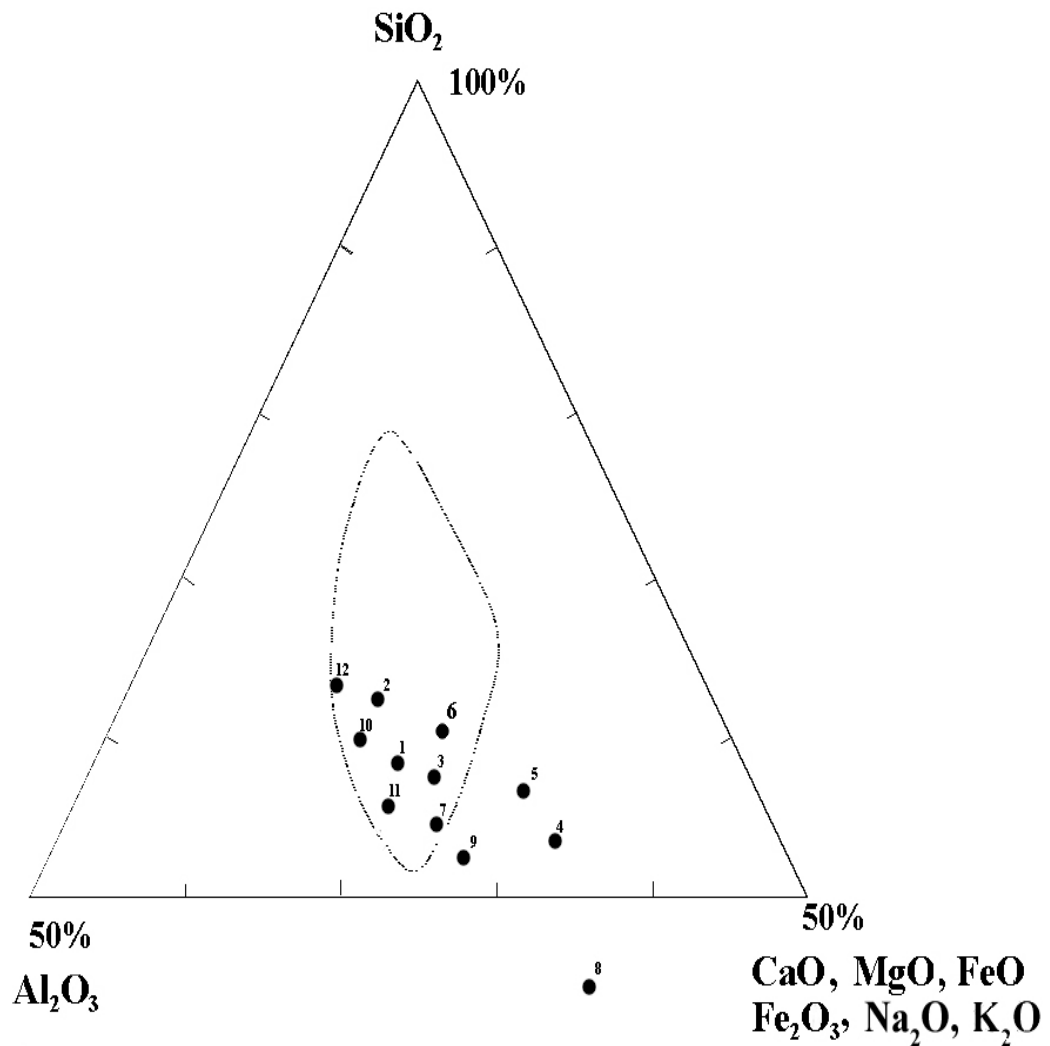
1:	AYD1	5:	HMD1	9:	CHM1
2:	AYD2	6:	KB	10:	OMK1
3:	BRD1	7:	CHB1	11:	SC100
4:	TFL1	8:	SHB1	12:	KFED1

Fig. 3   Ternary diagram showing the area of bloating after Riley (1951)

An organic matter of these clays is low, presented in the range of 0.21 % to 0.71 % (Table 3). The pretreatment at 350 °C is supposed to burn off a largest part of the organic matter. Therefore, the gases do not release during the bloating of the clays at higher temperature intervals. Organic matter appears to have no participated effect on bloating.

### 4.3. BLOATING OF THE RAW MATERIALS

It was well established that the swelling of fired balls occurs when the used materials produce high-temperature highly viscous glassy phases being able to trap gases. The used materials could also contain some minerals, which dissociate and release gases

during the melting process (Conley et al., 1948; Riley, 1951).

The used materials could be placed in the SiO_2/Al_2O_3/fluxing elements ternary diagram (Riley, 1951). Two groups of clays emerged, one within and one outside the area of bloating (Fig. 3). Those within the bloating area are the clays exhibiting favorable chemical composition to produce a suitable viscosity at the bloating temperature (Riley, 1951). According to the chemical analyses (Table 2) and mineralogical ones (Table 1), the gases evolved during the bloating, can be derived from the fluxes (calcite, dolomite, and iron derivatives) or from structural water of clay minerals and an organic matter. But the percentage of

Table 4 Technological properties of the obtained light weight aggregates

Samples	Apparent density (g.cm^{-3})	Point compression (Nm^{-2})	Water absorption (wt %)
AYD1	0.75	275	1.7
OMK1	0.82	263	7.7
KB	0.61	246	3.8
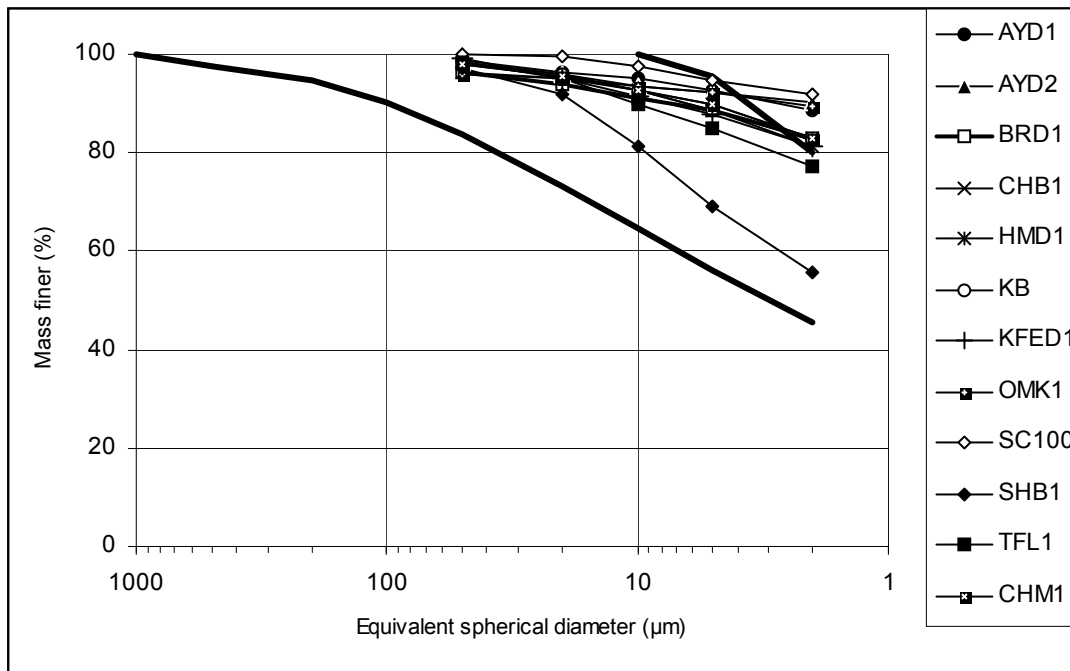
Fig. 4   Grain size spindle favourable for bloating. Thick solid lines indicate the limits of the spindle

organic matter is very low (0.21 % to 0.71 %) and it can be excluded (Table 3).

Among our clays presented within the bloating area, the intact swollen balls were obtained from KB and OMK1 using quick firing process and from AYD1 by slow firing. However, the others produced fractured or partially bloated balls and sometimes no bloating features were observed.

The clays lying outside the area of bloating are abundant in fluxing elements (21 to 28 %) and concurrently they have the lowest Si/fluxing figures 1.13 to 1.99 (Table 2). Consequently, a low viscosity of the liquid phase was unable to entrap significant amounts of gas and thus to bloat during firing (de Gennaro et al., 2004). The samples HMD1 and TFL1 slugged down into a "blob" due to their high contents of CaO (Riley, 1951; Dahab, 1980) which is known to be strong flux (Decleer and Viaene, 1993). When submitted to quick firing the balls prepared from SHB1 shrank while balls prepared from CHM1 exploded to small chips.

Beside the mineralogical and the chemical compositions, the particle size played an important role and a compromise between the drying process and retention of gas during the melting. Figure 4 presents the grain size spindle, and it will be used as a reference to classify our materials with good bloating properties (Cougny, 1990). Almost all of our starting clays (which exploded during quick firing) are situated in the upper part of this spindle and have the fine fraction (<2 μm) outside the plotted limits, with an exception for SHB1 material which is situated in the lower part of this spindle. It did not explode with quick firing due to its coarser grain size, which permitted the most complete drying of the sample before quick firing.

4.4. PROPERTIES OF THE FIRED MATERIALS

The brown selected aggregates prepared from OMK1, KB and AYD1 will be discussed. Those produced by quick firing (OMK1 and KB) had more regular and rounded shape than those produced by slow firing from AYD1. The latter were bright in surface due to the formation of a glassy film with few pores; the inner zone was inhomogeneous with large void in the centre of the balls (Fig. 5). However, the

Fig. 5   Internal structure of the aggregates prepared with AYD1

Fig. 6   Internal structure of the aggregates prepared with KB

LWA prepared from KB clay exhibited an internal structure similar to the industrial products with small irregular and homogeneously distributed pores (Fig. 6). This porosity affects directly the density and the strength of the expanded aggregates, and it is controlled by different parameters, such as the melt formation (amount and viscosity), gas expansion reaction on the glassy phase and finally on the neo-formation of minerals (especially mullite which is known for its high strength) (Decleer and Viaene, 1993).

Table 4 summarizes the data of density, mechanical resistance and water absorption of the prepared aggregates. The values are significantly appreciable. The apparent density is lower than 0.9 g.cm^{-3}, independent on the clays used, with point compression between 246 and 275 Nm^{-2}. The water absorption varied between 1.7 and 7.7 %. These results confirmed the suitability of these clays to be used in the production of lightweight aggregates.

5. CONCLUSIONS

Obtained data have indicated that some of Tunisian smectitic clays with specific characteristics are potentially favorable candidates for the production of lightweight aggregates with good physical properties. However, in the composition relatively much kaolinite (or partly illite) in addition to smectite seems to be favorable. The quick firing appears to be the most suitable process. However, the drying of these smectitic clays is the most crucial step.

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