

# **Integration of Agro-waste in Fibrous form in DIY Composites for Prospective Design Applications**

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## **Authors' contributions**

*This work was carried out in collaboration between all authors. Author CS designed the study, wrote the protocol and wrote the first draft of the manuscript. Author MFZ offered suggestions on esthetical and perceptive analysis of biocomposites. Author AP developed the project and carried out analysis on carrot peels. Author MP developed the project and carried out analysis on nutshells. Author MM developed the project and carried out analysis on orange skins. All authors read and approved the final manuscript.*

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## **ABSTRACT**

Different forms of waste from food production chain have been integrated into self-produced (DIY) matrices, based on starch or flour systems, to form biocomposites suitable for the prototyping of small design objects. The idea is to investigate the production of items with expressive and functional value, so that the waste materials, no longer suitable for nutrition purposes, can be "upcycled" into something that can have a "bond" with the user, therefore being durable. In particular, some examples are reported, which are related to waste with different "structural" nature, such as carrot peel, nutshells and orange skins. In the absence of significant literature on this particular use of these waste materials in DIY matrices, this is to be intended as a preliminary study. The possible application of the materials, which will need in the future full mechanical and thermal characterization, is in the meantime discussed and some design objects are proposed. Moreover,

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ways to prevent the formation of mould and therefore extending the use life on the material are also suggested, in the understanding that the use of waste would require coming to an application in a material intended not for "single use" purpose.

*Keywords: Agro-waste; carrot peel; nutshells; orange skins; starch matrices; design applications.*

## 1. INTRODUCTION

In the present context, we are striving towards obtaining a "circular economy", therefore with "zero waste" and subsequently aiming at a complete use of it according to more recent EU guidelines, such as Directive 2008/98/EC, which suggest defining waste more precisely a "secondary raw material" [1]. This reflects the possibility to give a "second chance" or rather a "second life" to it, often in a design-intensive application, which gives also a non-negligible added value to it, by studying its "expressive" possibilities [2].

Waste from food or agricultural productive systems can be divided into different categories, according to their main chemical component: ceramics (normally based on calcium carbonate), such as mussel valves or egg shells; protein-based, such as waste from milk and derivatives; ligneous, such as peanut shells or coffee waste; cellulosic, such as orange skins or banana peel. Another possible classification is based on the "fibrosity" of the waste obtained, which is well recognized for some particular materials for example in food technology, as for use as dietary fibre [3].

Not all of this waste is of course adapted to use in composites, hence to be introduced in a polymer matrix: in practice, an interface of some strength is needed, which is not always the case. Recently, in the design field, to obtain a wider knowledge of the possibilities of materials from waste, they are often introduced in a self-produced (DIY) biopolymer matrix, which may be based upon waste as well [4]. In this respect, a few examples of work carried out in the Experimentation on Innovative Materials for Design course in Università di Camerino and their prospected application in design prototypes are presented, with the unifying characteristic of having at least some fibrous character, as for filler, therefore not starting from a waste reduced into powder with the idea of ensuring some "visibility" of waste into the composite, as per most recent indications of materials design using this kind of "second life" material [5]. Beyond the results specifically obtained, which depend on a number of factors, among which the nature of

waste considered is obviously central, this procedure, which is recognized as "material tinkering", in that it concerns developing a sensitivity to the sensorial and experiential qualities of materials, which is useful to propose not only the function of possible objects, but also possibly to predict their wide acceptance among public [6]. Three characteristics are in particular recognized as useful to start an appraisal of these aesthetic-functional properties on natural materials, to identify in these "high quality" ones: these are "fiberness", which is more sensory-based than "fibrosity", "reflectiveness" and "roughness" [7].

In particular, three examples are presented, which concern particularly diffuse waste from the food production chain, namely carrot peel, nutshells and orange peel, variably mixed with other components in a composite suitable for the production of small design objects. A specifically developed DIY biopolymer was used each time as for matrix, using glycerol, sometimes in combination with milk, or egg white as plasticizers. The problem with these biomatrices can be the inclusion of air, therefore the production of voids, as well as the formation of mould, which all are briefly discussed in the examples presented. In the latter case, in particular, the aim was trying to avoid the use of unsustainable chemicals for the purpose, a concern which is diffused from few decades on starch-based products [8]. It is quite obvious that the three materials reported are not ready yet for structural application and therefore were not subjected yet to mechanical testing, also for the very reason of obtaining samples for the purpose. Initial characterization carried out though show that the possible improvement of these materials from waste unsuitable for consumption is a possible objective, which is moreover a formative aim for the education of design students towards the demonstrative self-production of materials from waste.

## 2. EXPERIMENTAL METHOD

In Tables 1-3 are presented the different recipes that offered some results in terms of formation of DIY composites from carrot peel, nutshells and

orange skins, respectively. All the above waste were obtained from the student services of Università di Camerino, after meal preparations and before dumping, in a perfect “km-zero” concept. Vegetable glycerol with 99.5% purity was instead purchased from Greensistem. Together with the preparation of the composites, which were moulded into silicone moulds with the idea of obtaining design objects, on which further details will be given later on, also micrographs and macrographs were taken to investigate the level of porosity and uniformity of the material. Moreover, some preliminary tests for durability were also performed, which involved the use of a Radwag MA110 thermobalance with the application of different conditions of temperature. In particular, tests with thermobalance involved a program of heating with 5 minutes at 80°C, 5 minutes at 100°C and 8 minutes at 120°C, hence for a total duration of 18 minutes. Heating was applied on square samples of 20 mm side on each material. Some further tests were also performed with the idea of ensuring the possibility to use these materials in an external environment. In the specific case of DIY composites from carrot peel, which for their particular cellulose-based nature, were considered the most critical from the point of view of ageing, also this kind of tests was performed. In particular, Fourier transform infrared spectroscopy (FTIR) tests were performed in this case, using a Perkin Elmer Spectrum 100 system in attenuated total

reflectance (ATR) mode. The system used a zinc selenide crystal, with contact area of 2 mm diameter, penetration depth of 2  $\mu\text{m}$ , spectral resolution equal to  $4/8 \text{ cm}^{-1}$  ( $0.1\text{-}32 \text{ cm}^{-1}$ ), spectral range of  $4000\text{-}650 \text{ cm}^{-1}$  equipped with a deuterated-triglycine sulphate (DTGS) detector, with possible sample thickness of up to 1 cm.

The material was aged during 30 days at room temperature with the objective to estimate whether it could be subject of further development, having a non-negligible life duration. Microscopy and macroscopy images were obtained using a Motic DS300 apparatus equipped with a maximal magnification of 100x.

In contrast, on nutshells bioplastics, which of the three, due to the higher lignin content, in the region of 50% [9], was considered the most suitable for possible application in outdoor environment, also preliminary tests involving contact with running water or immersion in it were carried out. The heating cycle of 18 minutes total in temperature, mentioned above, was repeated also in this case, after immersion in water overnight, considering that possibly absorbed water should be evaporated during heating cycle. This ageing cycle did not respond to any particular ISO standard, but was just intended to assess the possibility of further use of these self-developed materials.

**Table 1. Recipe of DIY composite obtained from carrot peel**

Ingredients	Quantity (no added water)
Carrot peel	81 g
Potato flour	38 g
Vinegar	5 mL
Milk	5 mL
Glycerol	5 mL
<b>Procedure</b>	Boil carrot skins in water, then mash them in a blender and mix with other ingredient, cooking finally at 160°C for 2 hours
<b>Modifications</b>	Adding 5 g of coffee grounds or 10 g of beetroot juice for colour change

**Table 2. Recipes for DIY composites obtained from nutshells**

Ingredients	Mixture A (no added water) (Fig. 5a)	Mixture B (no added water) (Fig. 5b)
Nutshells	70 g	70 g
Wheat flour	38 g	38 g
Egg white (albumen)	110 g	70 g
Chamomile infusion	50 mL	50 mL
<b>Procedure</b>	Mash nutshells in a blender, then mixing them with egg white and wheat flour until they look uniform. Finally add hot chamomile to the mix and stir well. Cook at 150°C for 1 hour.	

**Table 3. Recipes for DIY composites developed from orange skins**

<b>Composition A (per 100 ml water)</b>	<b>Composition B (per 100 ml water)</b>	<b>Composition C (per 100 ml water)</b>	<b>Composition D (no water)</b>
10 g orange skins	10 g orange skins	10 g orange skins	10 g orange skins
10 mL glycerol	10 mL glycerol	10 mL glycerol	10 mL glycerol
16 g rice starch	15 g wheat flour	15 g wheat flour	15 g wheat flour
2 g isinglass	8 mL vinegar		50 g orange juice

In the particular case of orange skins, only a “manual” chopping using a knife has been employed for all formulations, in the understanding that the “character” of this waste has to be preserved in the final material.

### 3. RESULTS AND ANALYSIS

#### 3.1 Carrot Peels

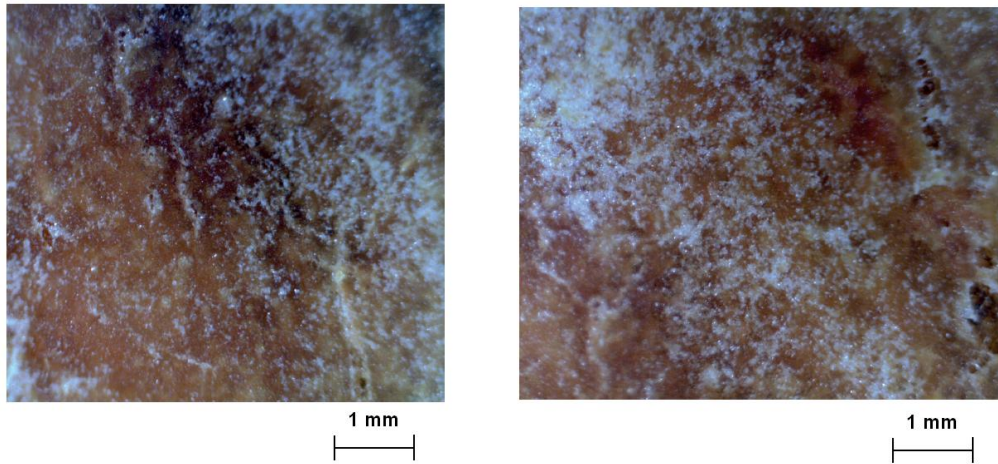
The high amount of carrot peels has offered a very distinct colour to the material, which results particularly stiff and does not present particularly significant air bubbles in its structure, as observable in Fig. 1, although some coalescent yet rare voids are present: no problems of mould formation were observed. It was suggested therefore that the use of carrot peels can be recommendable in a composite, in that carrot is recognized to have a wide anti-oxidant power as dietary fibre for the presence of  $\beta$ -carotene, which can be used also in the case of biocomposites as a natural anti-mould agent [10]. Compared FTIR analysis of the sample non-aged and aged for 30 days at room temperature, which is reported in Fig. 2, demonstrated that the position of peaks did not change, which indicated an absence of chemical reactions leading to disappearance of functional groups during the period and in particular no degradation of  $\beta$ -carotene and therefore suggested continuous anti-mould action, which in fact was visually perceivable from the samples. On the other side, carbonyl peak at  $1700\text{ cm}^{-1}$  intensified as an effect of aging, which can be recognized as the result of photo-oxidation [11]. As regards testing in temperature, the samples demonstrated having a constant weight loss with temperature, which amounted at the end of temperature cycle to around 3 ( $\pm 0.5$ )%. There has been no particular change in colour on the surface, although it assumed no distinctive smell with time and became more plastic with temperature (Fig. 3 middle). The colour of photoframes, obtained with silicone moulds in a chalk-made

external shell, was absolutely uniform with no air bubbles, and therefore it was proposed that the material could be coloured with other natural pigments, in that its structure may be suitable to allow this operation (Fig. 3 right). This proved also true with beetroot juice and waste coffee grounds, in the latter case introducing though some weakness due to the uneven distribution of coffee ground granules (Fig. 4).

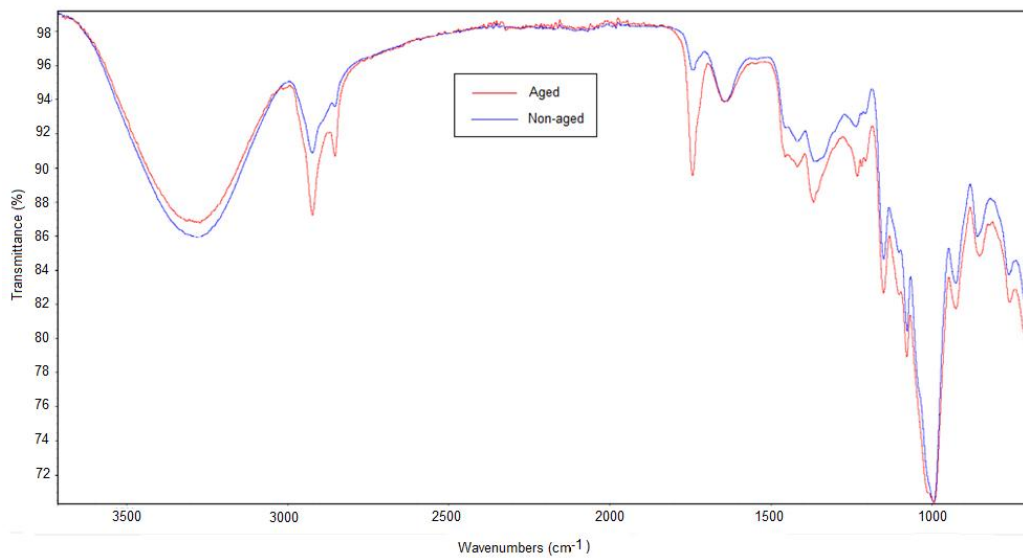
As a matter of fact, it has been recognised that carbonisation of coffee grounds leads to hierarchically porous materials, which might be an opportunity for the industrial production of very thin sheets [12]. In the present case, the control of material preparation in terms of heating time and temperature to achieve material uniformity proved quite cumbersome, although on the other side the colouring effect has to be considered fully effective.

#### 3.2 Nutshells

Initially problems were encountered in the creation of the recipes, especially due to the fact that ineffective mashing of nutshells results in the formation of air “pockets” inside the mixture with the material being too soft and glue-like and therefore in the gradual, yet very evident, formation of mould. However, after introducing the use of a blender, this issue was sorted. After establishing the recipe, the material after cooking appeared solid and apparently capable to withstand impacts. It is to be noticed, as from Fig. 5a, that the introduction of a lower amount of albumen i.e., 70 grams, in the mixture, all the other quantities remaining the same, brought the material to be more prone to flexure, due to the comparatively high weight of nutshell fragments and to the formation of voids. This problem was solved in the final mixture, whose surface is shown in Fig. 5b, where no voids are apparent any longer. The colour of the final material is slightly lighter than nutshells are, for the quite high amount of egg white.



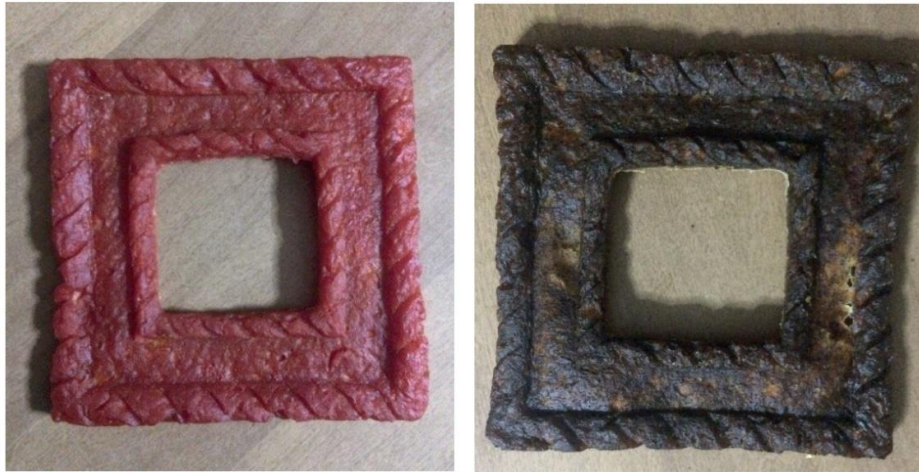
**Fig. 1. Surface colour of DIY composite from carrot peels**  
(left: front surface; right: back surface) (project by Agnese Petronella)



**Fig. 2. FTIR analysis of aged and non-aged DIY composite from carrot peels**



**Fig. 3. DIY composite developed from carrot peels for the production of photo-frames**



**Fig. 4. DIY composite from carrot peels for the production of photo-frames coloured with beetroot juice (left) or with coffee grounds (right)**



**Fig. 5. DIY composite from nutshells**  
(project by Michela Pelagatti)  
a. Side view of material from mixture A;  
b. Surface view of material from mixture B

A number of tests were carried out, which suggested that:

- Exposure under running water at room temperature for a period of 1 minute did not

bring to any alteration of shape or consistency of the material

- Immersion of the material in water overnight did bring in contrast to some alteration, in particular to a reduced stiffness and some weakness of the edges
- Thermobalance test did not result in particular absorption of water in the DIY composite, which was ultimately limited to less than 1% at 120°C.

On the other side, the development, yet with much more complex process, of a biomaterial based on albumen and wheat gluten is reported in literature, showing thermosetting potential [13]. It has to be considered also that the use of terpenoids, such as those obtained in this case from chamomile infusion, but also for example those obtained from thyme and cumin [14], does lead to a prolonged effect of natural preservation of the protein-based material from fungi causing mold appearance. This has a beneficial outcome especially in view of the design objects, such as those depicted in Fig. 6.

### 3.3 Orange Skins

In the case of orange skins, four different possible recipes (A-D) were comparatively assessed. As can be observed from Fig. 7, composition A appears not very homogeneous and has a considerable amount of voids, possibly due to the scarce compatibility between rice starch and isinglass, which in the end comes to the large formation of air bubbles. Composition B and C have both from visual inspection an elasto-plastic behaviour, and do appear at a first

glance quite mouldable and flexible, although in the former the orange skins are less visible than in the latter, which in the end is preferable. Vinegar was added to neutralize the basic effect of glycerol, yet in the end did not result in particular differences. Composition D is quite homogeneous and soft, although its thickness is not very uniform and there appears to be the possibility of detachment of small fragments overtime. Therefore, in the end it was decided to proceed with preparation of composition C and, as the consequence, some more analysis was performed on it.

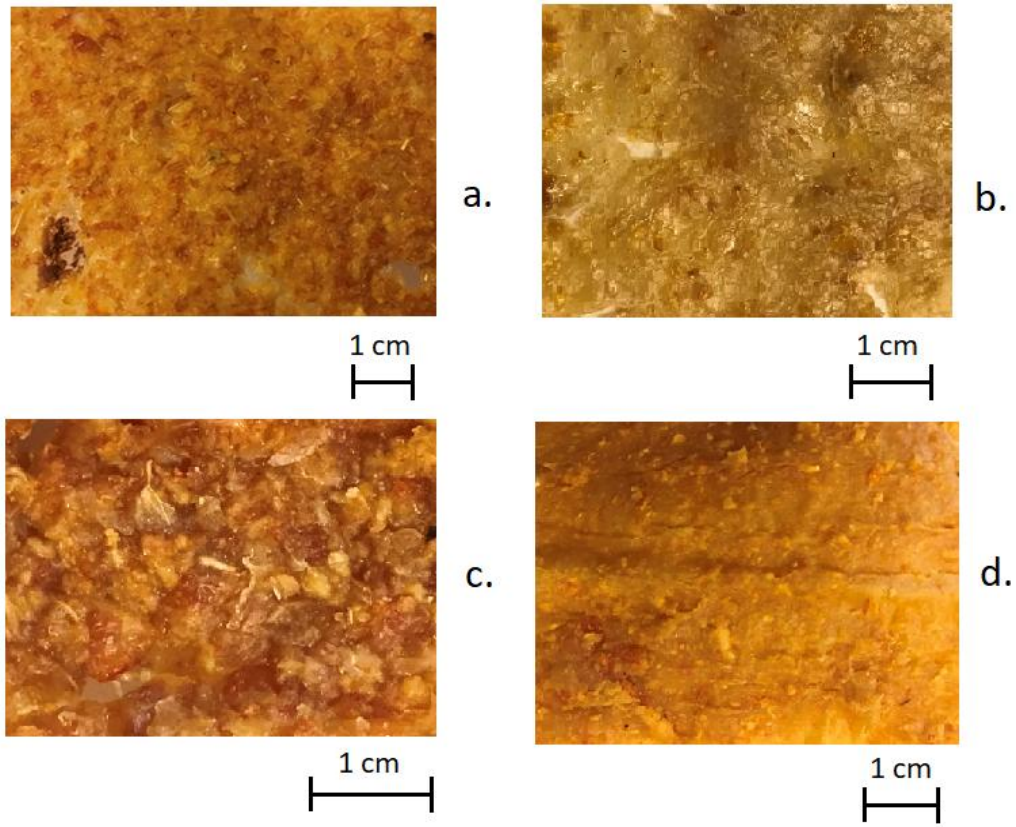
In particular, on DIY bioplastics with composition C, a sounder analysis from Fig. 8, concerning oven heating for some minutes at the temperature indicated, it is clear that weight loss at 80°C and above is very high, and it is therefore suggested that use above 60°C can be critical. Weight loss at room temperature was also measured and it offered an indication of the void content in the material, due to the formation of air bubbles during preparation, in the order of 2.8 (±0.3)%. Following this, immersion test for 30 minutes in cold water gave a lower weight gain, in the order of 1.4 (±0.2)%, while, as visible in Fig. 9, it did not create any strong change in the geometry and aspect of the material, apart from

some limited erosion of the edges. In general terms, it is suggested that the material could be used with moderate, hence accidental, exposure to water, whereas it cannot be guaranteed that it can resist, when immersed in water for a long time. It can be implied therefore that the objects are not washable, which might eventually be addressed for example with the introduction of a ceramic based waste, such as calcium carbonate from egg shells [15].

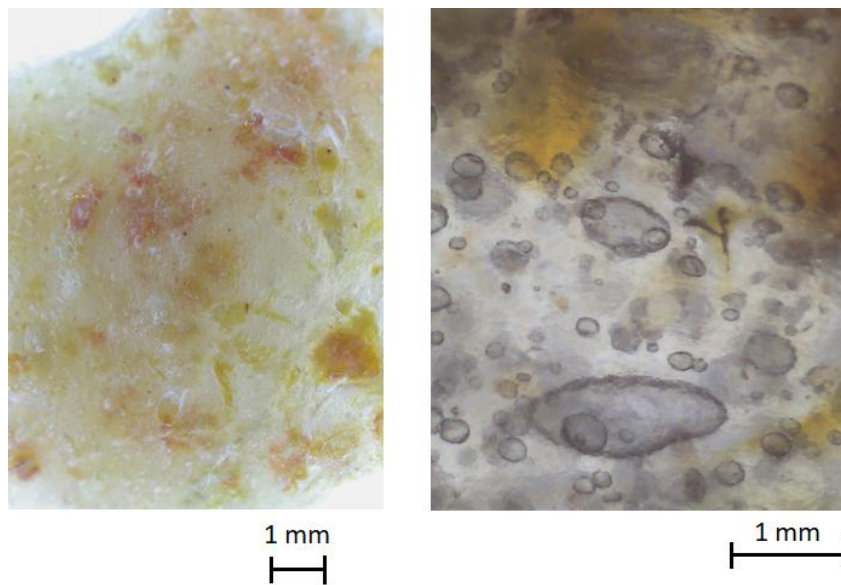
Orange peel is thus let to dry naturally for some time, then it is mashed and is mixed with the other ingredients in a silicone mould. Finally the material is left to cook in oven for 2 hours at 120°C. Orange peels are still visible, and the presence of some amount of voids and air bubbles are visible from macrographs and micrographs (the latter with a 20x magnification) in Fig. 10. Finally the design objects were obtained, as reported in Fig. 11, it is clear that elasto-plastic behaviour is still very visible, which brings to some geometrical mismatches between the samples, though with no particular detriment for their rigidity. The behaviour as regards mould formation did not present any particular problem: as a matter of fact, orange skins are already known for the effective extraction of essential oils [16].



**Fig. 6. DIY composite developed from nutshells for the production of various objects, including hangers, coasters, and CD holders**



**Fig. 7. Effect obtained with different compositions of DIY composites based on orange skins**  
(project by Matteo Marcucci)  
(the four sub-images a-d refer to the four compositions A-D, respectively)



**Fig. 8. Macrograph (left) and micrograph (right) of the surface of a sample of composition C**



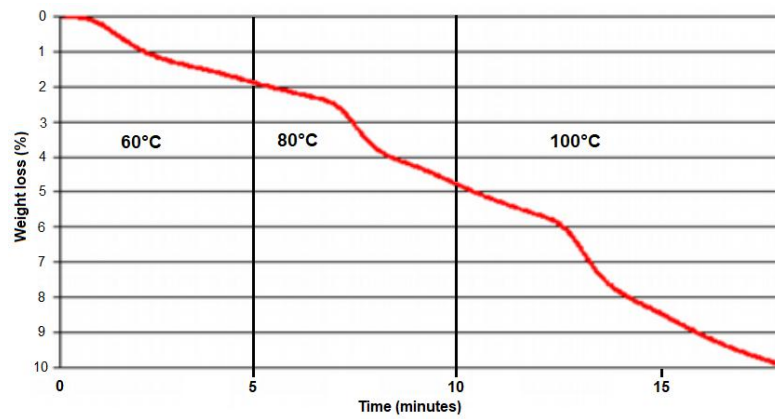


Fig. 9. Weight loss in a temperature cycle measured by thermobalance on composition C

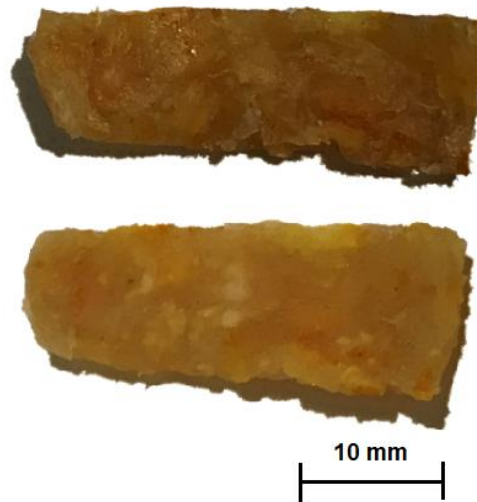


Fig. 10. Modification of the surface aspect of a sample of composition C

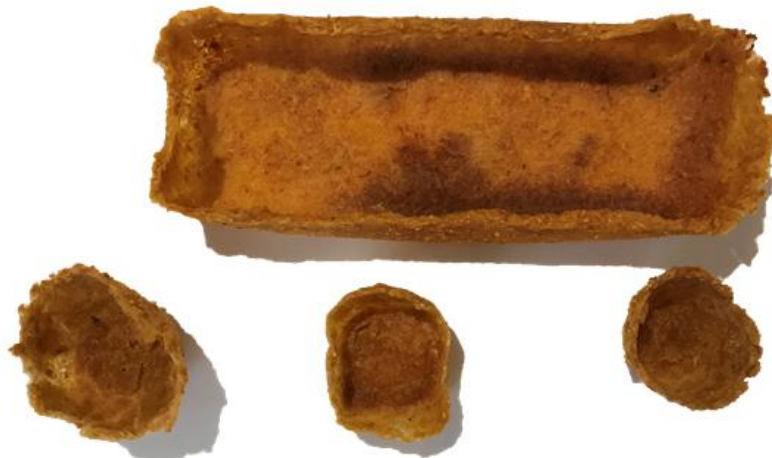


Fig. 11. Tray and cups obtained from the material

#### 4. CONCLUSIONS

The integration of different forms of waste from food production chain into self-produced (DIY) matrices, based on starch or flour systems, with different plasticizing systems, based e.g., on glycerol, egg white or vinegar, allowed forming biocomposites, which proved suitable for the tentative prototyping of small design objects. This procedure, which is intended as dynamical and susceptible of further development, is recently defined as “material tinkering”. This philosophy, which is inherent to learning materials design and has been recently defined as “material tinkering”, allows developing a sensitivity to the sensorial and experiential qualities of materials: this proves useful to propose not the function of possible objects, but also possibly to foster their wide acceptance among public. In particular, some examples are reported, which are related to waste such as carrot peel, nutshells and orange skins, producing some small design objects, mainly intended for use in-house, but prospectively developed also for a moderate use outdoors and in slightly aggressive environment. In this perspective, the possible application of the materials, which will need, whenever possible and reasonable, full mechanical and thermal characterization, is discussed and some design objects are proposed. Moreover, ways to prevent the formation of mould and therefore extending the use life on the material are also suggested, in the understanding that the use of waste would require coming to a durable application in a material not for “single use” purposes.

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#### COMPETING INTERESTS

Authors have declared that no competing interests exist.

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