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CHAPTER II  
ARTICLE 1

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# PAPER-CLAY AS A SOLUTION FOR CLAY RECYCLING AND REPAIRING

## ABSTRACT

One of the most remarkable problems with clay and ceramics is, once cooked, the incapability of being repaired in case of cracking, and linked to this problem, the recycling difficulty. Proceeding in the different stages of the manufacturing process, the problem presents more and more complex aspects, as not only the causes that generate it are more articulated, but also the process increasingly turns towards a condition of irreversibility. Moreover, the declination of the ceramic product (from an object of use, of an industrial nature, to an artistic object, sometimes of great value) is such that any damage affects, at various levels, the characteristics of the product: from those technological, until the economic value of the object is completely annulled. The only repairable options have so far been made possible by the use of resins, in particular two-component ones, which, in addition to not being biocompatible, are not even homogeneous with the ceramic material. Recently, under the pretext of a recycling hypothesis of a material with a prevalent composition of calcium carbonate, the old and neglected technique of paper-clay, namely a composite material of cellulose and clay, has offered some initial results, as well as some experiments on its components are highlighting some options of reparability and recycling for the ceramic product.

## KEYWORDS

Paper-clay, clay repairing, clay recycle.

## RESUMO

Um dos problemas mais marcantes do barro e da cerâmica é, depois de cozido, a impossibilidade de ser reparado em caso de fissuração, e ligada a este problema, a dificuldade de reciclagem. Prosseguindo nas diferentes fases do processo de fabrico, o problema apresenta aspectos cada vez mais complexos, pois não só as causas que o geram estão mais articuladas, como também o processo se volta cada vez mais para uma condição de irreversibilidade. Além disso, a decadência do produto cerâmico (de objecto de uso, de carácter industrial, para objecto artístico, por vezes de grande valor) é tal que qualquer dano afecta, a vários níveis, as características do produto: desde os tecnológicos, até que o valor económico do objeto seja totalmente anulado. As únicas opções reparáveis até agora têm sido possibilitadas pela utilização de resinas, em especial as bicomponentes, que, além de não serem biocompatíveis, nem sequer são homogêneas com o material cerâmico. Recentemente, a pretexto de uma hipótese de reciclagem de um material com uma composição predominante de carbonato de cálcio, a antiga e negligenciada técnica do papel-argila, nomeadamente um material compósito de celulose e argila, tem oferecido alguns resultados iniciais, bem como algumas experiências sobre os seus componentes destacam-se algumas opções de reparabilidade e reciclagem para o produto cerâmico.

## PALAVRAS-CHAVE

Papel-argila, reparação de barro, reciclagem de barro.

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# 1. ANALYSIS OF THE PROBLEM

## 1.1. DEFECTS IN CERAMIC PRODUCTION

As is certainly known, the complete ceramic process, referring to the most common one, consists of an air-drying phase, a first firing, glazing and second firing. This process, depending on the countless variations that can be made (materials, number of firings, temperatures, colors), can be intervened with a wide variety of product differentiation, according to technological, use, but also aesthetic characteristics. It is not at all unusual, however, that during some of the different phases of the clay production process for ceramic production, there may be damage, which essentially ranges from the injury, more or less accentuated, to the explosion of the piece.

The causes of these unfortunate events are manifold. Very often they are causes that originate from the moulding phase of the piece: differences and inconsistencies of thickness, air bubbles, impurities in the material (the most common of which are micro flakes of calcium sulphate dihydrate, incorporated during kneading of the clay material on the gypsum shelf), fatigue of the material, already recycled several times (in this phase raw, the material is still recyclable). Other times they originate from too fast drying or in presence of drafts. In particularly complex pieces, even a wrong architecture of the piece can give rise to a non-coherent contraction of some of its parts, creating internal tensions to the material, which in the cooking phase are exasperated and, if they reach the limit, give rise to more or less serious injuries, or even can lead to the destruction of the piece.

Some of these causes, in the pre-cooking phase, can be identified, prevented and resolved. Experience certainly plays a fundamental role, but sometimes even the most experienced ceramist must give up in the face of a certain unpredictability in the complexity of physic-chemical phenomena. These processes take place at temperatures close to 1000 °C and are therefore quite uncontrollable. However, before this phase, if some adjustments of the material are difficult, but nevertheless not impossible, as *extrema ratio*, the piece destroyed, not yet cooked, constitutes an amount of material still recyclable. It is only necessary to restart the production cycle with the proverbial patience of the potter.

In the industrial sector, where casting on a mould is predominant, the speed and controllability of the different parameters that contribute to the definition of the workpiece make it possible to minimize any production defects. Putting a raw piece back into production is in fact a simple automatism. The percentage of waste on the final product is extremely limited, many times it is made only on the basis of aesthetic defects, which sometimes also allow the sale at a lower price, in special markets dedicated to defective specimens in production. Finally, only for a small percentage, which is not part of the first cases, remains the problem of disposal as non-recyclable waste.

Part of this material is used in the field of aggregates: from draining soil for road surfaces or for the architecture of gardens, to the so-called "*chamotte*", terracotta in small-grained scales, to be mixed to other semi-refractory ceramic materials.

## 1.2. THE PROBLEM OF CALCIUM CARBONATE RECYCLING

The project PNRR (National Program of Recovery and Resilience), with its vast investment in scientific research, has seen the Polytechnic University of Bari as one of the significant partners, with several research operational units. In particular, one of the related units concerned the problem of the disposal of sawing sludge from stone material, the processing of which is one of the largest resources of the Apulian territory.

The fundamental problem in reusing this waste material is that of the sustainability and possible circularity of the material itself. The research unit is entitled "MICS - Made in Italy Circular and Sustainable" (see acknowledgements below). These two characteristics are therefore indispensable and obviously the most immediate hypotheses would allow the use of resins or polymers that would act as binders. A research hypothesis has instead glimpsed in the ceramic material a possibility of environmentally friendly solution, tradition and recyclability.

Compared to calcium sulphate, that is gypsum, calcium carbonate does not produce bursts at high temperatures, indeed its melting temperature ( $>825^{\circ}\text{C}$ ) would allow to predict that at the standard ceramic firing temperature ( $980^{\circ}\text{C}$ ) the cohesion between the two materials is guaranteed. During the first experiments, however, the prediction has been contradicted by the fact that during the mixing process with the liquefied- made clay, the so-called "slip" (in Italian, "*barbottina*"), as soon as rehydrated, the carbonate has rearranged in microgranules, visible to the naked eye, even after firing the different samples made with different percentages of carbonate (Fig. 1).

Hence the idea of resorting to additives, able to homogenize the distribution of the compound, avoiding the formation of lumps.

**Fig. 1.** (a, b) Clay mixture with calcium carbonate, in which the granules formed during the union of the two materials are visible; (c) The calcium carbonate granules still visible after firing three samples with different percentages of carbonate.



## 1.3. FROM PROBLEMATIZATION TO PROBLEM SYNTHESIS

In the technical-artisan literature of ceramics [1, 2, 5 & 6], there are numerous solutions that have allowed us to hypothesize the resolution of the listed problems, through a certain empiricism, at least in the initial phase, and subsequently by experimental way. The main solutions, and the analogies studied, on the basis of their use, that allowed their experimentation, have been the following.

**Pehatine.** The problem of preventing lumps is a well-known ceramic problem and solved thanks to a suspensive additive that is pehatine. It is a thick, sticky substance that blends well with powder glaze, and that adheres well

to the surface of the clay. Its usual use with glaze has revealed the possibility of extending its application to calcium carbonate powders.

**Rabbit glue.** Beyond its use to obtain the color "Saint John White", derived from the carbonation of hydrated lime, for prolonged exposure to air and subsequent grinding, calcium carbonate is used, together with animal glue to obtain the white background preparatory for the canvas to paint, the so-called "imprinting". Hence the idea that rabbit glue can act as a binder between calcium carbonate and clay.

**Agar gel.** Organic product of aquatic origin (it is produced by an alga, it is perfectly edible and is even used in the food industry), the agar gel has remarkable properties, as its liquefaction point (from left) does not coincide with its solidification point (from the right). In this part of the process, it tends to expel water and incorporate solid corpuscles. Also in this case, the idea comes from its agglutinating properties.

**Paper-clay.** The insertion of cellulose in the clay dough has always been under-evaluated, in the following chapter we will see the main reasons for this lack of consideration. It was a material that is often used in the field of child arts, for its immediacy, ease of use, good cost/quality ratio. This material also has excellent inclusive capabilities and are the ones for which it has been taken unexpectedly into account for a whole other type of scientific discourse.

## 2. PAPER-CLAY, FEATURES AND APPLICATION

### 2.1. GENERAL CHARACTERISTICS OF PAPER-CLAY

The last of the additives examined in the previous chapter was discussed at length with great interest, opening up a series of separate considerations independent of the research on calcium carbonate which we are now leaving out. Paper-clay is obtained, quite simply, by mixing cellulose gruel with clay and water. The first is already on the market, produced by industries for various uses, in a dry form that requires hydration (Fig. 2). It can also be obtained by softening cellulose paper in water. The recommended percentage does not agree with the different sources that speak of it, sources that actually are almost never scientific [2, 4]. They refer to percentages of weight or volume between cellulose and clay, which is very misleading because, depending on the percentage of water present in the clay, both the volume and the weight may vary. From all sources [3, 6], for example, those obtained are values ranging between 2% and 200% of the volume. For the rest the literature of documentation on paper-clay is really small, which is the main reason for the lack of consideration enjoyed by the material.

For the sake of comfort, from now on we make "paper" and "cellulose" coincide. In the face of a millennial technique, such as ceramics, paper is first and foremost much more recent. And even when it already existed and its use was claimed, the costs of the paper were decidedly high. Surely no one would use it as an excipient of a muddy dough. As a result, it is clear that no use of paper-clay has been consolidated, unlike in the case of ceramics, whose literature is millennial and whose technique has remained substantially unchanged.

In fact, both the pottery wheel and the mould technology have basically remained unchanged over the millennia, with the sole exception of a small technological update. Today we use the electric pottery wheel, at adjustable speed, but in the end until a few decades ago still potter masters from Faenza or Puglia used the pedal-operated pottery wheel.



Fig. 2. A basin containing a paper-clay dough made with 2.5% cellulose paste and white clay.

Regardless of the characteristics of paper-clay, for example how much this material is workable on the pottery wheel, it is perfectly understood that the process of refining the ceramic technique has been very slow and very long. While for obvious reasons in the case of paper-clay, evolutionary history, the exploration of technical and aesthetic possibilities have been very limited in time.

## 2.2. RECIPE BOOK USED IN EXPERIMENTATION

In the first trial six types of dough were obtained, symmetrical, in the sense that the dough are actually three, multiplied by the two types of clay with which they were mixed, the traditional Faenza and white clay. The three samples diverge by percentage of cellulose and in one was added the additive pehatine. The percentages of cellulose in the three Faenza and white clay samples were 1.25%, 2.5% and 5% of dry weight. The cellulose was added in the form of 100% cellulose paper, first macerated, then blended. So raw clay was added, perfectly dry. At this point the amount of water added is indifferent, since it will later evaporate or be expelled onto the gypsum shelf for drying. In the third sample the additive was added as a percentage of 5% by weight. Further samples will be taken in the course of the trials.

As for the other two additives, agar gel and rabbit glue, other samples were created, again with the percentage of 5% cellulose, to ensure the comparability of the result. The main difficulty in this case was in having to create the hot dough, which, however, once cooled, did not show great differences in consistency, perhaps due to the low percentage of the additive.

## 2.3. TECHNOLOGICAL CHARACTERISTICS AND SPECIFICATIONS OF PAPER-CLAY: WELDABILITY

Paper-clay is distinguished at the microscopic level by an alveolar structure that is obtained by sublimation during firing of the microscopic tubes with which the cellulose is presented. This alveolar structure considerably lightens the object, since the air takes its place, but this takes place according to a structural network of matter, so as not to weaken it.

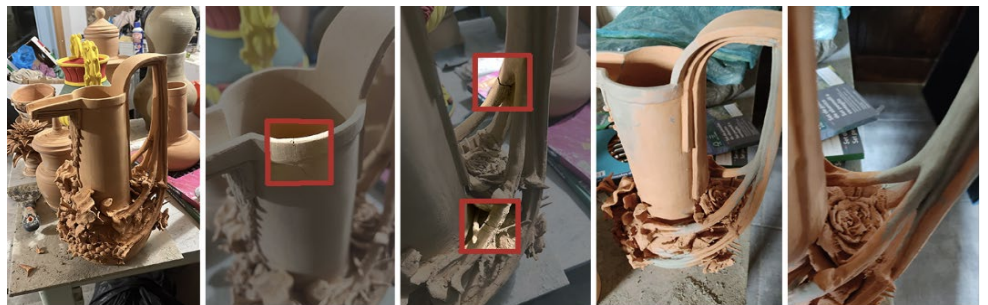
**Raw to raw weldability.** These tubes allow the material still raw, both in paste and in the state of slip (barbottina), to hook well to other material that has already reached the "leather hardness", or even is dry, level of drying with respect to which the clay is not in the state of concreteness of dough, nor that of barbottina, is able to depreciate. The sudden absorption of the water present in the material by the already hard parts, but for this very hygroscopic reason, means that the two materials do not bond together, indeed, in a good casuistry, just the second piece added hardens, comes off. In paper-clay, on the other hand, the hygroscopic tubes of the compound tend to keep the water in possession and the slow return by capillarity, so that the two parts of material can be perfectly welded. This parameter of "weldability" varies with the percentage of cellulose jelly in the compound, according to direct proportional methods. And it appears at first strange, but to a careful reason instead it turns out logical and clear, that the weldability stands out even more if the hard piece is in clay and the addition, or repair, in paper-clay. Precisely because the different hygroscopic parameter between the two surfaces in contact creates a difference in surface tension, such as to make the two materials bond better.

**Raw to fired weldability.** Although the previous point is also an important element in the manufacture of ceramics, the weldability of raw material on the already fired one is the most interesting aspect of the product. If the phenomenon described in the previous point succeeds in taking place thanks to the micro-particles of the material adhering to the boundary surfaces, at this point in the experiment the addition of additional additives has increased the adhesion. Fig. 3 shows the example of repair on the petal of a flower, broken by impact. The two flaps of the fracture coincided perfectly, and the paper-clay poodle was used as glue, that is, without the need to reintegrate missing parts of material. In Fig. 4, instead, the most difficult repair example is reported, as the shortening in cooking caused a fracture with lack of material. In this case we acted with a solid part of filling for the lack of more consistent and paper-clay poodle with additive for bonding to the edges.

**Raw weldability on glaze.** Extensive literature (as we said before, not very scientific) would confirm that the glazing can take place at this point in the process, just where the description arrives, ie by also disposing of a raw part and starting a single firing of the artifact. However, this part has not yet been sufficiently tested, so that we can boast of reliable data report here.



**Fig. 3.** Example of repair, perfectly successful even after glazing step, on a petal of a flower pine cone, accidentally broken.



**Fig. 4.** Example of remarkable fractures obtained in cooking and perfectly welded with paper-clay that integrates the missing part and adheres perfectly to the terracotta.

## 2.4. TECHNOLOGICAL CHARACTERISTICS AND SPECIFICATIONS OF PAPER-CLAY: MACHINABILITY ON THE POTTERY WHEEL

By subjecting the material to machinability at the pottery wheel, the results obtained were very different, depending on the different percentage of cellulose. The tendency has been to obtain better results on the pottery wheel for lower percentages of cellulose, as well as with the white clay, compared to Faenza clay. These data suggest that, although the presence of cellulose increases the mechanical strength of the product, it actually limits its elastic cohesion, a fundamental characteristic during turning, as it is the force that opposes the centrifugal strength of the wheel.

In some simple objects (two bowls, Fig. 5) made on the wheel with two different mixtures, it was possible to verify how the higher part, that is subject to greater peripheral speed and centrifugal force, was subject to exhaustion.

Moreover, compared to the time required for an object of comparable weight and shape (made in clay in January 2024) under the same conditions of temperature and humidity, the objects in paper-clay took more than twice as long to reach the leather hardness state.



**Fig. 5.** (a) Two bowls made on the pottery wheel in paper-clay with Faenza clay and white clay; (b) The bowl made on the wheel with Faenza clay; (c) A bowl made on the wheel with white clay. Note the exhaustion at the top of the bowl.

Finally, at this point, in addition to finding that the level of leather hardness tends not to be homogeneous in all parts of the object, due to the permanence of water in some parts, the finishing was particularly difficult, inaccurate and "dirty", because of some kind of hair brought by cellulose. Moreover, while the surface of the object tends to seal, due to the effect of finishing on the wheel, in the case of paper-clay, on the contrary, it tends to be more porous. The dry piece denotes greater lightness, even more once cooked, final phase in which even the sound of the object, in front of light percussion on the edge, bell-like, is more "woody", perhaps welcoming a suggestion dictated by the vegetable origin of cellulose.

## 2.5 AESTHETIC CHARACTERISTICS OF PAPER-CLAY AND ARTISTIC POSSIBILITIES

It was appreciated that the material offers interesting aesthetic characteristics, which vary with the different treatment that the material undergoes [7]. These differences in aesthetic performance, due to different exaltation of the technological characteristics of the material, were intended to highlight and unite all together in the same object.

In the example already seen of the bowl (Fig. 5b), moistening only one band and dropping on it the scrap of the finishing to the wheel (that in the case of the paper-clay is grainy), it adheres perfectly and definitively. Instead, depositing tufts of slip, it takes on a compact and non-porous appearance, due to the presence of water that is expelled very slowly.

Finally, slips of different colors (Fig. 5c) can be used as a bump to draw signs, in our experiment extremely hasty and crude, but that could definitely be improved.

Even the rough appearance and rough texture to the touch, even after finishing at the pottery wheel, could be exploited as an element of a primitive artistic poetics, while remaining the success of the material in the case of ceramic restoration with the integration of missing parts

### 3. CONCLUSIONS

This analysis reveals some extremely interesting points. The material, even in a very rudimentary phase of experimentation, already shows to contain in itself many potentialities that, in the continuation, will be transformed in more sure results. So far we have proceeded in such a way, as to be able to collect some first data, however comparable, and in any case certain indices of some behavior of the material.

Other experiments, as emerged in the course of this paper, are still to be conducted and others already conducted will certainly have to be repeated.

#### 3.1. POSITIVE RESULTS TO BE OPTIMIZED AND IMPROVED

**Different dosages.** Many experiments must be reconducted with different dosages and percentage, in order to get closer to optimized and reliable values.

**Different kind of clay.** The paper-clay made with porcelain, refractory, semi-refractory and pyrophilic products is certainly to be deepened.

**Interaction with glazes.** The interaction with the glazes must be verified, especially with regard to the opportunity of a single cycle of firing.

**Machinability parameter.** It will certainly be necessary to widen the possibility of additives which, by restoring the reliability of the material, increase its machinability parameter at the wheel.

#### 3.2. HYPOTHESIS STILL TO BE TESTED.

Another frontier, for now not very close and, for evident reasons, to be kept secreted by copyright, concerns the scientific interaction with another university that is developing the bacterial production of cellulose. If this road turns out to be possible, we will get even closer to a truly circular and sustainable material.

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