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DIY Bioplastics from Peanut Hulls Waste in a Starch-Milk Based Matrix

A self-produced (DIY) material has been developed from peanut hulls waste in a starch-milk based matrix, which has been referred to as Peanmat. This is obtained after some attempts, which are also shown, of including this waste in a DIY bioplastic. It shows some potential in terms of sustaining its own load and to make small objects, such as buttons, for design purposes. Its limits were clarified by thermal characterisation in terms of not being able to exceed temperatures of around 80°C and suffering non uniform deformation, especially in the case its thickness does not go beyond a few millimetres. Colouration tests proved effective. Of course, the material is in search of full mechanical characterisation though it proved suitable to be punched and did not suffer fragmentation or excessive porosity.

Keywords: *bioplastics, peanut hulls, starch, milk, thermal charac–terisation.*

1. INTRODUCTION

In the present context, striving towards obtaining a "circular economy", therefore with "zero waste" or more precisely aiming at a complete use of it according to more recent EU guidelines, such as Directive 2008/98/EC, which suggest that waste is rather defined as a "secondary raw material". This reflects the possibility to give a "second chance" or rather a "second life" to waste, a procedure which is well known in packaging design, where packaging re-usable as such or for other purposes, for example small pieces of furniture, is increasingly preferred nowadays [1].

Peanut (*Arachis hypogaea* L.) is one of the most largely produced and used agricultural species in the world. Though a very abundant waste, the use of peanut hulls waste has been limited in materials so far: apart from incineration, often in the form of biomass briquettes [2], it has been e.g., proposed to use it as a source of activated carbon for adsorption of metal ions, such as cadmium, copper, lead, nickel and zinc [3]. It was also shown that peanut hulls are physically and chemically suitable for biofiltration as an effect of the presence of bacteria and fungi in them, as indicated by a six month experiment using methanol as air pollutant, releasing only carbon dioxide and water [4].

Also the use of peanut hulls as mulching material was proposed, as they can also ensure a significant resistance to freezing temperatures, this can be added to the beneficial effect of mycoflora on mulching process, although this is heavily dependent on the climate, not being adapted to very hot ones [5].

However, in most cases the use of peanut hulls waste as energy recovery through combustion is still carried out, which can be considered very limiting in terms of the possibilities embedded in this waste, which is fibrous and high in lignin. This has also suggested the possibility to extract polyphenols from peanut hulls, to apply them for antibacterial and antioxidant purposes, although it is notable as this also appears promising in terms of their suitability to their application in the production of biocomposites [6]. In practice, the percentage composition of hulls waste is the following, with very limited differences for different cultivars, 8.2% protein, 28.8% lignin, 37.0% cellulose and 2.5% other carbohydrates [7].

In structural terms, exploiting in particular the amount of lignin, though quite limited, contained in peanut hulls waste, their use in very fine form, such as flour or husk, for the production of particleboards was proposed [8-9]. It was observed though that this possibility decreased the mechanical properties of particleboards, as far as a higher amount of peanut hulls materials was introduced. On the other side, a valorisation process of waste would require to introduce it in the matrix in an amount as high as possible.

Received: September 2017, Accepted April 2018
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doi:10.5937/fmet1804503T

In practice, it was demonstrated that the application of wood hulls waste would only be possibly suitable in a mixture with other material richer in lignin, such as pine wood chips, with adapted adhesive e.g., urea-formal–dehyde, to create an interface between the two [10, 11].

Due also to the previously mentioned limited success obtained in the introduction of peanut hull materials in wood replacement boards, their employment as fillers for softer matrices, such as rubber ones, was also proposed [12]. In this case, there was more attention to the way they mixed up with the original material, and this led to the conclusion that the introduction of peanut hulls waste was advantageous in this case in terms of maximum torque, for example.

The more recent attention towards sustainability [13] reports again the interest over the use of this kind of materials, such as peanut hulls waste, as fillers for biocomposites. This can be suitable in general terms, although it has been clarified in various occasions as their behaviour might be very different from what is obtained both in engineering composites, such as carbon or glass fibre ones, even if a biomatrix is used [14, 15], such as polylactic acid (PLA) [16]. Anyway, it can be suggested that the self-produced matrix, adequately filled with waste, is used for purposes of additive manufacturing: however, the experience suggests that the chances of success are limited, if one wants to compare the performance of the obtained material with others, such as acrylonitrile-butadiene-styrene (ABS) rubber wire [17].

To understand more about these differences and the inherent possibilities they conceal, a possibility that is explored recently is material tinkering as a practical and creative approach to develop the sensitivity to possible application of materials through experiential learning. In practical terms, this reflects the need for attention towards waste "personality", involving a sound reflection on possibilities "embedded" into its nature [18]. This process will foster creativity and provide inspirations for materials understanding and experimentation, which will result in facilitating communication and dissemination about the possibilities of waste materials.

In particular, the present paper discusses the possibilities involved into the re-use of peanut hulls waste to develop from them DIY bioplastics by means of a material tinkering approach as an important exercise to elucidate the assets and limitations of waste upcycling [19]. In practice, the result of material tinkering, through an exercise of expressive-sensorial evaluation of the material, is the production of objects that may serve to the valorisation of waste without hindering it into the whole structure of the product, yet leaving it apparent though valorised. It is important at this regard to offer value also to the sensorial-expressive experience taken on the material, therefore having the widest and largest approach to the "personality" of waste material, so that design itself could be driven by it [20].

In the specific case of this work, two kinds of matrices have been proposed for the inclusion of peanut shell waste. The first one was a potato starch based one, which proved in literature flexible in terms of possible introduction of fillers, passing from cellulose-based ones [21] even to ceramic ones, such as halloysite [22]. In both these extreme cases, potato starch showed a comparable level of success when it comes to having a sufficient interface strength preserving though some rheological properties during preparation, especially by the addition of an adequate amount of glycerol.

The second one was a milk-based one, on the example of galalith, a material that was originally deve–loped for the replacement of horn as a protein-based imitation [23]. This possibility was suggested by the fact that also peanut hulls do contain some amount of proteinaceous material and therefore might be more compatible with it.

2. EXPERIMENTAL

2.1 Preliminary experiments

Peanut hulls, as shown in Figure 1, were stored at 4°C, with the idea of mashing them to powder before use. In materials terms, it is significant to observe that with respect to other external nutshells, peanut tend to have cellulose (whiter) and lignin (browner) concentrated in different parts of the hull. The same occurs for fibrous content, which makes the thickness of the hulls uneven and far from constant. This may explain why the mechanical performance of peanut hulls as filler in materials was ultimately rather poor and of course is a factor to take into account also in the application discussed in this work.

Figure 1. Peanut hulls as received.

On the possible introduction of peanut hulls, a preliminary reflection was first of all carried out, in terms of the fact of using water in the mixture or not. A possibility was for example the inclusion of starch in a water-vinegar mixture, such as it is commonly used in some traditional food preparations [24]

The first attempt was defined as mixture n.1: the relevant recipe is reported in Table 1.

Table 1. Composition of mixture n.1.

Ingredients	Quantity (per 100 ml of water)
Peanut hulls waste	30 g
Maize starch	45 g
Vinegar	5 ml

The mixture including peanut hulls mashed to a size of around 100 microns was blended for 3 minutes, gradually adding water and starch and finally vinegar. A final blending of the whole mixture was then applied for 10 minutes. A slow cooking in cuplike mould was then applied for 1 hour at 180°C. Demoulding proved easy and effective.

The limits of the bioplastic obtained were basically the following:

- Thickness needs to be significantly high (over 10 mm) to withstand its own weight
- Use of industrial corn starch is recommended to optimise mixture with water and vinegar, which implies that it proved not possible to use other starch-based waste, such as for example starch from potato skins
- The peanut hulls fragments are excessively mashed, threfore only limitedly visible as such, being rather assimilable to any wood-like waste

Despite this, small objects are possibly obtainable with this preliminary procedure, as reported in Figure 2. In particular, the first attempts concerned the creation of a keytrays and waterproof coasters. In this case, the replaced material could most likely be cork, with the additional opportunity of having different colours on the two surfaces of the object.

Figure 2. Result (mixture n.1) from including peanut hulls in preliminary tests from water-vinegar-corn starch mixture.

Therefore a second attempt (mixture n.2) was performed, whose composition is reported in Table 2.

Table 2. Composition of mixture n.2.

Ingredients	Quantity (no water)
Peanut hulls waste	30 g
Starch from potato skins	40 g
Glycerol	30 ml

Peanut hulls were mashed at a size not lower than 100 microns and to ensure a sufficient consistence of the mixture, this high amount of glycerol was needed.

The mixture n.2 was cooked in fan-assisted oven at 180°C for 15 minutes. It was possible to slightly reduce the thickness up to around 6 mm. The result, which tended to be not very uniformly cooked, is reported in Figure 3.

To try to reduce the amount of glycerol, an alter–native preparation (mixture n.3) was also attempted using isinglass, but still using water and basically applying no oven cooking. The idea was also to be able to reduce the thickness of the final material down to a few mm. The composition is reported in Table 3.

Figure 3. Result from mixture n.2.

Table 3. Composition of mixture n.3.

Ingredients	Quantity (per 100 ml of water)
Peanut hulls waste	30 g
Potato starch from skins	50 g
Isinglass	8 g
Glycerol	3 ml

Peanut hulls are blended till powdery and then potato starch is added, isinglass is melted in warm water, after this the whole is mixed, adding also glycerol and spreading on the mould. Then the obtained preparation is left to refrigerate for 12 hours.

The advantages of the material obtained are an evident translucency, a characteristic which allows the visibility of waste. In addition, the thickness can also be quite low, down to a few mm. On the other side, for mixture n.3 to work peanut hulls had to be powdered, which does not allow these giving a sufficient contribution to the texture of the material. The result obtained is depicted in Figure 4.

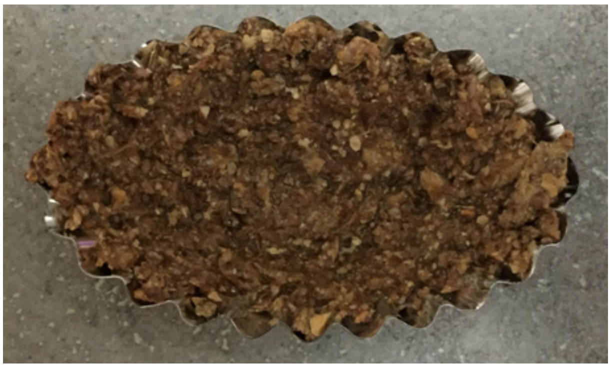

Figure 4. Result from mixture n.3.

2.2 Final preparation

A starch-milk mixture has been selected with the idea to produce a polymer gel from potato skins and lactose, obtained from milk whey, therefore two waste by-products of food industry. Their mixture results in a gel with properties that have been investigated already [25].

It is particularly interesting that these two wastes would show some mutual interaction in the material and would not show any glass transition phenomena at tempe–ratures below around 80°C. The above said interaction would possibly be improved as the effect of adding glycerol, though the resulting biopolymer would be likely to be quite brittle.

In practice, peanut hulls were mashed in pieces of dimensions around 100 microns to keep them recog–nisable in the mixture. The idea would be to obtain a mixture which is possibly to the best possible level workable by hand adding the maximum possible amount of peanut hulls waste to improve resistance without affecting the possibility to obtain a sufficiently sound interface between matrix and filler. In the end, a ratio between milk and potato flour in the region bet–ween 1.5 and 2 proved the most suitable for this purpose. Composition of Peanmat is reported in Table 4.

Table 4. Composition of Peanmat.

Ingredients	Quantity for a total of 100 g (no water)
Peanut hulls waste	17 g
Potato flour from skins	30 g
Whole milk (expired)	48 ml
Glycerol	5 ml

Measurement of the quantities was carried out within an accuracy of ± 1%. The mixed ingredients formed a compound that was cooked in a fan-assisted oven at 180°C for 13 minutes. The obtained material was called “Peanmat”.

Tests with Radwag MA 110.R thermobalance involved a program of heating with 5 minutes at 40°C, 5 minutes at 60°C and 8 minutes at 80°C, therefore for a total duration of 18 minutes. Heating was applied on square samples of 20 mm side with a thickness around 2.5 mm.

These tests were carried out on five samples of Peanmat compared with five samples of mixture n.2 and n.3 to see by comparison the effect of the addition of milk whey.

Tests on mixture n.1 were not carried out, because its excessive thickness and water consumption proved not effective for the purposes of the project. The heating program was selected according to the use envisaged (exposure to hot water for a limited period of time e.g., during washing) and relying on the absence of glass transition at these temperatures.

Ageing tests were carried out by exposing materials to an UV lamp for a period of 192 hours. Then FTIR-ATR (Attenuated Total Reflection) was carried out on five samples of Peanmat, comparing these for reference with five samples of mixture n.3, using a Perkin Elmer 100 spectrometer to compare new and aged samples of Peanmat plastics.

3. RESULTS

Despite the manual method used for the production of the material, the presence of voids in Peanmat appeared limited, being in the region of 3 (± 1) % of the whole volume, though in some cases not circular, but with some preferential orientation, which is likely to be due to the variable amount of filler in the composite: the appearance of the material obtained can be observed in Figure 5. The initial thickness of the material, which was produced with the process exposed above, was in the region of 2.5 (± 0.5) mm.

Thermal cycles carried out and monitored using the thermobalance, comparing mixture n.2 and n.3 with Peanmat (Figures 6 a-c), did indicate results in terms of weight loss which are reported in Table 5.

Table 5. Weight loss after thermal cycling of the different materials as measured by thermobalance.

Material	Weight loss (%)
Mixture n.2	7.6 ± 0.6
Mixture n.3	1.9 ± 0.3
Peanmat	6.5 ± 0.5

This indicated clearly that the effect of isinglass, present in the mixture n.3, was very evident in reducing the weight loss by providing an improved adhesion between the matrix and the filler. On the other side, the effect of the addition of milk in Peanmat, with respect to the starchy mixture provided as matrix in mixture n.2, did apparently lead to some minor improvements in thermal resistance.

However, whilst in mixture n.2 the weight loss, as from Figure 6a, was rather continuous and constant during the thermal cycle, in contrast in Peanmat, as from Figure 6c, weight loss progressed particularly in the initial period of time in which the material is brought from 40 to 60°C and then from 60 to 80°C, being more reduced then. This may suggest that the material becomes “accustomed” to heating, which may also indicate that a higher thickness, therefore a higher areal weight of the material, would possibly reduce the phenomenon.

On Peanmat, the global weight loss resulted in the appearance after heating of a white precipitate on the balance plate. The white patina observed after heating was only possibly removed with ethanol, not wiped out with water, thus possibly indicating that it did most likely originate from proteinaecous substance from the Peanmat material. Also, the colour of the sample after thermal cycling was no longer uniform, as it appears in Figure 7. Heating had another effect, evident from Figure 8, which is the non uniform dilatation: this, also to be attributed to the not high thickness, led to bending of the material and to the appearance of extensive voidage.

Concerning FTIR analysis, reported in Figures 9a and 9b, the graphs of pre-aged and post-aged mixture n.3 material, with isinglass, but without milk, were basically unchanged, an only modest variation of intensity of transmittance can be evidenced around 1690 cm^{-1}. This is likely to be referred to a modest reduction of the effectiveness of isinglass, as reported also in [26].

Also Peanmat material showed limited changes, although with larger evidence and range than for mixture n.3: in particular, the main changes refer to the range between 1545 and 1645 cm^{-1}.This variation are likely to refer to the denaturation of milk proteins present, which would also explain the white trace observed after heating [27].

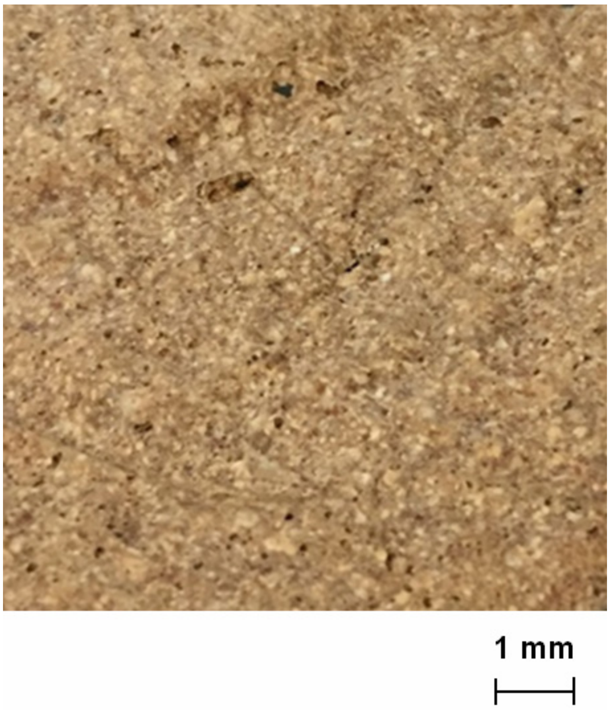

Figure 5. Surface aspect of the Peanmat material.

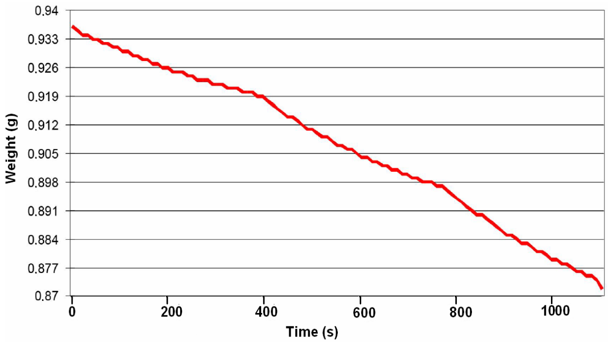

Figure 6a. Example of weight loss of mixture n.2.

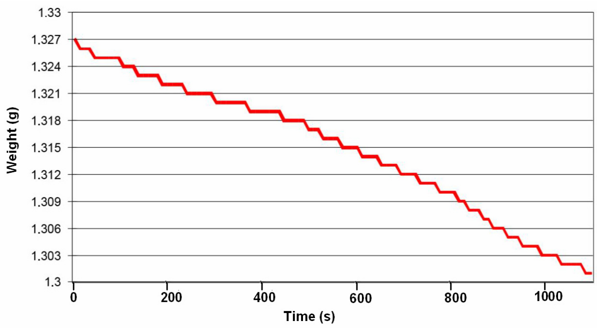

Figure 6b. Example of weight loss of mixture n.3.

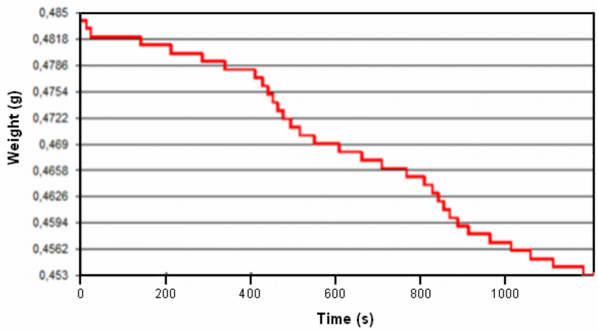

Figure 6c. Example of weight loss for Peanmat.

These initial results have indicated that this material is applicable in elements of sufficient thickness, possibly exceeding 5-6 mm. Its non-uniformity tends to discourage application at temperatures above 80°C: however, the interest for the material is confirmed in that it is able to sustain its own weight, does not show excessive porosity and is able to re-utilise waste, a process to which design can offer substantial oppor–tunities [28, 29]. This is the aim of the following part of this paper.

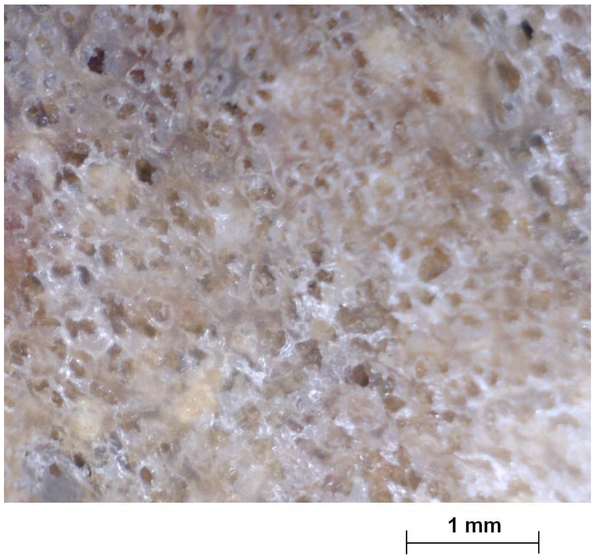

Figure 7. Microscopic appearance of Peanmat samples after thermal cycling.

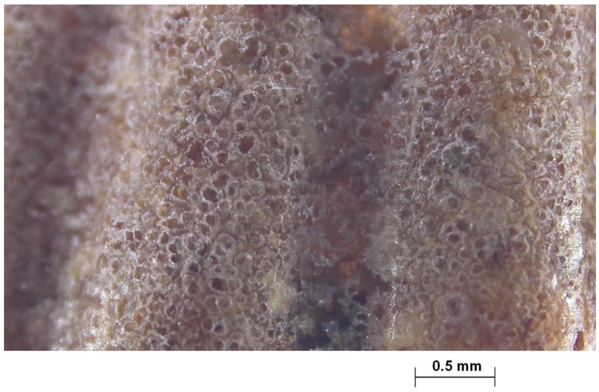

Figure 8. Non-uniform bending of Peanmat sample after thermal cycling.

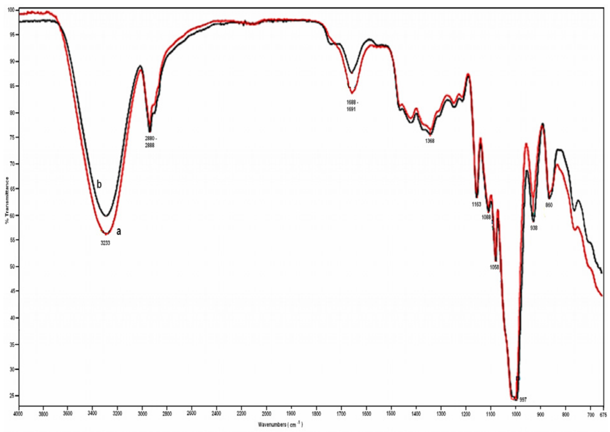

Figure 9a. FTIR-ATR spectra of newly produced mixture n.3 (a) and aged one (b).

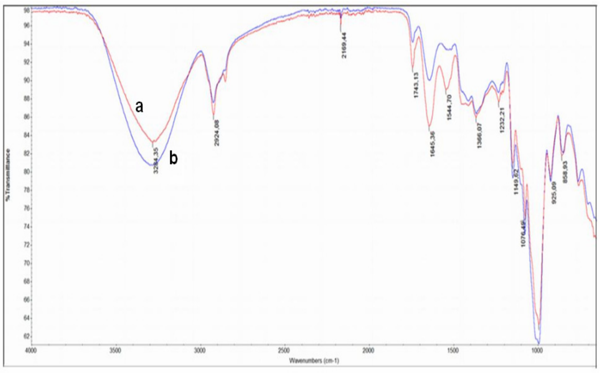

Figure 9b. FTIR-ATR spectra of newly produced Peanmat (a) and aged one (b)).

4. DISCUSSION AND DESIGN APPLICATION

To better elucidate the effect of possible development and to test the material consistence, the example was followed of what was attempted as for use of material obtained from mixture 1, which is depicted in Figure 10, though with considerably higher thickness. For this purpose, small cake cups have been initally created by means of a silicone mould and countermould. Demoul–ding proved easy and fast and, as suggested by Figure 11, the mashed waste is still visible in the texture.

Colouration tests were also carried out by the addition of a small amount, in the order of around 7 wt. % of blackberry juice, 1 wt.% of saffron powder and around 4 wt.% of mashed beetroots. From experiments carried out it is suggested that, to have an effective application of colouring agents, they need to be concentrated enough, such as colouring with blackberry juice did not produce a sufficient effect, in that the woody colour of Peanmat proved still prevalent.

It is widely recognised on the other side that the stability of colouring with natural substances, therefore suitable also for food, depends on a number of factors [30]. In this specific case, the need to maintain the waste visible from the material texture did worsen the prob–lem. The effect is positive though for the two colouring agents indicated above and is shown in Figure 12.

The positive results obtained with the objects exposed above did suggest the possible application of Peanmat material for approximately cylindrical objects, with a quite low ratio between base diameter and height, such as buttons. For the purpose, a cylindrical smaller mould was used, which is depicted in Figure 13. The use of countermould would avoid the possible irregularities of the edges. As reported in Figure 14, from initial moulding, different shapes can be obtained by hand, in which holes can also be punched during oven heating when the material is still sufficiently soft, by means of a toothpick, for sewing. In this way, the buttons can be applied on different items, such as garments and bags (Figure 15 and 16). Some criticity can be referred to the fact of machine washing, especially at high temperature, which may induce their damaging or fragmentation. It is therefore suggested they might be either hand washed or removed and re-applied after washing, the latter applying particularly to use for bags.

In general terms, this work can be explained as an attempt to re-use waste in a new DIY material, which is definitely in search of a fuller mechanical charac–terisation in the future, but it is of particular interest for the possible inclusion of different wastes into a context of "revived beauty". This requires a new kind of aesthetical and functional appreciation, which on the other side enters into a new perspective in which waste is clearly distinguished from by-products and needs to be re-used in materials as such and made visible as much as possible [31]. The consequence of this procedure is that waste is defined as such only in case it is explicitly declared it is.

This causes the attention to move towards the way waste can be prevented to be produced. It is therefore important to offer to waste a use and a time perspective that is compatible with its nature and confers to it some added value: this procedure is commonly defined as "upcycling" [32]. It is clear that the creation of "use and throw" objects is excluded in this context and therefore it is suggested that what has been performed with Peanmat enters in this general frame.

The natural reference of Peanmat is galalith, the casein-based plastic from which buttons, but also for example dice, were obtained in the 40s [33]. The advantages of Peanmat over galalith would also likely to be a higher resistance to the development of fungal moulds, a common problem in casein materials, as known also from glues used in historical heritage [34]. In addition, also the possibility to use natural colouring, such as those experimented in this work (e.g., saffron) would contribute to this anti-mould process.

5. CONCLUSIONS

This work concerned a self-produced material has been developed from peanut hulls waste in a starch-milk based matrix, in the idea to upcycle waste, therefore offering it higher value not using it in "use and throw" object, but rather for design items having some emotional, other than physical value. This material showed some potential to produce small objects, not hiding the waste from which it originated, being possibly coloured with natural pigments. Thermal cycling proved critical for the material, leading to the denaturation of the proteins present, most likely those from milk. Another indication of this work is the possibility to rediscover milk-based plastics, such as galalith, for the production of commodity objects, such as trays, cups and buttons. This would be done in a waste upcycling perspective, the limits of the material being the fact of not being able to exceed temperatures of around 80°C and suffering non uniform deformation when moulded at low thicknesses. Initial investigations indicated that the material, indicated as Peanmat, is able to be punched and shaped with different geometries without suffering either excessive fragmentation or excessive porosity. Following the same eco-friendly trend as that of green composites, which have had their mechanical properties thoroughly validated in recent works [35-36], in a future study it would be desirable to quantify tensile, compressive and impact properties of the Peanmat in order to precisely understand its potential applications.

ACKNOWLEDGMENTS

This work has been developed in the frame of the course of “Experimentation on innovative materials for design” at Università di Camerino, School of Architecture and Design. The course has been based over years 2015-16 and 2016-17 over the upcycling of waste into new materials, elaborating over a large range of different waste.

Figure 10. Example of object obtained from mixture n.1.

Figure 11. Example of cupcake sample.

BLACKBERRY

SAFFRON

BEETROOT

Figure 12. Colouration tests of the “cupcake” samples.

Figure 13. Cylindrical mould for buttons.

Figure 14. Peanmat buttons of different shapes.

Figure 15. Application of Peanmat buttons on garments.

Figure 16. Application of Peanmat buttons on a bag.

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DIY БИОПЛАСТИЧНИ МАТЕРИЈАЛИ ИЗ ОТПАДА ЉУСКЕ КИКИРИКИЈА У МАТРИЦИ ЗАСНОВАНОЈ НА СКРОБУ

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Само произведени (DIY) материјал развијен је из отпадака љуске кикирикија у матрици заснованој на скробу, који се назива Пеанмат. Овај материјал се добио након више покушаја, који су такође приказани, укључивањем овог типа отпада у биопластику DIY. Овај материјал показује потенцијал у погледу одржавања сопственог оптерећења и ства–рања малих предмета, као што су дугмад, за потребе дизајна. Његове границе су објашњене термичком

карактеризацијом у смислу да не могу да пређу температуру од око 80° C и да не трпе неједнаке деформације, посебно у случају када његова дебљина не прелази неколико милиметара. Тумачења су се показала ефикасним. Наравно, материјал је у потрази за потпуном механичком карактеризацијом, иако се показао погодним под ударом и то без фрагментације или претеране порозности.