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Development of an acrylic emulsion paint added with cellulosic dispersion treated with an electron beam accelerator

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ABSTRACT

Paint is a prevalent type of surface of coating well known to most people. It is an easy-to-apply product, with excellent physical and chemical resistance and a wide range of applications. The function of a paint is to protect and beautify amongst other properties. Paints are formed using five components: resin, pigments, fillers, solvents and additives. This work deals with a specific type of paint, composed of a water-based acrylic emulsion, whose film formation is by coalescence and used to coat walls and other surfaces. The aim of this innovative research was to evaluate the effect of the addition of cellulosic dispersion of waste paper tissues treated by electron beam irradiation to an acrylic emulsion-based paint. The methodology used was a case study carried out at the Nuclear and Energy Research Institute that consisted of adding aqueous dispersions of cellulosic wipes with mass concentrations varying from 0.1% to 10% in deionized water, and treated by electron beam processing with absorbed doses from 5 kGy to 50 kGy. The results obtained followed the parameters of the Abrafati Sectorial Quality Program. The main sample parameters analyzed were specific density, which reached an average of 1.35 g/cm^3; and covering power, whose value of 93.55% was above the specified limit (minimum of 90%). Among other results obtained, improvements in appearance, applicability and functionality were significant. It was concluded that this research constitutes an incremental improvement to the acrylic emulsion paints segment, and environmental sustainability, through the reuse of cellulosic tissue residues.

Keywords: **acrylic emulsion, architectural paint, ionizing radiation, circular economy, sustainability.**

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1. INTRODUCTION

The paint market today is extensive, mainly due to the variety of paints and their diverse applications. Paint can be applied to numerous substrates such as metal, wood, plastics, glass and masonry amongst others in order to fulfill various coating purposes. The present work will limit itself to polymeric paint coatings.

According to Nascimento[4,29], paint is generally composed of resin, pigments, fillers, solvents and additives. The main component of paint is resin, the element responsible for forming the protective layer of the substrate and anchoring the other layers of paint. Pigments are elements that confer color and opacity, thereby masking the substrate. Fillers are mineral elements responsible for giving body, physical and chemical resistance to the formed film. The penultimate element, the solvent, is inserted into the paint to solubilize the paint resin and enable its application. Finally, additives are substances used in small amounts to confer specific properties to the paint.

Paints in relation to the film formation mechanism are classified into[19, 29].

Thermoset paints - those paints whose film does not change when applied and polymerized. These are divided into: a) Paints dried by air oxidation - polymerization (film curing) occurs by oxidation of the paint film, as with oil paints and synthetic enamels. The main applications of this type of paint is to paint doors, windows, railings or furniture by brush or by spray gun; b) Heat cured paints - the paint film is formed by the action of heat, resulting in crosslinking (polymer reticulation),. These paints are also known as oven enamel or oven polyester enamel. In general, they are applied to household appliances, bicycles, motorcycles and steel cabinets, among others; c) Catalyzed or two-component paints - polymerization (film curing/film formation) occurs through the reaction between two components, polymer “A” and curing agent/catalyst “B”, supplied in two packages. These are known as epoxy and polyurethane paints and are applied to buses, planes, oil and gas tanks, water pipes, oil platforms and hydroelectric gates, among others [27,28, 29];

Thermoplastic paints - those paints whose film is formed by the evaporation of the solvent. With this type of paint, even after its application and polymerization, the film can be removed by the action of a solvent. These are the so-called “nitrocellulose lacquers” and “acrylic lacquers”, used to paint wooden furniture and in the automotive refinishing segment [27,28, 29].

Coalescence curing paint - the third and last type of film formation mechanism is the object of this research. These are paints that form a film by coalescence, that is, the formation of the film occurs by fusion of the particles after the evaporation of the solvent. Once the film has formed, it is no longer possible to solubilize the paint layer. These paints are used to cover walls and buildings, known in the market as PVA paint and water-based acrylic emulsion paint [4, 19, 21, 29].

The paint manufacturing process, in general, involves a set of complex discrete operations as follows: separation and weighing of raw materials, dispersion, grinding, completion, quality control, filtration, packaging, storage and distribution. In this study, the approach is specific to water-based acrylic emulsion paints, also known as real estate paints. However, it is worth mentioning that there are other types of architectural paints [13, 33], which includes the following: PVA latex paints, acrylic paints, vinyl-acrylic paints and textures. Depending on the environment in which these paints are applied, internal or external, at least one of the following grades must be chosen: economic, standard or premium [13].

Cellulosic paper wipes - According to the Brazilian Technical Association of Cellulose and Paper [14], and IBA [24] cellulosic wipes represent one of the products in the manufacturing line of cellulosic materials [36]. Known as “tissue paper”, moistened cellulosic paper tissues are made of low weight sheets or rolls, used for personal hygiene and domestic cleaning, toilet paper, handkerchiefs, paper towels and napkins. In addition to virgin fibers, they typically have good quality recycled shavings included in their composition. These products are generally used by children and adults for cleaning and removing dirt, impurities and cosmetics, including waterproof products, with no need for rinsing. Another example are baby wipes, which are, according to one of the major manufacturers, hypoallergenic for sensitive skin and dermatologically tested, making them perfect for all skin types. Basically, they are composed of sheets of cellulosic paper moistened with purified water. The main characteristics that these products must have to act as cellulosic tissue paper are weight, absorptivity, softness, specific volume, resistance to traction, whiteness, elongation and appearance.

Ionizing radiation - Electron beam technology (ionizing radiation) is widely used in the industrial processing of materials, and can function in the crosslinking and curing process of polymers. . This technology is used in the manufacture and modification of many products, such as rubber, paints and adhesives, sterilization of medical-surgical devices, preservation and disinfection of food, among other applications. Among its advantages are controlled application, operating flexibility, high processing capacity, reasonable cost, clean technology designation, among others [21, 29, 31, 15].

Electron accelerators are divided into low (150 keV to 300 keV), medium (300 keV to 5 MeV) and high energies (5 MeV to 10 MeV). Low and medium energy accelerators are used in the treatment of combustion gases (removal of SO_2 and NO_X), in the processing of composite materials, nanocomposites and biodegradable composites, in the irradiation of carbon fibers and heat-shrink tubes, in grafting ion exchange membranes for application in fuel and hydrogen cells, in the production of nanoparticles in electrocatalysts and in obtaining natural polymers, among other applications [29, 30, 15, 31, 1].

Applications of high-energy accelerators include radiosterilization of medical-surgical products, the induction of color in gemstones, the treatment of industrial and domestic effluents and sludge, the preservation and disinfestation of food and agricultural products, the irradiation of lignocellulosic material as a pre-treatment to produce biofuel ethanol, decontamination of pesticide containers, solid waste remediation, the removal of organic compounds in wastewater, and the treatment of effluents in oil production units, among others [29, 31, 5, 1].

It is noteworthy that radiation technology is an excellent instrument used to reduce the microbial burden in organic materials. Studies demonstrate the efficiency of using electron beam technology as a tool to decontaminate or eliminate microorganisms from organic materials, mainly from paper and cellulosic residues. In addition, this technology can be applied to the functionalization of cellulose fibers to improve their interaction with an acrylic emulsion, contributing to the optimal performance of paint.

Cellulose is a natural organic polymer and there are several studies that describe the use of ionizing radiation to accelerate biodegradability. In this way, electron beam technology used in aqueous cellulosic dispersions contributes to the functionality of the water-based acrylic emulsion under study, improving its flexibility, processability, applicability, physical-chemical resistance and covering power, with the added advantage of using a clean technology [1, 18, 20, 21, 26].

2. MATERIALS AND METHODS

2.1. Purpose of the study

The objective of this study was to improve the properties of a premium water-based acrylic emulsion paint by the addition of cellulosic industrial waste from moistened paper tissues in an aqueous dispersion in concentrations by mass of 0% (control), 0.1%, 0.2%; 0.3%; 0.4%, 5% and 10%.

The cellulosic dispersion is followed by treatment with an electron beam, at absorbed doses of 5 kGy and 50 kGy. This research sought to develop a new paint based on an incremental innovation of acrylic emulsion in accordance with the requirements established by the Sectorial Quality Program (PSQ), defined and practiced by the Brazilian Association of Paint Manufacturers [13]. Additionally, this work provides a new destination for cellulosic waste from moistened tissue paper, making it possible to increase profitability, reduce costs and reduce environmental impact. Another relevant contribution is that the project can be registered as positive evidence of successful application of clean radiation technology in the areas of architectural paints and wet wipes.

2.2. Justification of the study

In this study, a premium water-based acrylic emulsion was selected, allowing for the application to both indoor and outdoor environments. Furthermore, in the industrial segment of cellulosic wet wipes, a major problem is how to handle the large amount of waste from the production process. The amounts discarded are considerable and there is very little reuse of these materials in the production process due to the complexity of this waste, resulting in their deposit into industrial landfills [30,17]. In the case of architectural paints, a factor that should be highlighted is the degree of biodegradability .

The addition of raw materials that contribute to this end make these paints more sustainable, as is the case with the addition of plant fibers. Based on this information, it is worth remembering that a new green wave of thought focused on sustainability is influencing the European and American continents, contributing to the concept of a circular economy. According to these principles, treating waste, keeping it within the production cycle and providing it with a new application in the manufacturing cycle contributes immensely to sustainability and minimizes the consumption of natural resources[16].

The accelerators used in industrial processes produce electrons in the energy range of 0.1 to 10 MeV. In the application of radiation dosage, this study employed the Industrial Electron Accelerator of 1.5 MeV and 25 mA present at the Radiation Technology Center of IPEN and manufactured by Radiation Dynamics Inc., .Samples were irradiated in batches using pyrex glass containers. The irradiation parameters were 4.0 mm maximum thickness of the sample, 112 cm (94.1%) width in the scanning system and 6.72 meters/minute in transporting the containers.

2.3. Sampling

The processes are based on NBR [5,7]

2.3.1 **Preparation of cellulosic dispersions** – Samples of aqueous cellulosic dispersions were prepared in deionized water, at concentrations by mass of cellulosic tissue residues of 0.1%, 0.2%; 0.3%; 0.4%, 5% and 10%

2.3.2 **Treatment of cellulosic dispersions with ionizing radiation** - samples of aqueous cellulosic paper tissue dispersions were treated with ionizing radiation at absorbed doses of 5 kGy and 50 kGy .

2.3.3 **Preparation of paint samples** – Water-based acrylic emulsion paints were prepared by adding cellulosic aqueous dispersions in deionized water, previously treated with ionizing radiation, to a final level of 0% (control), 0.1%; 0.2%; 0.3%; 0.4%; 5% or 10% by weight.

The paints were splitted into samples, based on the additive composition and sent for analysis, in accordance with the requirements of the Sectorial Quality Program (PSQ), of the Association of Paint and Varnish Manufacturers[13].

2.3.4 Preparation of paint samples for analysis and application on a ceramic substrate

In this final phase, the paint samples were sent for laboratory analysis and paint application, in accordance with the requirements of Abrafati's PSQ, to the Paint and Varnish Laboratories of the Senai Mario Amato School, and to the Sherwin Williams Paint factory.

2.3.5 Standards used to determine the tests

The standards of the Brazilian Association of Technical Standards [6, 7, 8, 9, 10, 11, 12] and American Society for Testing and Materials [2, 3], used in the analysis of acrylic emulsion-based paint with added electron beam-treated cellulosic dispersions are shown in Table 1.

Table 1 - Tests carried out with paint based on acrylic emulsion with added electron beam-treated cellulosic dispersions, according to Abrafati's PSQ requirements

Item	Test	Methodology
1	Paints for civil construction - Method for evaluating the performance of paints for non-industrial buildings - Determination of wet abrasion resistance without abrasive paste	NBR 15078:2006
2	Determination of dry paint hiding power and theoretical yield.	NBR 14942:2022
3	Determining the hiding power of wet paint	NBR 14943:2018
4	Determination of color and color difference by instrumental measurement.	NBR 15077: 2004
5	Resistance to UV radiation and water condensation by accelerated test.	NBR 15380:2015
6	Standard Practice for Xenon-Arc Exposure of Plastics Intended for Outdoor Applications	ASTM D2565-16
7	Standard Practice for Operating Xenon Arc Lamp Apparatus for Exposure of Materials	ASTM G155-21
8	Determination of the specific mass	NBR 5829:2014

3. RESULTS AND DISCUSSION

3.1. Moisture test

Testing was performed to determine the moisture content of the sample supplied by the partner company, the manufacturer of cellulosic wet wipes. A moisture content of approximately 70.41% was obtained. An electronic analytical scale from Mettler Toledo, with a maximum capacity of 500 g and precise to 0.01 g, and an oven manufactured by Fanem, with a maximum temperature of 300ºC, were used for this analysis.

3.2. Microbiological test

The microbial burden of the cellulosic dispersion was determined to avoid possible contamination of the paints following addition of the cellulosic wipes. The results of the microbiological analysis are presented in Table 2. The methodologies used in the determination of possible microorganisms were a surface swab for molds and yeasts[1] and a surface swab for total microorganism count[2].

Table 2 - Results of microbiological tests of cellulosic dispersion

Samples	**Bacteria**		**Fungi**	
	CFU/100 cm^3	**Result**	CFU/100 cm^3	**Result**
1	0	Conform	0	Conform
2	0	Conform	0	Conform

The cellulosic dispersions were free of fungal or bacterial contamination, with no risk when added to premium acrylic emulsion-based paint.

3.3. Test of wet hiding power of paint

Tests were carried out as determined by Abrafati's PSQ [13]. This standard establishes that a premium acrylic emulsion type paint must have a wet covering power ≥ 90%. The aqueous cellulosic dispersions, up to concentrations of 10% by mass and irradiated by electron beam at absorbed doses of 5 kGy and 50 kGy, occurred in 2 (two) phases:

a) Phase 1 - cellulosic waste concentrations of 0.1%, 0.2%, 0.3% and 0.4% by mass;

b) Phase 2 - cellulosic waste concentrations of 5% and 10% by mass.

The results of the wet paint hiding power tests, in Phases 1 and 2, are shown in Table 3.

[1] **Mold and yeast surface swab -** *POPMB - UNI174, AOACO Official Method 997.002 - POPMB - UNI184. ISO 21527-1:2008 Microbiology of food and animal feeding stuffs – Horizontal method for the enumeration of yeasts and molds – Parte 1 Colony count technique in products with waters activity greater than 0,95.*

[2] **Total Microorganism Count Surface Swab** - *POPMB - UNI057, AOACO 2000. Official Method 990.12 – POPMB - UNI178. ISO 4833:2003 - Microbiology of food and animal feeding stuffs – Horizontal method for the enumeration of microorganisms – Colony count technique at 30ºC.*

Table 3 - Covering power of wet paint, according to NBR 14943:2018 [9]

Analyzed Samples	CP 1	CP 2	CP 3	Results	specification
Phase 1					
Control (*)	95,88	96,48	96,73	**96,36**	≥ 90 %
FCN 0,1% RC 5 kGy	96,51	96,17	97,24	**96,64**	≥ 90 %
FCN 0,1% RC 50 kGy	96,44	96,03	96,07	**96,18**	≥ 90 %
FCN 0,2% RC 5 kGy	96,14	96,15	97,44	**96,58**	≥ 90 %
FCN 0,2% RC 50 kGy	95,55	96,76	96,66	**96,32**	≥ 90 %
FCN 0,3% RC 5 kGy	95,58	96,73	96,50	**96,60**	≥ 90 %
FCN 0,3% RC 50 kGy	95,84	95,87	95,80	**95,84**	≥ 90 %
FCN 0,4% RC 5 kGy (**)	-----	-----	-----	-----	not analyzed
FCN 0,4% RC 50 kGy	94,74	94,86	95,84	**95,15**	≥ 90 %
Phase 2					
Control (*)	95,69	95,43	96,24	**95,79**	≥ 90 %
FCN 5% RC 5 kGy	94,20	94,41	88,93	**92,51**	≥ 90 %
FCN 5% RC 50 kGy	95,65	94,04	90,76	**93,48**	≥ 90 %
FCN 10% RC 5 kGy	93,88	91,23	94,79	**93,30**	≥ 90 %
FCN 10% RC 50 kGy	93,25	96,06	95,35	**94,55**	≥ 90 %

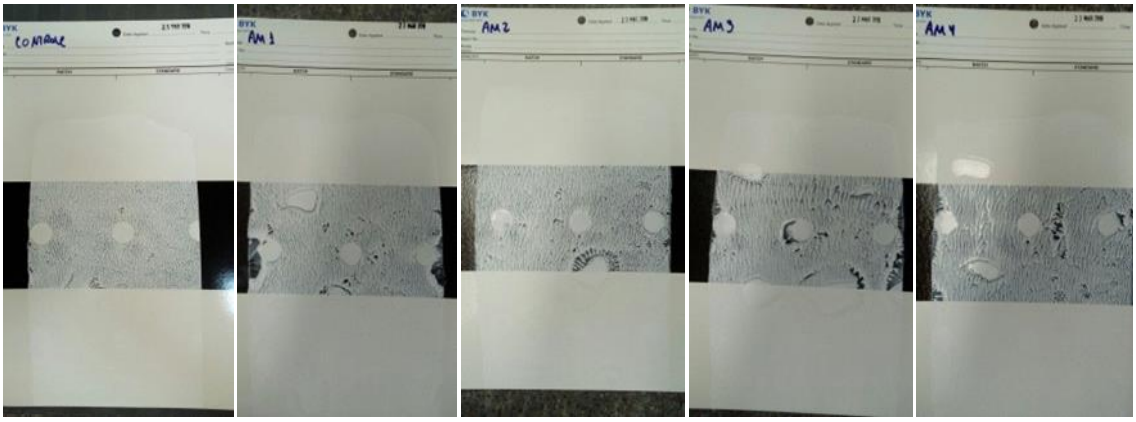
CP - Specimen of samples in triplicate.

(*) Premium acrylic emulsion based paint in white color, without addition of cellulosic residue and without radiation treatment.

(**) Sample FCN 0.4% RC 5 kGy presented a problem in the preparation and was disregarded for analysis.

Figure 1 shows the results of the wet paint hiding power test. It is observed (in white color) that there is practically no difference between the control sample (without added irradiated aqueous cellulosic dispersion) and the samples of premium acrylic emulsion-based paints with added cellulosic dispersion.

Figure 1 - *Results of wet paint covering power tests: (a) control; (b) FCN 5% RC 5 kGy (Am1); (c) FCN 5% RC 50 kGy (Am2); (d) FCN 10% RC 5 kGy (Am3); and (e) FCN 10% RC 50 kGy (Am4)*



(a) (b) (c) (d) (e)

The factor that most of the features to a paint present a higher or lower covering power is the pigment, especially white. The most used is rutile titanium dioxide. One of the factors for this phenomenon to occur is its average refractive index, which is around 2.73 for the rutile titanium dioxide and 2.55 for the anatase type [19]

Mineral fillers are widely used to replace part of the white titanium dioxide pigment, which is very expensive. These generally have a covering power of less than 1.7, such as barium sulfate with 1.64, calcium carbonate (1.57) and calcium sulfate (1.59), among others [19]

In the case of this research, the incremental innovation has been chosen by using an aqueous dispersion formed by cellulose fibers, treated with ionizing radiation (electron beam), as an additive for water-based architectural paints, replacing part of the fillers [22,23, 32, 34,35].

This replacement did not interfere with the covering power, both wet and dry. Possibly, due to the average refractive index of the cellulose fibers, which is around 1.6. It is very close to the mineral fillers that are used for providing protection and contribute to the covering power, and to the cost reduction.

The calculation of the wet covering power described in item 3.2 and the results presented in Table 3 follow the standards determined by NBR 14943:2018 [10], whose minimum expected limit for a polymeric coating is 90%, to be used in the real estate and masonry area.

It can be noted in Table 3 that all samples of this research reached and even exceeded the minimum limit of the coverage power. However, a practical way to observe this characteristic is to apply the paint on a black and white card, with a single coat of paint.

The wet coverage power is one of the characteristics most perceived and required by painters. Through this test, the painter's vision can be simulated in relation to the behavior of the paint when applying it on a substrate, which one wants to protect.

Figure 1 demonstrates the appearance of the coating on the substrate with the film still wet. The paint samples have showed results of the wet hiding power, such as the control sample, once again confirming the fulfillment of this requirement.

Thus, after analyzing the results presented in Table 3, it is observed that all samples of premium acrylic emulsion-based paints with additives (white color) are within the technical specifications established in NBR 14943:2018 [10] ; that is, with a wet covering power ≥ 90%. We highlight the arithmetic mean values of the results of all samples in Phase 1 equal to 96.19% and in Phase 2 to 93.46%.

3.4. Specific mass determination test

The objective of this test is to verify if the raw materials were added according to the technical specifications of the paint. Accordingly, the results of additions of aqueous cellulosic dispersions in deionized water, treated by electron beam (5 kGy and 50 kGy), in the determination of the specific mass, in Phases 1 and 2, are presented in Table 4. Sample tests in triplicate were carried out at the Paint Laboratory of the Senai Mario Amato School, which is accredited by Inmetro, in compliance with the NBR 5829:2014 standard - Determination of the specific mass.

Table 4 - Determination of specific mass, according to NBR 5829:2014 [6]

Analyzed samples	**Results (g/cm^3)**	**Average specification (g/cm^3)**
Control (*)	**1,39**	1,20 a 1,50
Phase 1		
FCN 0,1% RC 5 kGy	**1,40**	1,20 a 1,50
FCN 0,1% RC 50 kGy	**1,39**	1,20 a 1,50
FCN 0,2% RC 5 kGy	**1,40**	1,20 a 1,50
FCN 0,2% RC 50 kGy	**1,39**	1,20 a 1,50
FCN 0,3% RC 5 kGy	**1,38**	1,20 a 1,50
FCN 0,3% RC 50 kGy	**1,37**	1,20 a 1,50
FCN 0,4% RC 5 k Gy(**)	-----	not analyzed
FCN 0,4% RC 50 kGy	**1,35**	1,20 a 1,50
Phase 2		
Control (*)	**1,39**	1,20 a 1,50
FCN 5% RC 5 kGy	**1,36**	1,20 a 1,50
FCN 5% RC 50 kGy	**1,35**	1,20 a 1,50
FCN 10% RC 5 kGy	**1,34**	1,20 a 1,50
FCN 10% RC 50 kGy	**1,36**	1,20 a 1,50

(*) Premium acrylic emulsion based paint without addition of cellulosic residue and without radiation treatment..

(**) Sample FCN 0.4% RC 5 kGy presented a problem in the preparation and was disregarded for analysis.

Following analysis of the results presented in Table 4, it is observed that all samples of premium acrylic emulsion-based paints with additives, are within the technical specifications established in the NBR 5829:2014 [6] standard; that is, in the range of 1.20 g/cm^3 to 1.50 g/cm^3. It is noteworthy that the addition of the treated aqueous cellulosic dispersion does not interfere with the test of the specific mass of the acrylic emulsion.

3.5. Preparation processo of masonry tiles

Acrylic emulsion paint added with added irradiated cellulosic dispersions was applied to masonry tiles, after sanding of the tiles with #100 and #120 sandpaper and dust cleaning, .

The results of applicability were excellent, especially when compared to the control sample, in view of the functionalization of cellulose promoted by ionizing radiation [25]. At this moment of the research, the samples are being exposed to natural weathering to check if there is any kind of degradation. No degradation has yet to be detected.

4. CONCLUSIONS

Due to the results achieved, it has been concluded that the research project presents an exceptional possibility of becoming a new and innovative business for the companies involved, as a result of the technology of irradiation of an aqueous cellulosic dispersion by electron beam.

The samples of paints based on premium acrylic emulsion with additives are within the technical specifications established in NBR 14943: 2018 [9], with wet covering power ≥ 90%. The arithmetic mean values of the results of all samples are equal to 96.19% (Phase 1) and 93.46% (Phase 2).

In addition, the samples are within the technical specifications established in the NBR 5829:2014 [6] standard, in the range between 1.20 g/cm^3 to 1.50 g/cm^3. It is noteworthy that the presence of the treated aqueous cellulosic dispersion does not interfere with the test of the specific mass of the acrylic emulsion.

The results related to applicability were extraordinary. The samples are currently being exposed to natural weathering to check if there is any kind of degradation, none has yet registered.

Thus, architectural paints based on an aqueous acrylic emulsion with added aqueous dispersions of irradiated cellulosic waste, up to 10% by volume meet the quality requirements of Abrafati's PSQ. In addition, there is a contribution to environmental sustainability and the good of the planet, by minimizing costs, natural resources and retaining industrial products within the production chain, as established by the Circular Economy technical cycle and the philosophy of Product Life Cycle Analysis.

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