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A sustainable bioplastic obtained from rice straw

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ABSTRACT

A new bioplastic was produced from rice straw, an agricultural waste that generally is not recovered. For the synthesis the sample was treated by using the Naviglio extractor, then it was dissolved by trifluoroacetic acid. The material exhibits good mechanical properties, with tensile strength and elongation at break equal to 45 MPa and 6.1% and 10 MPa and 63% for dried and wet dumbbells, respectively. It results that the mechanical properties of the produced bioplastic, in its dry state, are comparable to those of polystyrene, while cast bioplastic in wet state is similar to plasticized poly(vinyl chloride). This demonstrates the high mechanical performance of the newly obtained bioplastic both in dry and wet status. The morphology of bio-based material, investigated by scanning electron microscopy, showed a uniform and compact surface structure. 2D X-ray Diffraction analysis reveals that bioplastic is essentially amorphous. Mass loss test noted that it is completely decomposed after being embedded in soil for 105 days. Industrial and environmental advantages of the newly obtained biomaterial are evaluated in terms of embodied energy and CO_2 footprint production and in comparison with thermoplastic starch and other plastics. Finally, shape memory test revealed promising dual shape effects of the biomaterial, with a partial but significant shape recovery. In summary, depending on the environmental humidity, the material shows a dual mechanical behaviour that can be exploited to obtain shrink films and sheet or to drive shape memory effect. Therefore, we suggest rice straw bioplastic as a new potential eco-material for different application fields.

1. Introduction

Plastics are used to make a wide range of products in different application fields thanks to their versatile properties. As a result, the production of plastics has increased substantially over the last 70 years from nearly 0.5 million tons in 1950 to over 365 million tons in 2016 worldwide, and nearly to 66 million tons are produced in Europe (PlasticsEurope, 2017; Thompson et al., 2009). Conventional polymers contain additives such as stabilizers, pigments, and toxic chemical components such as flame retardants, antioxidants, and blowing agents (Bach et al., 2013; Hahladakis et al., 2018; Hermabessiere et al., 2017). Although there are a lot of advantages of plastic application compared to other materials (Andrady and Neal, 2009) their drawbacks are becoming increasingly evident. Currently, the huge amount of plastic waste production is one of the most-faced issues over the world both for environmental problems and human health threat. Plastics are the major components in municipal waste. Another concern is due to their non-biodegradability. Since plastics are persistent for many years, their waste should be addressed to an adequate management treatment (Teuten et al., 2009). For this reason, it is mandatory to drive the demand for new and innovative material solutions, which should be cost-effective and environmentally biodegradable. Eco-friendly bio-composites from plant-derived fibre (natural/bio-fibre) and crop-derived plastics are novel materials produced by vegetable waste (Bayer et al., 2014). Bioplastics are defined as a new plastic generation that significantly reduce the environment impact

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in terms of greenhouse effect and energy consumption (Mohanty et al., 2002).

Rice straw is a vegetable waste with abundant cellulose (32–47%), hemicellulose (19–27%) and lignin (5–24%) (Garrote et al., 2002; Saha, 2003). It is known as a potential feedstock for fuel ethanol production (Binod et al., 2010). It is reported that for nearly each kg of crop harvested, approximately 1–1.5 kg of straw is produced. Since rice is known as the third-most produced grain crop, it is possible to estimate a global rice straw production of approximately 1000 million tons per year (Maiorella, 1985). However, rice straw is considered an agriculture waste, and in several countries, it is either dumped into rivers or burned in the field causing greenhouse gas emissions, contaminations, and pollution (Sangon et al., 2018). Currently, several agricultural by-products and inedible food waste are considered in the production of some eco materials: potato peels, sugar cane bagasse, whey protein, shrimp shells, egg shell, lignocellulosic fibres derived from apple and orange fruit juice extraction, are some examples (Tiimob et al., 2017; Chiellini et al., 2001; Moro et al., 2017). It is also important to remember that different food waste categories are difficult to separate; therefore, the possibility to obtain bioplastic from these residues must be coupled with a suitable waste management strategy. Concerning rice straw, this waste can be easily managed because it does not require separation from other waste (Dominguez-Escriba and Porcar, 2010).

Application fields of bioplastics are numerous: food packaging, medical care, horticulture, agriculture, electronics, etc. Food packaging is the most important field (Gilbert, 2017), which requires resistance to moisture and fat or oil. The performance expected by bioplastics in this application field is protecting the food from environment and guarantee food safety and quality (Siracusa et al., 2008). Although significant improvements have been achieved, there are still some restrictions to be overcome. Some properties such as poor heat sea lability, thermal instability, high water vapour, brittleness, and low melt strength need to be optimized. Indeed, the abovementioned drawbacks have opened up research opportunities to improve the functionalities of the bioplastics. Coating, blending, addition of nanoparticles, addition of cellulose, and chemical/physical modification are some of the methods proposed to overcome the aforementioned limitations of bioplastics (Sartore et al., 2015, 2016). However, it is extremely difficult to propose a low-cost bioplastic with mechanical properties comparable to plastic material, such as polyethylene (PE) and polypropylene (PP).

Different tests performed on bioplastics show another advantage compared to conventional plastics, their shape memory capabilities. This means that they are able to change their shape upon application of an external stimulus. This phenomenon is observed in various polymers with significant differences in their chemical composition, but having common elements on a structural basis. In fact it is known that molecular structure is responsible for the shape memory properties of polymers and in the case of typical shape memory polymers it is composed of reversible and fixing phases. The reversible phase, which is in charge of shape variation, may be crystalline or amorphous, while fixing phase, which is responsible for the permanent shape, can be either physically or chemically cross-linked points (Bai et al., 2016). Based on literature studies, heat, light and immersion in water are used as external stimuli to cause shape change (Behl and Lendlein, 2007; Huang et al., 2005; Lendlein and Kelch, 2002). It is already proved that shape memory polymers undergo deformation and recovery cycles under much higher strain levels compared to metallic alloys, where the maximum deformation is only 8% between the permanent and temporary shapes (Lendlein and Kelch, 2002; Wei et al., 1998).

In this study, rice straw is used to produce new cellulose-based bioplastic materials. After an extraction pre-treatment, cellulose present in rice straw interacts with trifluoroacetic acid (TFA) (Bayer et al., 2014), to produce cellulose-based bioplastic materials. TFA is a volatile organic acid and has already proved to be one possible non-aqueous solvent for cellulose swelling (Zhao et al., 2007). Moreover, it is easily recyclable by distillation and is miscible with many organic solvents, as well as water, a quality rarely observed in other mineral acids. As far as we know, no one has attempted to obtain bioplastic from rice straw through the proposed approach. Mechanical properties are studied through the tensile test of dried and wet dumbbell specimens. Shape memory capabilities are also considered. Material loss test under soil moisture is carried out. Finally, sustainability of the proposed bioplastic in terms of energies and emissions required for the synthesis is evaluated.

The aim of this work is to propose a method to realize new bioplastic from rice straw and to investigate the new obtained eco-materials properties.

2. Experimental section

2.1. Extraction procedure

Approximately 200 g of dried rice straw samples were placed in a membrane bag and transferred to the chamber of the Naviglio Extractor, 2000 mL (Naviglio extractor®, Atlas Filtri Ltd., Padua, Italy). The extraction is based on the generation of a negative pressure gradient from the inside to the outside of the solid matrix. Approximately 1 L of Milli-Q water was added. The two extraction chambers are connected by a pipe with an electric valve, which is closed during the hydration process and opened to evacuate the liquid from the system (Naviglio et al., 2007, 2013). Total extraction was performed using a rapid dynamic solid-liquid extractor by applying cyclical pressure to the liquid in contact with the rice straw material for approximately 3 h and with 30 cycles and 12 strikes per cycle and the static phase for 10 min, as reported in the instrument manual. A pressure gradient ranging from 0 to 10 bar is applied, and the system reaches the equilibrium at a pressure of 8.5 bar. After the extraction, the sample was washed and completely dried under laminar hood at room temperature. It is important to highlight that this process does not damage the plant cells and is faster, easier and more complete with respect to maceration. From an economical point of view, this instrument is cheaper and requires a minimum consumption of energy compared to extraction performed by other techniques such as supercritical fluids (Motohashi et al., 2000).

2.2. Sample preparation

Dried rice straw was ground into fine powder using an agate mortar and sieved through 300 μm sieve. Approximately 10 g of the powdered rice straw was mixed with 200 mL of TFA. The solid/liquid ratio was optimized to 1:20 (w/v). Then, it was maintained under magnetic stirring (about 800 rpm) at room temperature for three days in a glass laboratory flask (Pyrex®, 1 L) and poured into a low edge crystallizing container (Pyrex®, 19.2 cm in diameter and 2 cm in height). After that, the container was maintained under laminar hood and covered using a glassware case to slowly remove TFA.

2.3. Characterization

2.3.1. SEM analysis

The morphology of the material was characterized by a Zeiss LEO EVO 40XVP (Carl Zeiss Microscopy GmbH, Oberkochen, Germany) scanning electron microscope (SEM) equipped with a Link

letter of commitment — B-1
B-2 — CV of experienced research
capacity of participating organ.
Ethical Aspect

Analytical probe for energy dispersive X-ray spectroscopy (EDXS, Oxford Instruments NanoAnalysis, Buckingamshire, UK). To characterize the composites, the morphology of cross section and surface of rice straw bioplastic were scanned under secondary electron mode.

2.3.2. 2D-XRD and FT-IR measurements

2D X-ray diffraction (2D-XRD) measurements were performed with a D/Max Rapid diffractometer equipped with a Cu anode (Cu Kα radiation) operating at 40 kV and 40 mA and a cylindrical image plate detector, to collect data from 0° to 160° (2θ) horizontally and from −45° to 45° (2θ) vertically.

Fourier Transform Infrared spectroscopy (FT-IR) was performed with an Equinox 55 (Bruker) spectrometer operating in transmission mode. Each sample was analysed in triplicates and the spectra were recorded from 4000 to 400 cm^{-1} (average of 128 scan per sample and 4 cm^{-1} of resolution). The obtained spectra were baseline corrected using OPUS software, version 5.0.

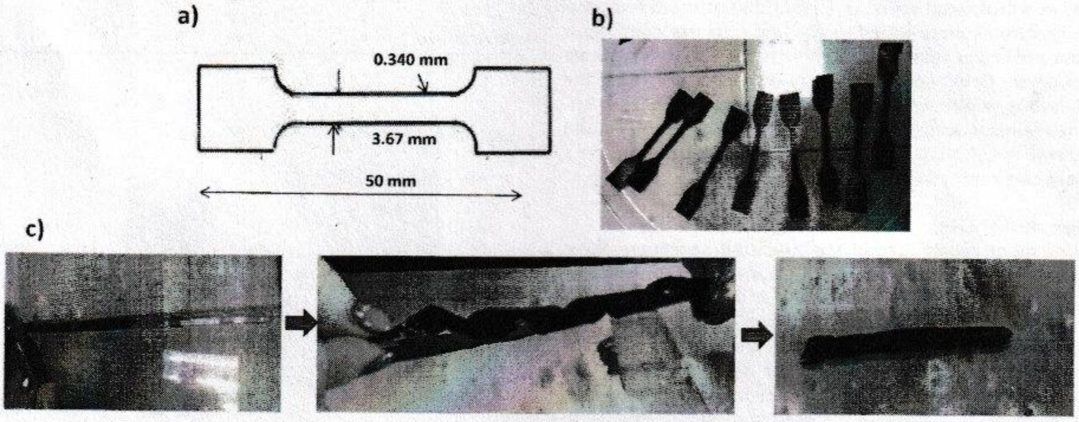


Fig. 1. a) Scheme of dumbbells; b) dumbbells cut rice straw bioplastic specimens; c) preparation of cellulose-based material with a helical shape.

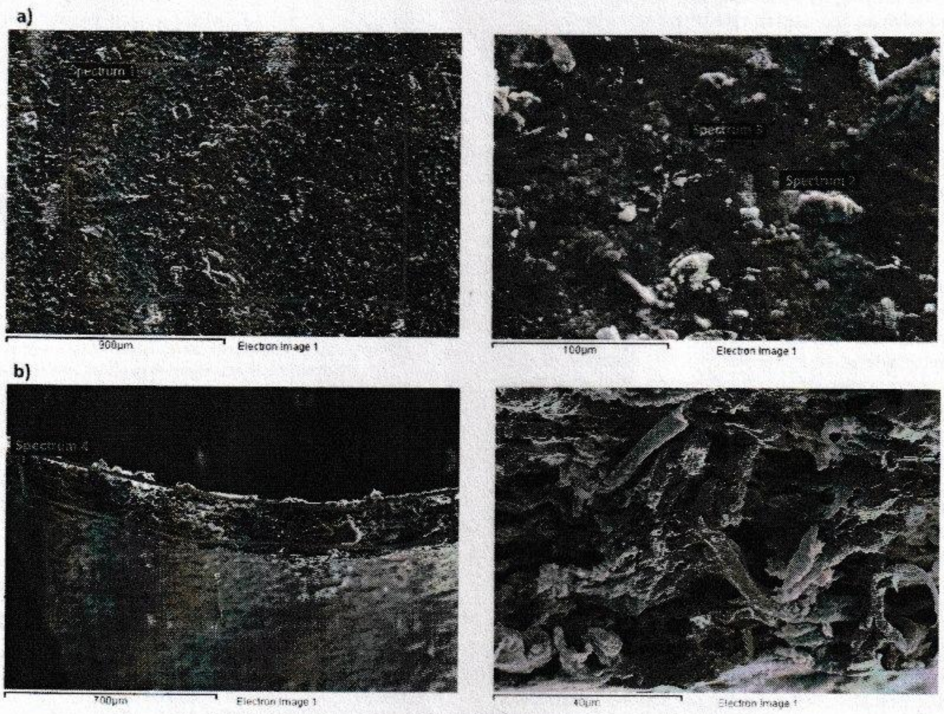


Fig. 2. SEM images of the surface (a) and cross section (b) of rice straw bioplastic. Images were collected in secondary electron mode, at the following magnifications: a) 60× and 450×, b) 75× and 1200×.

2.3.3. Thickness and width measurements and tensile tests

Samples were cut into dumbbell specimens (Fig. 1a) according to ISO 527-2/1BB, 1996. Thickness measurements were performed by a digital micrometer Mitutoyo 1P65 Coolant Proof (0–25 ± 0.001 mm). Three different points were measured along their narrow section, as described in the D 638–02a Standard (ASTM D638: 2003). A travelling length microscope Leica MS 5 was employed to measure the dumbbell's width at three different points. According to D 638–02a, the width of this specimen is the distance between the cutting edges of the die in the narrow section of dumbbells.

An electromechanical dynamometer Instron Mod 3366 Tensile Tester equipped with a 500 N load cell is employed. The tests were conducted at a crosshead speed of 10 mm/min at room temperature. The specimens were tested under two different conditions: after drying and in wet state. At least three specimens for each state were considered. Dried specimens were obtained by cutting the prepared casting, while wet dumbbells were measured after immersion in deionized water for 1 h. The force on the specimen and its displacement were continuously monitored and plotted on a stress-strain curve until failure.

2.3.4. Shape memory test

Shape memory capabilities of the as-casted specimens were investigated by taking into account the transformation between two specific shapes: (i) transformation of a flat specimen into a helical shape and its recovery to a flat shape and (ii) transformation of a helical specimen into a flat one and its recovery to the helical shape. In the former case, a flat specimen (length 15.5 cm) was immersed in water (1 h) to obtain an adequate wetting and softening of the material, wrapped in the wet state around a glass rod, and dried while kept fixed in this position. Hence, shape recovery was determined by immersing the specimen in water at room temperature and monitoring its shape shifting during the time by means of a camera with an acquisition rate of 1 frame every 30 s (2 frames per min). The other case was studied in a specimen originally with a helical shape, obtained by spiral cutting of a tubular specimen which was prepared by casting the solution on a glass rod. Fig. 1b shows preparation of the helical specimen. The specimen with a helical shape was immersed in water for 1 h, flattened in its wet state and dried while kept flat. Once the specimen was fixed in the temporary configuration, its recovery to the initial shape was performed by immersing it in water and monitoring its shape shifting by means of a camera (acquisition rate 1 frame/s).

2.3.5. Material decomposition test

Paper (cellulose-based) sample, rice straw bioplastic material, a piece of plastic from a certified compostable bag (Mater-bi) and a conventional poly(ethylene terephthalate) (PET) sample of the same dimension were taken into consideration. Samples were weighed separately and buried in soil. Material mass loss test was performed during the winter time from November to January. The mass of each sample was weighed on alternate days.

3. Results and discussion

3.1. Bioplastic preparation and characterization

Cellulose regeneration from its derivatives usually requires extra time and costly chemical purification steps. In the present study, we used TFA as a unique solvent to dissolve rice straw powder; it co-solubilizes cellulose with other organic matter. Solvent cast films and sheet were obtained after TFA evaporation. Solvent-casting is a century-old film-making process that has taken many years to develop into a high precision technique (Sata et al., 2004; Saha, 2003). Today, it is used to produce engineering plastics, optical films, medical films and sheet forming for electronic applications. In addition, being the solution and polymer film exposed to relatively low thermal and mechanical stress throughout the entire

Table 1
Results of energy dispersive X-ray spectroscopy (EDXS) performed on rice straw biofilm.

Sample	Spectrum	Elemental Concentration (%)				
		C	O	Si	Ca	Ti
Surface	1	37.1	51.1	11.8	–	–
	2	31.5	32.1	34.6	–	1.7
	3	50.0	34.4	14.5	1.2	–
Cross-Section	4	39.6	51.4	7.7	1.4	–

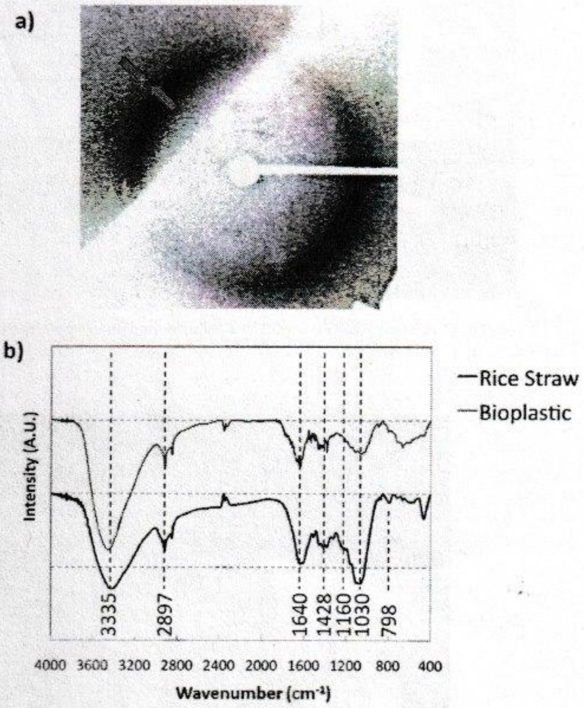


Fig. 3. a) 2D-XRD image of bioplastic. Arrows indicate the position of crystalline peak at about 24°. b) FT-IR measurements of rice straw (black line) and bioplastic (blue line).

Table 2
Thickness, width and mass of dried and wet dumbbells of rice straw.

Parameters	Dumbbell					
	Group I			Group II		
	1	2	3	1	2	3
	Dried			**Dried**		
Thickness (mm)	0.344	0.331	0.247	0.313	0.326	0.327
Width (mm)	3.458	3.429	3.542	3.576	3.451	3.415
Mass (g)	0.201	0.223	0.191	0.203	0.218	0.228
	Wet (After immersing)					
Water Absorption (%)	42.6	41.6	40.7			
Thickness (mm)	0.414	0.437	0.402			
Width (mm)	3.683	3.724	3.621			
Mass (g)	0.287	0.316	0.269			

production process, degradation or adverse side reactions are not significant. For these reasons solvent-casting is ideal for manufacturing films containing heat-sensitive ingredients and polymers such as poly(vinyl chloride) (PVC). The bioplastic from rice straw showed the main requirements for the solvent casting manufacturing process. In fact, (i) the row material was soluble in a volatile solvent, (ii) a stable solution with a reasonable minimum solid content (the solid/liquid ratio was 1:20) and viscosity was formed, (iii) formation of a homogeneous film and release from the casting support was possible. The obtained film and sheet were continuous, flawless, flexible and resistant to tearing. The solvent casting technique, applied in this work in a laboratory-scale production, could be improved to an automated production processing (Siemann, 2005). The browning of the obtained samples (see Fig. 1a and b) is probably due to the presence of a high number of organic compounds in the matrix. Indeed, TFA is capable to catalyse dehydration reaction (Zhao et al., 2007). The morphological analysis performed on the cast film (Fig. 2) clearly shows a regular distribution of microfibers within the matrix and the absence of any stratification of the fibres.

Fig. 2 shows the SEM images collected from surface (a) and cross section (b) of dried rice straw bioplastic specimen. The SEM images demonstrate a compact and uniform surface structure, characterized by the presence of small inorganic aggregates, (see Fig. 2a magnification 450×, where small aggregates are evident). Cross section images of rice straw bioplastic (Fig. 2b) show the cellulose fibres. Table 1 presents the results of SEM analysis performed on spectrum 1, 2, 3 and 4 (see Fig. 2). Semi-quantitative analysis shows that Si is observed in the granular area. This can be due to the high amorphous silica content in the rice straw (Sangon et al., 2018).

The 2D-XRD image of the bioplastic shows the presence of a very broad peak at $2\theta = 20°$, which confirmed the amorphous nature of the organic material present in the rice straw bio-film (see Fig. 3a). Moreover, 2D-XRD analysis shows a sharp crystalline peak at approximately 24°, which can be attributed to calcium oxalate monohydrate (whewellite, $Ca(C_2O_4)\cdot(H_2O)$), in agreement with EDXS results. Its presence may be due to the formation of crystals in specialized cells during the calcification process, named crystal idioblast (Franceschi and Horner, 1980). Other studies confirmed the presence of whewellite in cell walls (Malainine et al., 2003) and plants with high amount of cellulose (Mannai et al., 2016).

The normalized IR spectra of rice straw and bioplastic (Fig. 3b) presents the typical bands associated with cellulose, namely, O-H stretching mode at 3335 cm^{-1}, C-H stretching mode at 2898 cm^{-1}, adsorbed water at 1641 cm^{-1}, and C-O stretching mode at

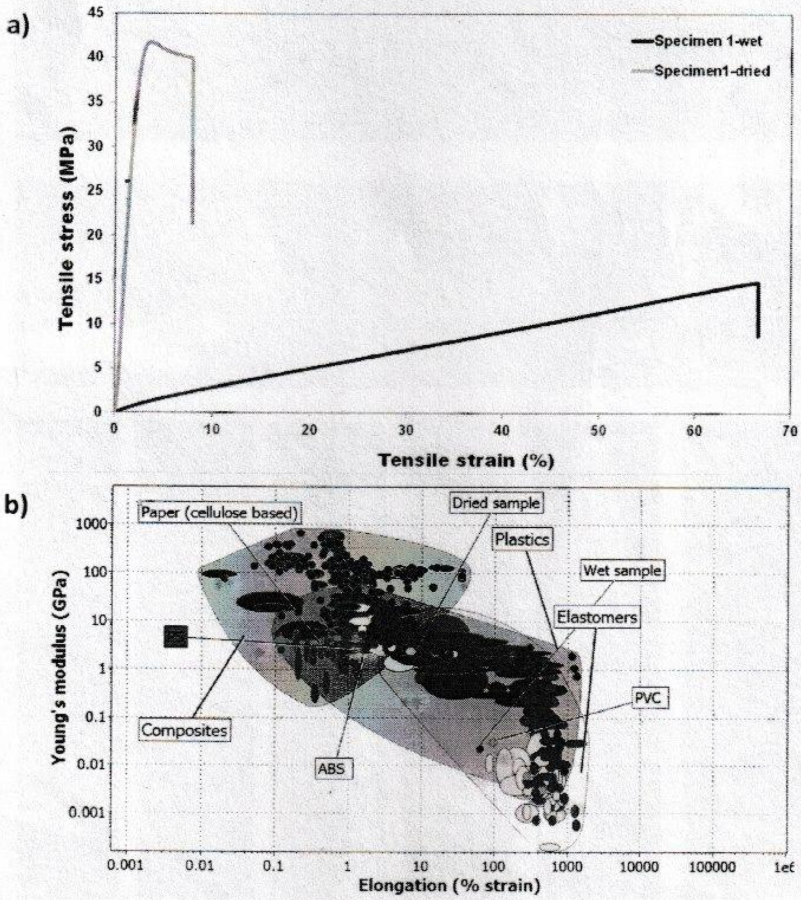


Fig. 4. a) Representative tensile stress-strain curves of dried and wet dumbbells of bioplastics; b) Young modulus vs elongation (%) of the plastics using CES Selector software (Granta, 2016).

Table 3
Stress strain analysis of dried and wet dumbbells. The results are reported as the mean ± S.D. of three replicates.

Samples	Dumbbell	E (MPa)	σ_b (MPa)	ε_b (%)	σ_y (MPa)
Flat	Dried	2330 ± 220	43 ± 3	6.1 ± 1.6	45 ± 3
Flat	Wet	23 ± 4	11 ± 3	63 ± 3	–

S.D. – standard deviation.

1030 cm^{-1} (Garside and Wyeth, 2003). The peak at 798 cm^{-1} is related to the presence of silica in raw rice straw. Moreover, the peak at 1428 cm^{-1} presents the C-H deformation both for lignin and cellulose, and the peak at 1160 cm^{-1} can be ascribed the C-O-C asymmetric stretching of cellulose, hemicellulose and lignin. (Agustin et al., 2014).

Bioplastic from rice straw exhibits excellent water absorption properties: Table 2 reports mass, thickness and width of dried samples and of the same samples after water immersion. As expected, higher values are obtained for wet dumbbells. Nevertheless, volumetric expansion do not cause cracking or breaking of the specimens.

3.2. Tensile properties

Tensile tests provide important information regarding the mechanical features of a material, including elastic and ultimate properties. The results of tensile tests are reported in terms of nominal stress vs nominal strain relationship for both dried and wet specimens. Other mechanical parameters such as elastic modulus as well as elongation at break are considered. Fig. 4a shows tensile stress-strain curves of dried and wet dumbbells.

Results of tensile test performed on dried and wet dumbbells prepared from rice straw bioplastic are reported in Table 3.

As expected, a stiffer response is found for the dried specimen, which presents a higher strength and relatively lower strain at break, approximately 6%. By contrast, wet samples revealed a lower stress and strength, but a significantly higher strain at break, approximately 63%. These results suggest increased compliance and ductility due to the plasticizing effect of water. Such an important effect may be employed to enhance chain mobility in a

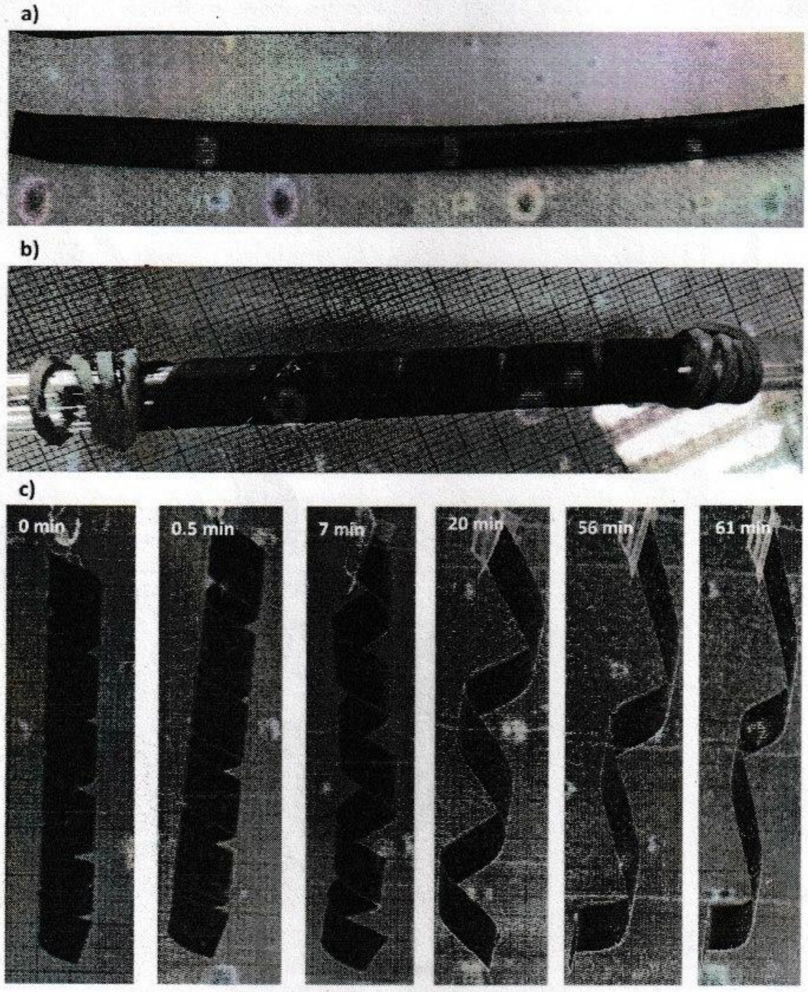


Fig. 5. Flat shaped biomaterial: a) initial shape of a specimen prepared as flat; b) temporary helical shape; c) water-driven recovery test from helical to flat shape.

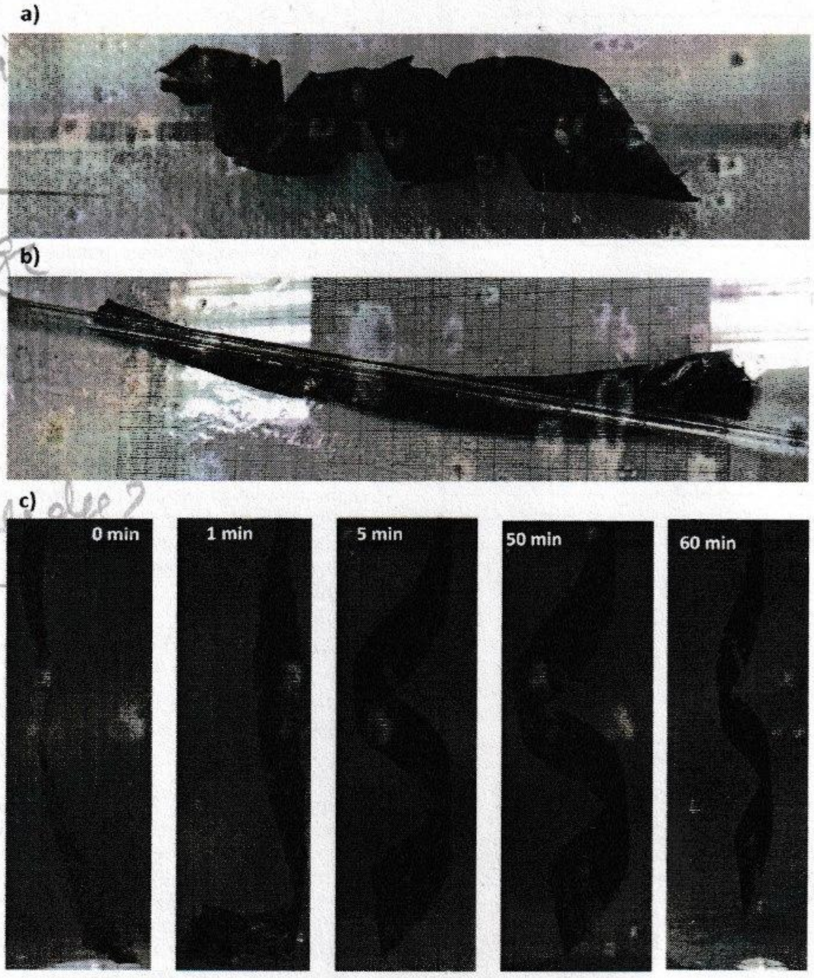


Fig. 6. Helical shaped biomaterial: a) initial shape of a specimen prepared as helical; b) temporary flat shape; c) water-driven recovery test from flat to helical shape.

potentially environmentally – or humidity –driven shape memory effect as discussed later.

Comparison of the mechanical properties of rice straw bioplastic with other material categories is shown in Fig. 4b. Being a cellulose-based bioplastic, it is not surprising that the mechanical properties of the produced biomaterial in its dry state, are comparable to that of cellulose. On the other hand, it is very interesting to notice that the mechanical behaviour of the produced biomaterial in its dry state, is comparable to polystyrene (PS) while cast bioplastic in wet state is similar to plasticized PVC. This demonstrates the high mechanical performance of the newly obtained bioplastic both in dry and wet status. Depending on the environmental humidity, the material showed a dual mechanical behaviour that can be exploited to obtain shrink films and sheet or to drive shape memory effect.

3.3. Shape memory test results

Shape memory behaviour of rice straw bioplastic was investigated by deformation-recovery cycle using water diffusion as a triggering stimulus for the shape transformation and exploring the material performance under these conditions. The specimen appearance during the shape memory cycles are reported in Figs. 5 and 6 for the materials prepared as flat and helicoidal specimens, respectively.

The so-called "programming step", i.e., fixing the temporary shape of the specimen, is here depicted, by showing the pristine, "permanent" shape (Figs. 5a and 6a) along with the temporary configuration (Figs. 5b and 6b). The temporary shape was set starting from dried flat (Fig. 5a) and helical (6a) specimens, immersing them in water and deforming in the temporary configuration, which was maintained until the specimens were completely dried (Figs. 5b and 6b). Pictures of Fig. 6 also show that the bioplastic formed on burette surface is not as uniform as that obtained by casting deposition (Fig. 5). Moreover, the figures illustrate the recovery process, which was activated by suspending the specimen in a beaker containing distilled water. When they are immersed in water, water diffuses into the polymer samples and acts as a plasticizer, resulting in shape recovery. The pictures of Figs. 5c and 6c represent various instants of the recovery sequence of the originally flat and helical specimens, respectively.

Based on the sequence pictures of the water-driven test, a quantitative description of the recovery process as a function of

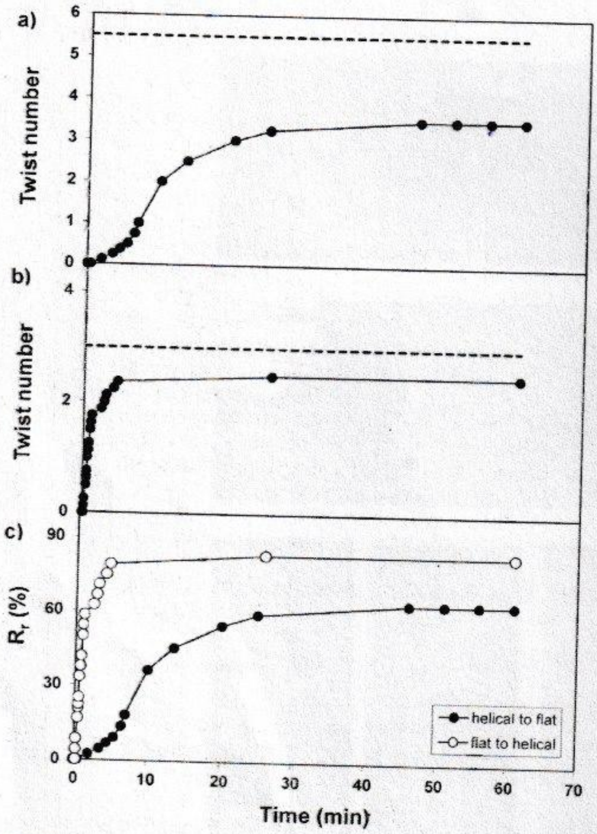


Fig. 7. Cycle number changes during the water-driven test for the a) flat-helical specimen; b) helical-flat specimen. The dashed lines represent the angle of rotation applied when deforming the specimen, as representative of the full recovery condition. c) shape recovery of flat and helical specimens.

time was attempted, and it is represented in Fig. 7a and b for the originally flat and helical specimens, respectively. In both figures, the shape variation is represented in terms of number of twist that the specimen undergoes during recovery. Each twist represents a 360° rotation of the free specimen extremity. Fig. 7a shows the gradual recovery process from the helical to the flat one, and it represents the number of twists applied to the material when providing it in the temporary configuration as a dashed line; this curve also represents the number of twists that the deformed specimen should undergoes to display full recovery. During the first 25–40 min shape changing is faster, and then, it remains almost constant with time.

Similarly, Fig. 7b represents the recovery process from the flat to helical shape in terms of twists as a function of time, showing a fast shape evolution in the first 10 min and a higher degree of recovery with respect to the originally flat specimen of Fig. 7a.

To properly compare the different shape memory response of the two specimens, the raw data in Fig. 7a and b were used to evaluate a shape recovery parameter, termed recovery rate (R_r). R_r is a term traditionally employed in literature to express the overall shape memory capabilities in terms of a percentage of recovery, and it is usually defined under tensile or compressive uniaxial condition (Behl and Lendlein, 2007). Here the typical recovery rate equation is adapted to the twisting/untwisting case as expressed in Eq. (1):

$$R_r = \frac{\Delta\gamma}{\gamma_{initial}} * 100 \quad (1)$$

where $\Delta\gamma$ is the rotation of the specimen free extremity during the recovery process, while $\gamma_{initial}$ is the degree of rotation provided during the programming step.

R_r changed as a function of time (Fig. 7c) for both specimens, with a shape recovery ratio for the material between 64 and 83%, depending on the specimen and the different recovery speeds of the two specimens. Our results suggest a higher shape recovery of the originally helical specimen, possibly due to its morphology: the originally helical specimen has a different thickness and is generally thinner than the originally flat specimen, thus allowing a faster absorption of water. These R_r data are lower compared to R_r values reported by other studies (Bai et al., 2016), where cellulose acetate was shown to almost fully recover the original shape. The measured lower recovery ratio may be ascribed to the low temperature used in our water activation tests. However, these data are comparable with those reported for other types of polymers (Lendlein and Kelch, 2002). These results suggest that the proposed bioplastic demonstrates promising dual shape memory shifting capability.

To further investigate the effect of water absorption on both specimens, absorption test was carried out. After immersing in Milli-Q water at room temperature, water absorption was calculated using Eq. (2):

$$water\ absorption = \frac{(m_2 - m_1)}{m_1} * 100 \quad (2)$$

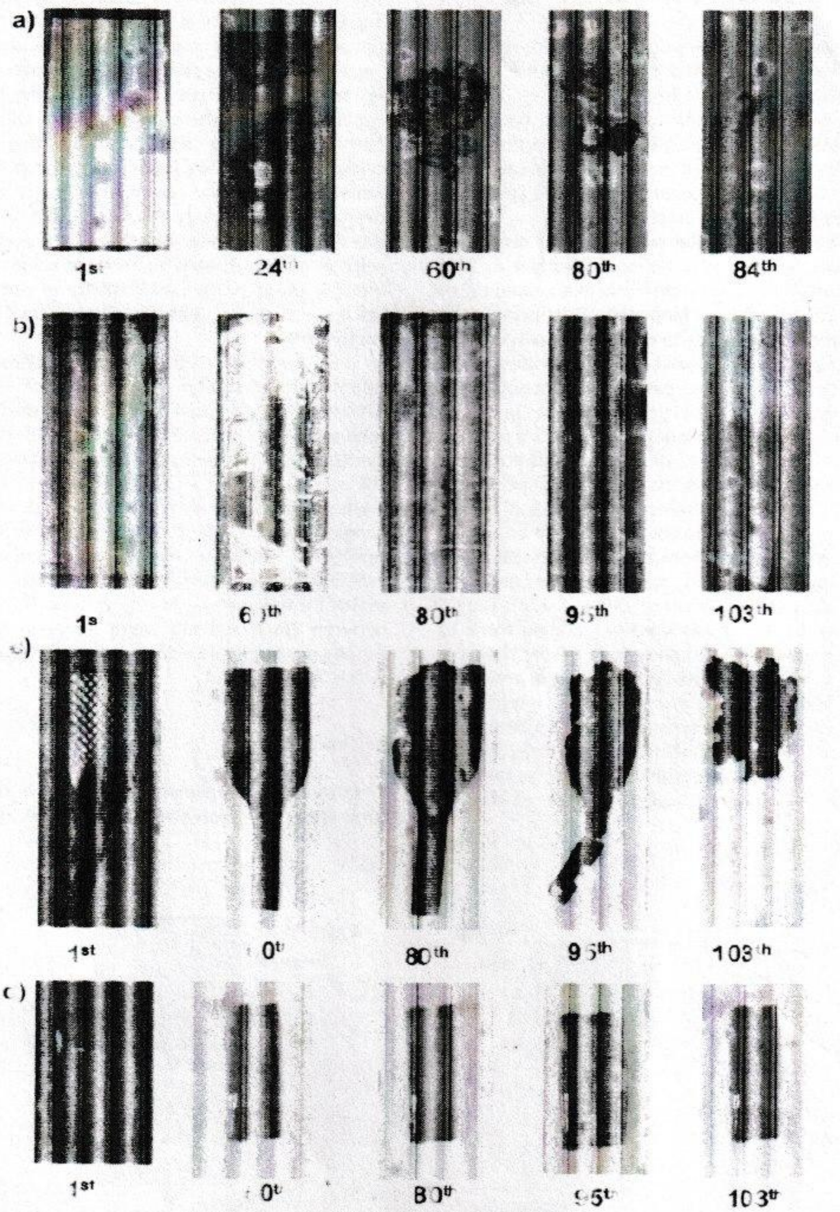
where m_2 and m_1 stand for the specimen mass after and before water absorption, respectively.

The originally helical shape shows higher water absorption percentage compared to the originally flat one, with values of approximately 89% and 61%, respectively. Thus, justifying the faster and more effective process. As reported in the literature, there is a linear relationship between the increase of glass transition temperature and decrease of the water absorption rate of shape memory polymers (Huang et al., 2005). Based on our preliminary results, agro-food packaging may be proposed as a possible application field of this type of biomaterial. Further studies will be necessary to investigate other parameters, which will improve the shape memory effect of this type of biomaterials.

3.4. Material mass loss test

Degradation phenomenon results from the action of naturally occurring microorganisms, such as bacteria, fungi, and algae. During this process, the bioplastics must biodegrade in specific environments such as soil, compost, or marine environments. Carbon dioxide, water and biomass are biodegradation products under aerobic conditions, while under anaerobic conditions, methane, water and biomass are produced.

Cellulose-based bioplastic in distilled water showed a neutral pH (6.02) close to tap water (6.12), highlighting that this biomaterial does not leach any toxic component related to TFA. On the first day, all materials, except for PET plastic, have increased their mass. This phenomenon is probably due to the absorption of soil moisture by the materials. As expected, degradation was observed first for cellulose paper. Specifically, it was broken into two parts after 24 days, and its mass became almost half after 10 days, decreasing from 0.23 g to 0.12 g. Mater-bi compostable plastic showed a great ability to absorb moisture, doubling its mass after some days. From the 50[th] day, its mass remained almost constant, and it appeared to be thinner from a qualitative point of view. PET



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3.5. Industrial and environmental aspects of the technology

The main advantage of the recovery and reuse of rice straw is its availability. Indeed, this type of waste does not require complex management strategies to separate it from other wastes. The proposed technology is based on the use of an already assessed instrument: the Naviglio extractor, which is available on the market and currently successfully employed in the food, cosmetic and pharmaceutical industries. The novelty of the proposed approach is the application of this device to pre-treat a waste.

Fig. 9 depicts the synthesis of the bioplastics. As reported in other studies on its use for material dissolution (Dong et al., 2009; Quartarone et al., 2014), TFA can be completely evaporated off and recovered for the synthesis of new bioplastic. This increases the process efficiency and reduces its cost. In terms of the sustainability of the new proposed process, the evaluation of embodied energy (MJ/kg) and CO_2 footprint (kg/kg) for primary production of bioplastic vs other material categories was performed (see Fig. 10). The embodied energy (EE) is the energy required to produce materials from ores and feedstock. It includes direct and indirect energy consumed during the production process of 1 kg of a specific material. The CO_2 footprint (CF) represents the equivalent mass of greenhouse gases in kg, released into the atmosphere as a consequence of the production of 1 kg of the material. The first parameter represents energy and the second represents the emissions, involved in material synthesis (Bontempi, 2017a, 2017b). The CES Selector software (Granta, 2016) was used to evaluate these parameters. Generally, materials that require high EE for their synthesis (Bontempi, 2017a) are produced in lower quantities in respect to materials requiring low EE. In general, high energies and emissions involved in the synthesis correspond to production of the corresponding materials in reduced quantities. Therefore EE and CF, which are material-dependent constant values, are suitable parameters for comparison of different materials, either in terms of sustainability or economic advantages in their production. Low EE and CF may enhance a high market demand. Obviously, the functional properties of materials are fundamental. As we can see in Fig. 10, EE of the proposed materials ranges from 16.84 to 22.0 MJ/kg, and CF ranges from 1.01 to 1.62 kg/kg. These results are comparable to those of thermoplastic starch and cellulose-based paper. Both EE and CF are high for synthesizing plastic from fossil resources. The SUB-RAW index for rice straw bioplastic compared to other materials was calculated according to the previously described methodology (Bontempi, 2017b). SUB-RAW index is a quantifiable parameter that allows the comparison between two selected materials based on the mean values of EE and CF. When the index is positive, the sustainability of the material proposed as reference is higher than the conventional materials selected for comparison.

Table 4 lists the SUB-RAW indices calculated for the new biofilm with respect to the other conventional bioplastics. These data indicate that, in terms of energy and emissions involved in material production, the aforementioned materials derived from rice straw can be considered comparable to a bioplastic obtained from starch (EE = 25.25 MJ/kg and CF = 19.88 MJ/kg). The new bioplastic material is much more sustainable and industrially convenient compared with other plastic materials such as PP and Polylactic Acid (PLA). Therefore, environmental impact can be reduced by substituting the aforementioned materials with the proposed bioplastic from rice straw. Finally, considering the inverse correlation between EE/CF and the world materials production (Bontempi, 2017b), an industrial advantage in the synthesis of these materials is also expected.

4. Conclusions

In this study, application of rice straw, classified as an agricultural waste, for bioplastic production is reported. Rice straw, a

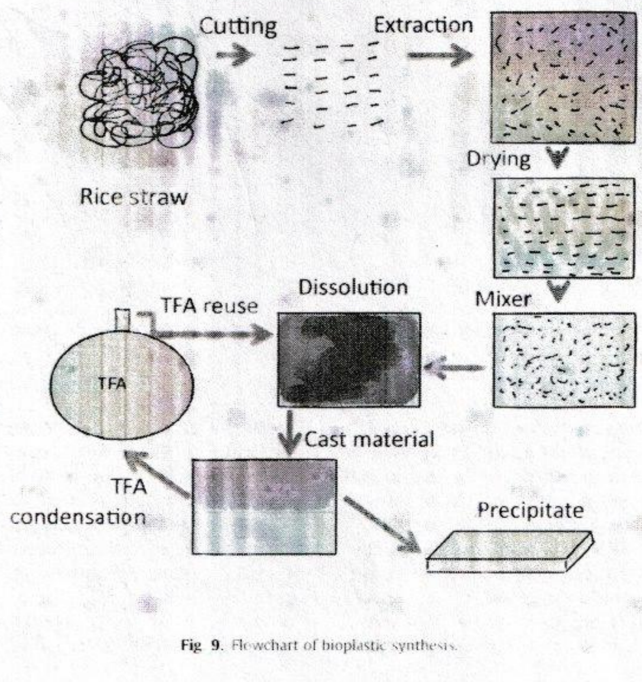
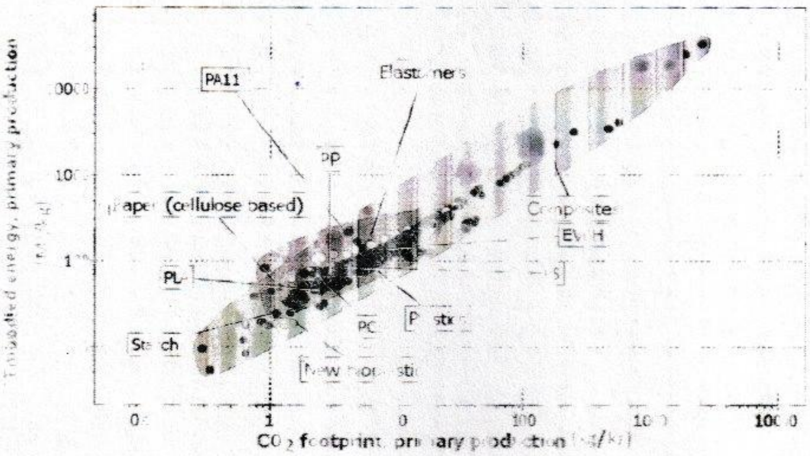


Fig. 9. Flowchart of bioplastic synthesis.



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