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RAW MATERIALS FOR THE HEAVY-CLAY INDUSTRY IN EMILIA-ROMAGNA AND MARCHE (CENTRAL-NORTHERN ITALY)

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Abstract: Within a research program concerning raw materials and products of the Italian heavy-clay industry, 75 clays used by producers in the regions of Emilia-Romagna and Marche (central-northern Italy) were characterized by chemical, mineralogical and grain size analyses to relate them to their utilization in the various brick and tile productions. From a geological point of view, the clays considered are mainly belonging to Quaternary alluvial deposits and to Pliocene - Pleistocene marine sediments. Raw materials show distinct chemical and mineralogical compositions, with the most significant variations due to the content of carbonates. As regards the grain size distribution, all the samples can be classified as silty clays or clayey silts. Moving on to the technological field. Roofing tiles and paving bricks are produced through complex bodies which are mainly constituted of non-carbonatic clays, while carbonate-free clays are never employed for the hollow products. As for soft-mud bricks, grain size distribution of the clays is the most important feature.

Key words: ceramics, heavy-clay products, raw materials, heavy-clay industry.

Introduction

A research programme concerning the raw materials used by the Italian brickmaking industry is getting on; it is focused on the compositional characterization of the clays in order to point out the parameters which are conditioning the behaviour of the raw materials and their suitability to a certain production.

In the ambit of this project, the present paper deals with the clays utilized in the plants of the regions of Emilia-Romagna and Marche (central-northern Italy). On the whole 56 factories are operating here, distributed quite uniformly on the territory (41 in Emilia-Romagna and 15 in the Marche). The production is characterized by a strong diversification regarding both the typology of the bricks and the technological process. All the types of heavy-clay products are represented, firstly masonry bri cks and blocks, then hollow materials, roofing tiles and "cotto" paving bricks. As it regards the processing, it can be pointed out that up-to-date (equipped with fast dryers, tunnel kilns, etc.) and conventional plants (hand shaping, Hoffmann kilns and so on) have more or less the same frequency.

Material and methods

Forty-eight brickmaking plants were taken into account, more precisely 36 in Emilia-Romagna and 12 in the Marche; in each factory the clays utilized were sampled (75 on the whole) together with the more representative heavy-clay products (totally 70 samples). Furthermore, the most important remarks about the technological process were collected, along with geological information regarding the raw materials.

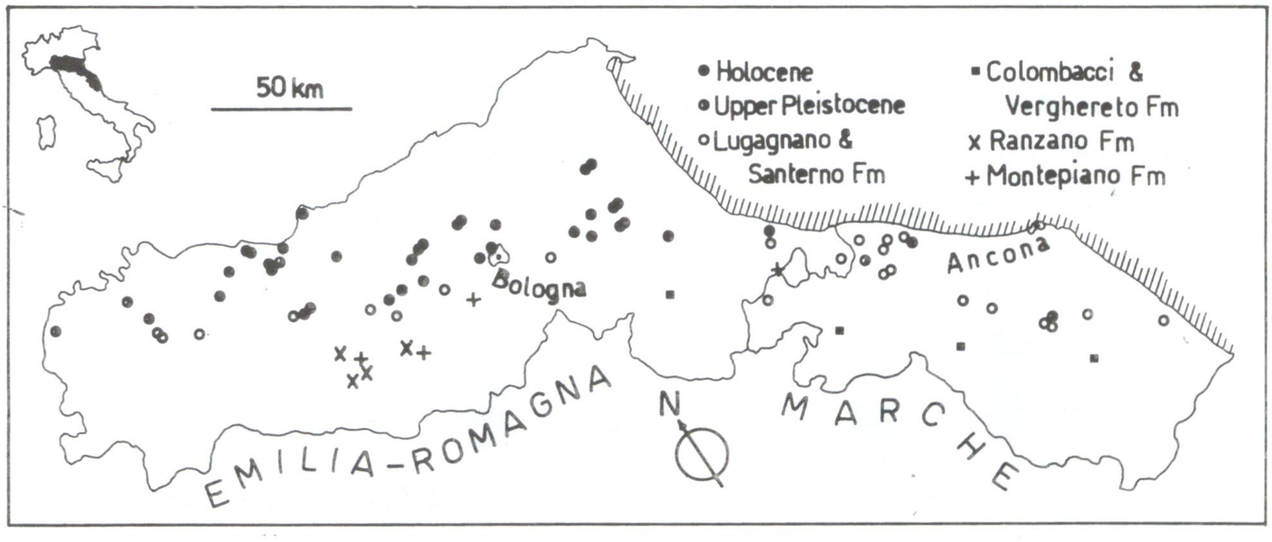

Fig. 1. Location of the sampling sites in Emilia-Romagna and Marche regions, with the indication of the geological unit the samples belong to.

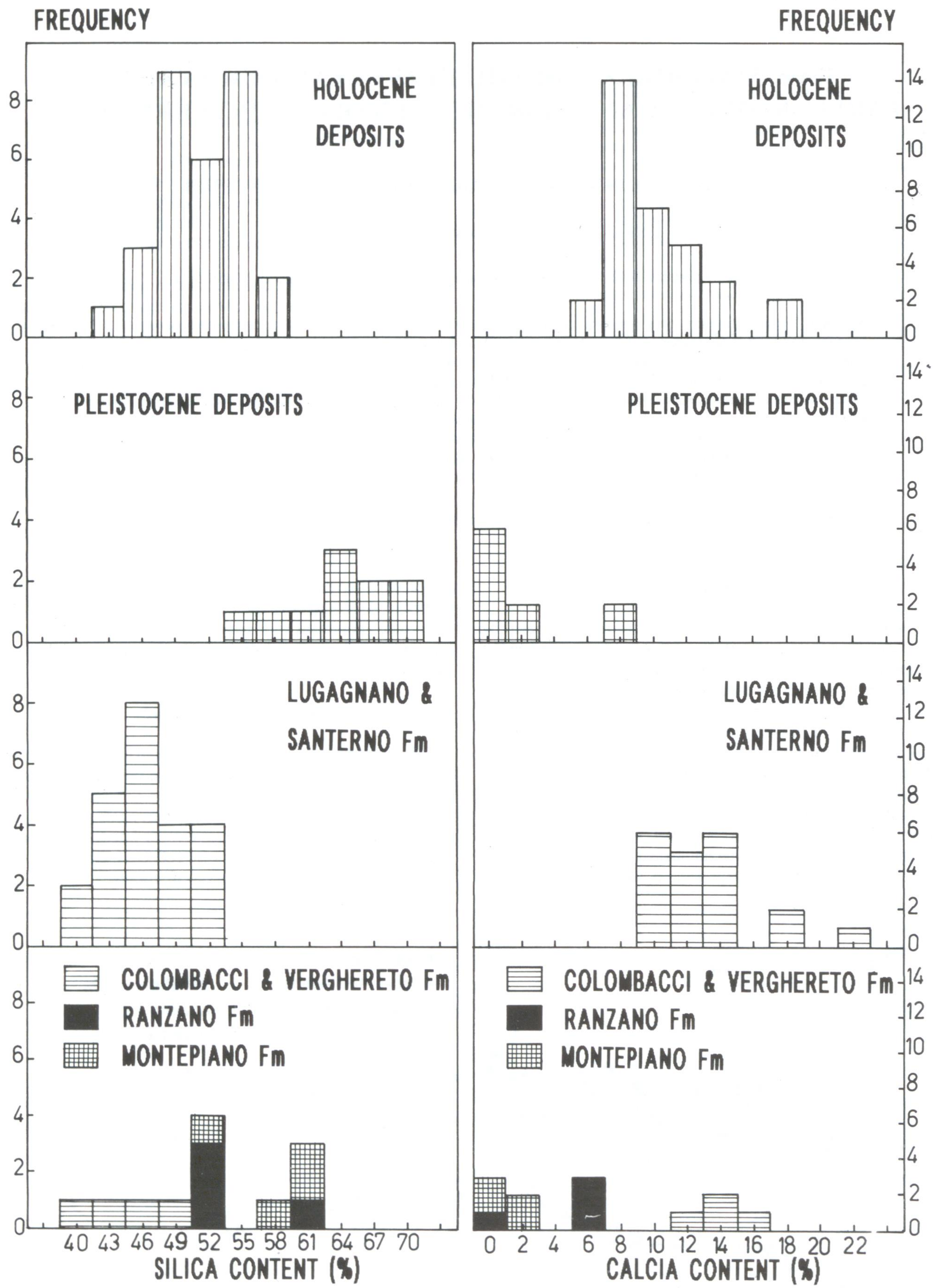

Fig. 2. Frequency histograms for CaO and SiO_2 concentrations relative to the various geological units.

The clays were analyzed by the chemical, mineralogical and grain size point of view. Chemical analyses were performed by means of a X-ray fluorescence spectrometer (Philips PW1480) using pressed powder pellets. Qualitative mineralogical investigations were carried out by means of a X-ray diffractometer (Rigaku Geigerflex with CuKα radiation) on random powder samples; quantitative mineral composition was then calculated through rational computing methods (Fabbri et al. 1986). The grain size distribution was determined by means of a Micromeritics Sedigraph 5000ET apparatus and wet sieving for the coarser fraction.

According to Dondi et al. (1982), the ceramic clays studied (Fig. 1) pertain to 6 main stratigraphical units, which were sampled according to their industrial exploitation. Samples and geological features are:
a - Holocene incoherent deposits of fluvial-deltaic environment, 30 samples; *b* - Upper Pleistocene alluvial deposits, 10 samples; *c* - Lower Pliocene to Lower Pleistocene marly sediments (Lugagnano and Santerno Formations), 23 samples; *d* - Upper Miocene pelites of the Colombacci and the Verghereto Formations, 4 samples; *e* - Lower Oligocene to Lower Miocene pelitic member of the Ranzano Fm., 4 samples; *f* - Middle Eocene - Lower Oligocene red clays (Montepiano Fm.) , 4 samples.

Units from (c) to (f) are essentially deep-sea pelites with a variable carbonate content, while (a) and (b) deposits are continental.

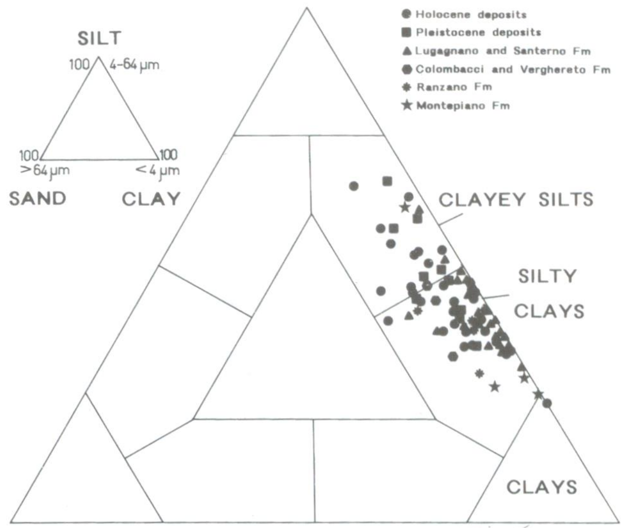

Fig. 3. Granulometric classification of the samples in the Shepard's diagram.

Table 1: Average chemical and mineralogical composition of the clays used by the brickworks in Emilia-Romagna and Marche regions.

	Holocene (n=30)		Pleistocene (n=10)		Lugagnano (n=23)		Colombacci (n=4)		Ranzano (n=4)		Montepiano (n=4)	
	X	s	X	s	X	s	X	s	X	s	X	s
SiO_2	53.30	3.55	65.78	5.12	48.11	3.69	46.02	4.07	56.44	4.73	60.05	4.27
Al_2O_3	12.02	1.04	13.67	2.03	11.54	1.09	11.73	0.80	13.75	0.50	16.38	1.89
TiO_2	0.62	0.05	0.69	0.10	0.62	0.05	0.59	0.09	0.66	0.01	0.71	0.03
Fe_2O_3	4.71	0.51	5.01	0.62	4.53	0.50	4.34	0.62	5.59	0.29	6.05	0.29
MnO	0.12	0.01	0.15	0.06	0.10	0.01	0.11	0.01	0.11	0.03	0.18	0.02
MgO	2.72	0.47	1.80	0.41	2.78	0.31	2.89	0.74	3.63	0.58	2.88	0.52
CaO	10.55	2.43	2.86	3.18	14.10	3.24	15.39	1.64	5.70	2.81	1.89	1.55
Na_2O	1.24	0.31	1.02	0.32	1.11	0.24	1.09	0.14	1.46	0.28	1.24	0.35
K_2O	2.24	0.26	2.00	0.30	2.23	0.30	2.06	0.12	2.75	0.12	3.16	0.55
P_2O_5	0.14	0.02	0.10	0.04	0.15	0.01	0.14	0.01	0.11	0.00	0.12	0.02
L.O.I.	12.97	1.75	6.99	2.73	15.25	2.33	16.33	1.77	9.40	2.46	7.31	2.19
Quartz	31	3.6	42	5.4	27	2.5	25	3.2	30	5.8	34	6.0
Feldspars	10	3.0	9	6.2	9	2.2	9	1.4	13	2.2	11	2.6
Calcite	17	4.3	traces*		25	5.2	25	2.2	9	4.8	traces	
Dolomite	traces		traces		traces		6	5.2	traces		5**	
Illite	22	2.5	22	3.5	21	2.1	22	2.2	25	1.7	31	3.8
Chlorite	8	2.3	6	2.4	8	2.1	6	2.6	10	3.8	6	2.2
Kaolinite	traces		traces		traces		traces		4	1.7	5	5.0
Smectite	4	3.4	12	4.9	traces		traces		traces		traces	
Fe-oxides	3	0.5	4	0.7	3	0.4	3	0.5	4	0.0	5	1.0
Accessories	5	3.0	5	4.2	7	3.0	4	1.2	5	3.6	3	1.3

Legend: * with the exception of 2 samples which contain about 15% calcite; ** 2 samples contain 10% dolomite and the others 2 no dolomite. L.O.I. - Loss on ignition; X - average concentration; S - standard deviation.

Results

The chemical and mineralogical data are summarized in Tab. 1, where significant statistical parameters for each group of samples are reported. Frequency histograms for CaO and SiO_2 concentrations are shown in Fig. 2, and grain size distributions are schematically evidenced by the Shepard's diagram (Fig. 3).

a - Holocene incoherent alluvial deposits

All the samples included in this geological unit come from Emilia-Romagna. From a chemical point of view, the most conspicuous characteristic seems to be the variation of the CaO content, about in the range 8 - 18%, even if the main part of the data is comprised between 8 and 14%. These variations correspond to analogous fluctuations of the carbonate contents, which are mainly represented by calcite; most of the samples containing significant amounts of dolomite come from Romagna area. According to the carbonate contents the Holocene raw materials are classified as marly clays or clayey marls.

Silica concentrations range from 48 to 60%, except one sample with about 45% SiO_2, and they are inversely related to calcium contents: the higher the carbonates, the lower the quartz and the silicates. Among the latter phases quartz prevails on plagioclase, while the phyllosilicates are principally constituted by illite, chlorite and smectite (Tab. 1).

In agreement with the Shepard's classification, the Holocene materials can be classified as silty clays and clayey silts, the former being more abundant.

b - Upper Pleistocene alluvial deposits

These raw materials are used in Emilia (7 samples) and Marche (3 samples), but no difference can be emphasized between the two areas of provenance. They generally show a low content of carbonates and high concentrations of silica, which vary in a wide range from about 55% to about 73% SiO_2, due to high percentages of quartz. Among the phyllosilicates it can be stressed the significant amount of smectite, which is the second in order of abundance after illite (Tab. 1). Upper Pleistocene sediments can be classified as clays or marly clays by considering the carbonate content, or as silty clays or clayey silts according to the Shepard's diagram.

c - Lugagnano and Santerno Formations

Plio-Pleistocene sediments are largely used by brickworks both in Emilia-Romagna (10 samples) and in the Marche (13 samples). These raw materials are characterized by rather narrow ranges of variation of the compositional and granulometric parameters, with the exception of calcium concentrations, and of carbonate minerals as a consequence. As a matter of fact, there are 4 samples coming from the Marche which contain more than 18% CaO and 36% calcite plus dolomite, instead of an average content of about 13% and 24% respectively. The main group of samples is represented by marly clays and clayey marls, while the four Ca-rich samples are marls. The silicate fraction is constituted by quartz and illite prevailing on plagioclase and chlorite (Tab. 1). All the Plio-Pleistocene materials are silty clays with reference to the grain size distribution.

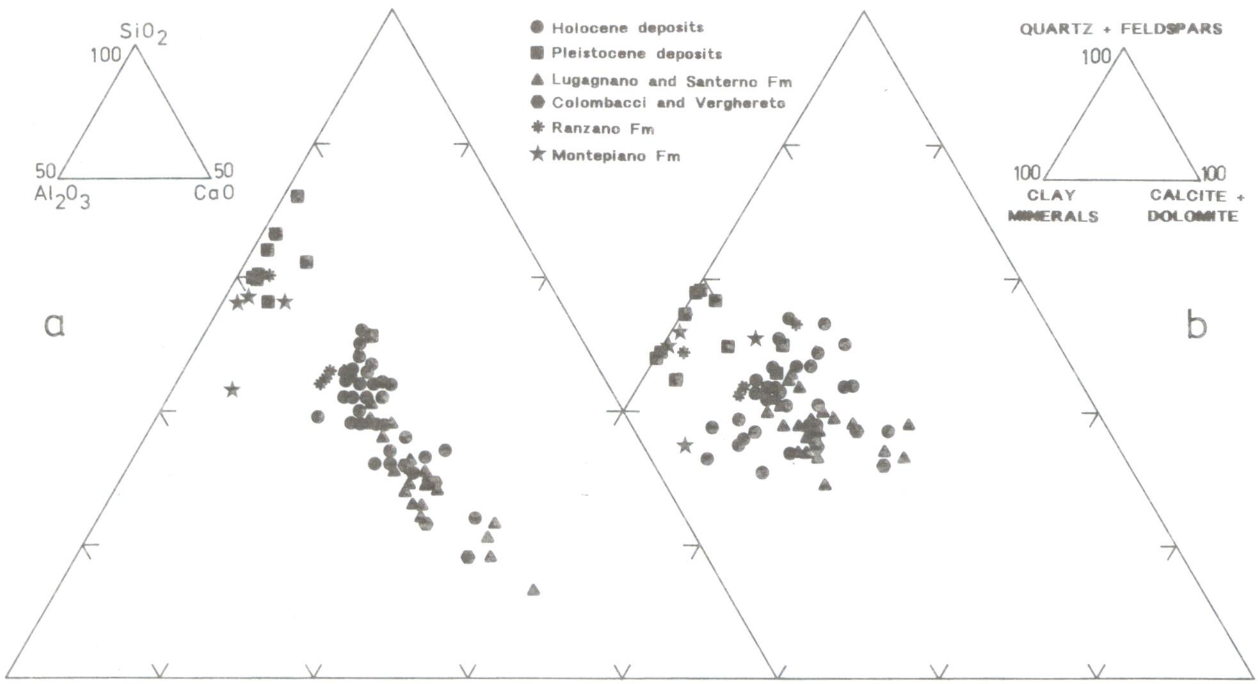

Fig. 4. Representation of the clays in the ternary diagrams:
a - SiO_2-Al_2O_3-CaO and **b** - clay minerals-carbonates-quartz plus feldspars.

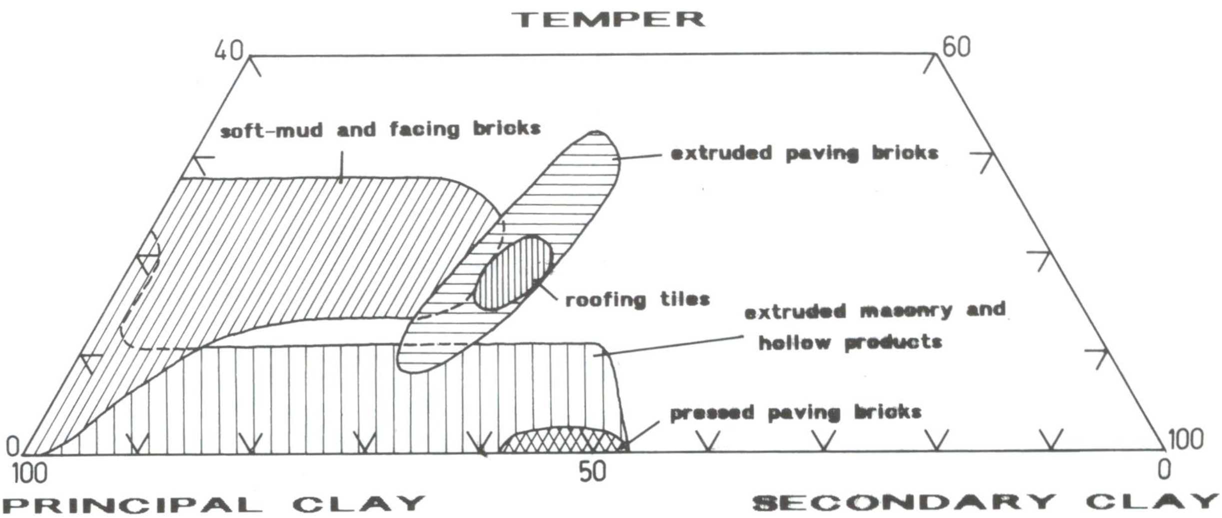

Fig. 5. Schematic representation of the combination among principal clay, secondary clay and temper in the bodies for the different heavy-clay products.

d - Colombacci and Verghereto Formations

Messinian raw materials have compositions consistent with those of marly sediments. About the chemical data, silica values range from 41 to 51%, alumina and calcia in the intervals 11-13% and 13-18% respectively. Mineralogy points out high contents both in calcite and dolomite as well as relatively low concentrations in quartz and feldspars (essentially plagioclase). In the <4 μm fraction, illite is predominant over chlorite, while kaolinite and smectite are present in traces. Grain size characteristics correspond to silty clays in the Shepard's sketch.

e-f - Ranzano and Montepiano Formations

The pelitic member of Ranzano Formation and the red clays of Montepiano Formation are well known in the Italian ceramic tile industry, because they have been largely used in the production of the classic "cottoforte" and red stoneware tiles respectively in the district of Sassuolo. The carbonate content of these raw materials is always quite low, but calcite is typical of Ranzano, and dolomite of Montepiano; silica and alumina values are high and quite constant. Illite is usually very abundant, while chlorite is more concentrated in the Ranzano Fm.; smectite is always negligible (Tab. 1). From the grain size viewpoint, the samples of both the Formations are classifiable as silty clays.

In summary all the variations of the three principal chemical oxides (SiO_2, Al_2O_3 and CaO) are well evidenced in the ternary diagram reported in Fig. 4a. It is clear that the main variation range is relative to calcium concentrations, so that the points in the diagram individuate a line along which the SiO_2/Al_2O_3 ratio is constant and equal to about 4.5. The points representative of the clays belonging to Eocene and Pleistocene Formations are located near the SiO_2-Al_2O_3 side; Pliocene and Upper Miocene samples are the richest in calcium.

The indications above are well confirmed by the ternary diagram based on the mineralogical composition, arranged by grouping the phases as follows: clay minerals, carbonates, quartz plus feldspars (Fig. 4b). Furthermore, an average ratio close to 1 can be deduced between clay minerals and quartz plus feldspars, while the carbonate content can vary up to 40%. These observations are in good agreement with what previously reported in the literature about both Italian and foreign clays for the manufacture of heavy-clay products (Fiori et al. 1974; Lach 1979).

From a granulometric point of view, most of the samples are silty clays, and they are suitable for the production of hollow blocks (perforation > 40 %) according to Winkler's diagram.

Discussion

As regards the use of the clays examined, some interesting observations can be made considering Tab. 2, which simultaneously takes into consideration the number of clays and the presence of temper in the body on one hand, and the type of products obtained on the other. The body formulation for each type of products can be schematized by a ternary diagram, whose vertexes represent the amount of two clay raw materials and temper respectively (Fig. 5). The more abundant clay is named "principal clay" and the less abundant, if present, is named "secondary clay".

Paving bricks and roofing tiles

Neglecting pressed paving bricks, for which there are few samples, it is worth searching for specific compositional ranges of the clays for each kind of products.

The extruded paving bricks are made of bodies which consist of two clays, with a ratio from 1:1 to 2:1, besides temper (Fig. 5); the principal component is a non-calcareous iron-rich clay, while the other one contain up to 20% of carbonates: the higher the carbonate content, the lower the proportion of this latter raw material.

The bodies for roofing tiles look like those for paving bricks, as far as their formulation is regarded. In both cases a non-carbonatic clay is mixed with a carbonate-bearing one, and the two clays are different also as far as the concentrations of aluminium and iron are concerned, being these elements more concentrated in the non-carbonatic clay.

Table 2: List of the routes to compose the bodies for different heavy-clay products.

Geological unit**	Number of clays	Temper	Type of products* A	B	C	D	E	F
Holocene	1	no	X	XXXXX	XXX			
		yes	X	XXX	XX			
	2	no		XXXXXX	XXXXX			
		yes	X	X	X	XX		
Pleistocene	1	no	X	X	X			
	2	no		XXX				X
		yes	XX	XX		XX		
Lugagnano and Santerno	1	no		XX	XXX			
		yes	X	XXX	X			
	2	no		XXXX	X			XX
		yes	XXX	XXX	XX	XXX	X	
Colombacci Verghereto	1	no	X	XX		X		
Ranzano	1	yes					X	
	2	yes				X	XX	
Montepiano	2	yes				XX	XXX	

Legend: *letters indicate: A-soft-mud bricks; B-extruded masonry bricks and blocks; C- hollow materials; D-roofing tiles; E-extruded paving bricks; F=pressed paving bricks. The number of "X" represents the frequency of each type of product.
**see text for the description of the geological units.

Soft-mud bricks

The situation regarding soft-mud bricks is more complex, because the bodies can be built up by using either only one clay or a mixture of two clays and temper; in the former type of body, moreover, temper can be present or not. This situation depends on the fundamental needs of these products, which are a suitable colouring and an abundant coarse fraction. As a matter of fact, the clays used without temper contain about 30% of >20 microns fraction, while the other clays only contain about 15% of it; the addition of temper, in a proportion around 20%, makes all the bodies similar with reference to the grain size distribution. In conclusion, it results that the clays, on the whole, present compositional features which are variable in wide concentration ranges and it is impossible to individuate well defined compositions.

Masonry and hollow materials

The chemical and mineralogical composition of the clays for the bodies used to produce extruded masonry and hollow materials does not point out any specific characteristic relative to the different products: common and facing bricks, masonry light-weight blocks, hollow bricks, hollow floor blocks, hollow slabs. This situation resembles that one already seen for the body formulation of the same artifacts (Fig. 5), since the mixture can be made by using one or two clays, with or without temper. Nevertheless, if we analyze the results more in detail, we can observe some features to be evidenced. Carbonate-free clays, for example, are never used in the production of all the hollow products, and those used for slabs and floor blocks show similar compositions which fall in a restricted variation range, with average values for SiO_2, Al_2O_3 and CaO around 53%, 12% and 11 % respectively. The clays used to produce hollow bricks are slightly richer in calcium carbonate, so that the average data for SiO_2, Al_2O_3 and CaO are 47.5%, 11% and 15% respectively, but the silica/alumina ratio is always the same and close to 4.5.

Most of the clays for masonry bricks and blocks lie in the same compositional field for hollow materials, and the silica/alumina ratio is the same too; the remaining clays are practically carbonate-free.

In conclusion, analogously to what concluded by Schmidt-Reinholz (1986) about German raw materials for heavy-clay industry, it can be seen that the clays of each geological unit are used for different products; in particular, the Ranzano and Montepiano clays are exclusively utilized for the production of roofing tiles and extruded paving bricks, always in complex bodies built up of two clays and temper. Another singular case is represented by the production of pressed paving bricks, which are obtained by using a mixture of Pliocene and Pleistocene raw materials, always without temper.

The production of soft-mud bricks is done by utilizing Holocene to Upper Miocene clays; in most cases, the body is obtained by mixing two different clays and temper as well. The situation for extruded masonry and hollow materials (B and C in Tab. 2) is almost the opposite, since you do not generally add temper and the frequency of one or two clays in the body is practically the same (Fig. 5).

Masonry and hollow bricks and blocks are produced by employing different geological types of clays, from the most recent to the Upper Miocene ones.

Conclusion

Emilia-Romagna and Marche are two regions where several brickworks are operating. Most of the products fall within the cathegory including masonry and hollow bricks and blocks; the production of soft-mud bricks is developed too. This industry employs clays of various geological provenance, from Eocene to recent alluvial deposits.

The main common characteristic of the clays is, from a compositional point of view, the SiO_2/Al_2O_3 ratio, around 4.5, while the more consistent variations regard the calcium and carbonate content (up to 22% and 40% respectively).

With reference to the different types of artifacts, the most characteristic clays seem to be the few used to produce roofing tiles and paving bricks. Within the larger family including extruded masonry and hollow bricks and blocks, the most uniform of the clays are those for hollow slabs and hollow floor blocks. As for soft-mud bricks, the most important feature of the corresponding clays is the grain size distribution, which appear to be characterized by an amount of >20 microns fraction as high as 30 %; temper is added if this amount of coarse fraction is not present in the raw clay.

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Our company offers its services for home and foreign clients. In years 1972 - 1990 **Nafta** was involved in drilling projects in Iraq (oil fields Maynoon, North Rumaila and Hamrin) and in 1985 in Sweden for the hydrothermal programm.

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- On-shore oil and gas drilling up to 6 000 m (20 000 feet), completion and well testing jobs included
- Work-over jobs
- Water and geothermal drilling, including well completion
- Injection & production well drilling and completion jobs for gas underground storages

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Gas is injected into depleted beds by six turbocompressor units with a total power of 18 000 kW with a pressusre of 9.0 MPa.

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The main gas reservoirs are located in the Vienna Basin as well as first of all in the East Slovakian Lowlands with natural gasoline gas deposits.

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Nafta plans on improving its carbonhydrate production with enhanced secondary and tertiary stimulation methods in the future.

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