
Process and cell for generating electric current

Specification

The invention relates to a process of generating an electric current according to the independent process claim, wherein melanin, melanin precursors, melanin derivatives or analogues in aqueous solution within a container absorb energy obtained by an energy source, whereby melanin, melanin precursors, melanin derivatives or analogues dissociate water molecules by dissipating the absorbed energy, whereby high energy electrons are transferred to a primary electron acceptor, and whereby melanin, melanin precursors, melanin derivatives or analogues (Melanins) are used the reverse reaction that comprises the union of atoms of hydrogen and oxygen generating water molecules and an electric current. In addition the invention relates to a cell for generating an electric current according to the independent product claim, wherein the cell comprises a container in order to permit at least electromagnetic radiations to pass through, melanin, its precursors, variants, derivatives, or synthetic or natural analogues,

which are mainly dissolved in an aqueous solution in the cell, an inner energy source within the container, a cathode and an anode.

FIELD OF THE INVENTION

The invention relates to processes or methods for obtaining alternative energy, particularly by interaction of Gamma Radiation with melanin, melanin precursors, melanin derivatives or analogues to alter its oxidation-Reduction Potential that results in electric current production, whereby hydrogen and oxygen atoms are obtained by means of the separation or partition of water molecule with which hydrogen and oxygen atoms are generated. Because the reactions occur in both ways, the invention can be applied to electricity generation, for our method permits to bind hydrogen and oxygen atoms forming water molecules, and collaterally generating electrical current.

BACKGROUND OF THE INVENTION

About the related art, nowadays, the known processes used up to now to separate the water molecule in hydrogen and oxygen atoms are, among others:

- a).-The application of intense electrical currents.
- b).-The heating of water until two thousand degrees centigrade.
- c).-The separation of water molecule by solar electrochemical method.
- d).-Another method to separate water is by solar energy concentration (with mirrors for example), with the object to elevate water temperature until two thousand ° C. This is the required temperature used in laboratory to divide the water molecule.
- e) One further method is by using photosynthetic microbes as green algae and cyanobacterium, those produce hydrogen from water as part of metabolic activities using light energy as main source. This photobiological technology is promising, but as oxygen is produced as well as hydrogen, the technology must solve the limitation that is the sensibility to oxygen in the enzymatic systems. Besides, hydrogen production from photosynthetic organisms is currently too low to be economically viable.
- f).-Another method is water electrolysis, using electricity to separate the water molecule in its compounds (hydrogen and oxygen atoms). At present time, two kinds of electrolyzers are used for commercial production of hydrogen: the alkaline, and the membrane of protons

interchange, but these approaches cannot compete now from an economic point of view with the hydrogen produced from natural gas. (Source: U.S. Department of Energy, Efficiency and Renewable Hydrogen fuel cells and Infrastructure Technology Program Hydrogen Production & Delivery).

A natural material that can also divide or separate the water molecule and that has been studied is chlorophyll but because its affinity with light is between 400 nm and about 700 nm the rest of the light energy is lost. That is why it is estimated that 80 per cent of used energy is wasted. Moreover, its production is complex and expensive, requiring for example temperatures of -8°C .

The problem of the invention is to find an energy source that gives energy to an electrolyzing water element to create a process and a device to separate the water molecule in hydrogen and oxygen atoms, other than the aforesaid, so an electric current is generated. In particular one problem of the invention is to find an energy source to start an electrolyse of the water, other than the aforesaid energy sources.

This problem is solved according to the invention by all the features of the independent process claim and by all the features of the independent product claim. The dependent patent claims specify possible embodiments.

In particular the invention is characterized in that the energy source is an inner energy source within the container and whereby the energy obtained from the energy source is electromagnetic radiation that is absorbed by melanins.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENT

These are the reasons by which the present invention proposes to use the melanins as electrolyzing water element by absorbing electromagnetic radiation for generating an electric current.

Melanins are complex polymers found in species of all biological kingdoms with a multifaceted utility related to physiology such as protection from visible and UV light [1,2], decreased oxidative stress [3], energy transduction and Fe(III) reduction [4-6]. However, the most fascinating and the least explored function of melanin is related to its interaction with ionizing radiation (gamma radiation). Melanin plays a role in decreasing radiosensitivity of human melanoma cells [7] and melanized microbial species thrive in highly radioactive environments such as cooling pools of nuclear reactors, the stratosphere, space stations and inside the damaged nuclear reactor at Chernobyl [8]. Furthermore, certain melanized microbes seem to dominate the environments characterized by elevated levels of ionizing radiation such as pyomelanin-producing bacteria found in uranium-contaminated soils [9] and melanized fungi in radio-contaminated soils showing directional growth towards radiation sources (radiotrophism) [10]. Recently it could be demonstrated that ionizing radiation changes the electronic structure of melanin and enhances the growth of several melanized fungal species, suggesting a role for melanin in this process [11], specifically the physico-chemical interaction between melanin and ionizing radiation. Melanin pigments are diverse in structure and function in regard to effects from ionizing radiation [7,12] and offer potential as manufactured radioprotective materials [12].

Melanin pigments are composed of quinone moieties that are believed responsible for its redox behavior. The polymeric structure of melanins permits oxidation and reduction to occur simultaneously. Perhaps the most interesting aspect of any radio protective material is its requirement to withstand the oxidizing impact of ionizing radiation indefinitely, without bleaching.

Preferably the invention consists essentially in obtaining under normal temperature until to 500°C, and using electromagnetic radiation, as the only source of energy, the division of water molecule to obtain hydrogen and oxygen atoms as well as electrons of high energy or join hydrogen or oxygen atoms to obtain water and electric current; using as main or central electrolyzing melanins: polihydroxyindole, eumelanin, feomelanin, alomelanin, neuromelanin, humic acid, fullerens, graphite, polyindolequinones, acetylene black, pyrrole black, indole black, benzene black, thiophene black, aniline black, poliquinones in hydrated form sepiomelanins, dopa black, dopamine black, adrenalin black, catechol black, 4-amine catechol black, in simple linear chain, aliphatics or aromatics; or their precursors as

phenoles, aminophenols, or diphenols, indole poliphenols, ciclodopa DHI Y DHICA1, quinones, semiquinones or hydroquinones. L-tyrosine, L-dopamine, morpholin, ortho benzoquinone, dimorpholin, porphirin black, pterin black, ommochrome black, free nitrogen precursors, any of the above listed with any size or particles. (from 1 angstrom to 3 or 4 cms.). All aforementioned compounds are preferably electroactive, in suspension, solution, in gel, that absorb the ultrasound in the interval of one MHz, natural or synthetic, with vegetal, animal or mineral origin; pure or mixed with organic or inorganic compounds, ions, metals, (gadolinium ,iron, nickel, copper, erbium, europium, praseodymium, dysprosium, holmium, chromium or magnesium, lead selenure, and so on). Gadolinium is a very effective metal. The metal is incorporated into the melanin in ionic form or as a particle, as well as drugs or medication energizing the electrochemical design with electromagnetic radiation (natural or synthetic), though other energy types, for example, the kinetic, also are efficient in various grades, according to the rest of the conditions (pH, temperature, pressure, and so on). To this kind of designs magnetic fields from soft to significant intensity can be applied in addition.

The events in this design may occur to a greater or lesser extent under internal or external physical or chemical stimuli.

Preferably the use of melanins (as mentioned before) is propound as the electrolyzer material of the water molecule, using electromagnetic radiation as main or sole energy source for the hydrogen production systems known as electromagnetic radiation/electrochemical methods. As aforementioned, these systems integrate a semiconducting material and a water electrolyzer inside a monolithic design to produce hydrogen atoms directly from water, using electromagnetic radiation as the main or sole source of energy, though sound, ultrasound, in an interval of one MHz, mechanical stir, magnetic fields, etc. can also be used.

At least two basic criteria had to be met to find a material that could withstand the whole process: one was the electromagnetic radiation absorbing system or compound had to generate enough energy to start, lead and support completely the electrolysis reaction, and it had to be low cost, stable and long lasting in a water environment.

Melanins can meet reasonably and efficiently the above mentioned requirements and this represents a progress to solve the central problem of electromagnetic radiation designs.

Moreover, as it is shown in the Figures it can be demonstrated that irradiation of melanin resulted in melanin oxidation over time as measured by current production as a function of melanin concentration and time of exposure of gamma radiation. An increased oxidation of melanin in the presence of ascorbate during exposure to ionizing radiation is due to the reductive properties of ascorbate. Maintaining the radioprotective properties of melanin-like materials is key for long-term functioning of the material.

The shape of the container holding it in the appropriate equipment can be very varied: cubic, cylinder, spherical, polyhedral, rectangular, etc. Being one of the main requirements, to be transparent, in order to prohibit the electromagnetic radiation to pass through. The walls could be made of plastic for example with a special lead alloy, so that the walls of the container do not emit radiations by the emitting energy source. The material of which the container is made could be of a color that allow maximum transparency or absorption of the wavelength from the electromechanical spectrum, so that beside the inner energy source within the container an outer energy source preferably light, to produce protons in the photolysis. The walls can be made of glass or of any other polymer whose permeability characteristics of the electromagnetic radiations fit to the final needs of the electromagnetic radiation design, so that no radiation escapes.

Inside the cell, the main material, the essential solute, is preferably mainly dissolved in water. The basis of the design is the notable capacity of melanin to absorb the radiation from the inner electromagnetic radiation source, probably by the surrounding portions of the molecule, followed by the generation of high energy electrons from low energy electrons. These high energy electrons go to the centers of free radicals of the compound where they are probably captured by an element for example: a metal such as iron, copper, gadolinium, europium, etc. from where they are transferred to a primary electron acceptor from a nature that is uncertain up to now because the union is complex and comprises ionic interactions depending on the pH. This electron transfer liberates energy which is used to establish the protons gradient.

The combination of the melanin molecule with water forms a system, which captures electromagnetic energy using at least two interrelated activities: removal of electrons from water and generation of a protons gradient.

The melanin components are in very close contact among them which makes a fast transfer of energy easy. At three picoseconds of electromagnetic radiation, the melanin reaction centers respond transferring an electromagnetic-excited electron to the primary electron receptor.

This transference of electrons generates a donator, positively charged and a receiver negatively charged. The importance of the formation of two species with opposite charges is seen when we consider the reduction capacities of these two species, because one of them is deficient in electrons and can accept electrons which makes it an oxidizing agent. By contrast, the other compound has an extra electron that can be lost easily, making it a reducing agent. This event - the formation of an oxidizing agent and a reducing agent from the electromagnetic radiation- takes less than billionesimal of second and is the first essential step in the process of generating an electric current.

Because they are charged in an opposite way, these compounds show an obvious mutual attraction. The separation of charges is (probably) stabilized by their movement to opposite sides of the molecule; being the negative compound the one that first gives its electron toward a quinone (Q1) and possibly then the electron is transferred to a second type of quinone (Q2), this producing a semi reduced form of the quinone molecule which can be strongly linked to the reaction center of the melanin molecule. With each transfer, the electron gets closer to the reaction center of the melanin molecule. The portion of melanin positively charged is reduced, thus preparing the reaction center for the absorption of another photon. The absorption of further electromagnetic/ionizing radiation sends a second electron along the way (melanin negatively charged towards the first and second quinone molecule - Q1 and Q2 -). This second molecule absorbs two electrons, and thus combines with two protons. The protons used in this reaction could derive from the same melanin molecule or from the surrounding water, causing a decrease in the concentration of hydrogen ions of the photosystem, what contributes to the formation of a protons gradient. In theory the reduced quinone molecule is dissociated from the reaction center of melanin,

been replaced reaction by a new quinone molecule. These reactions occur at normal temperature but when you modify for example the temperature you can favor the reaction in one or other way, depending on the control of the other variables: (pH, magnetic fields, concentrations, gases, partial pressures, shape of cells, etc.) and the main objective of the process.

The separation of water molecules into hydrogen and oxygen atoms is a highly endergonic reaction due to the very stable association of hydrogen and oxygen atoms. The separation of the water molecules (in hydrogen and oxygen atoms) in the laboratory requires the use of a strong electric current or high temperature of almost 2,000 °C the above (water electrolyzing) is obtained by melanin at room temperature, more preferably the boiling temperature of the melanin solution that is close to 500°C, using only the energy obtained from electromagnetic radiation, either from natural or artificial source. It is estimated that the redox potential of oxidized form of quinone is approximately +1.1 V, what is strong enough to attract the firmly united low energy electrons from the water molecule (redox potential of +0.82), separating the molecules in hydrogen and oxygen atoms. The separation of the water molecule by absorbing electromagnetic/ionizing radiation emitted by the emitting radiation source that is called radiosynthesis is nearly the same as the separation of the water molecule by photopigments via photons that is named photolysis. It is believed that the formation of the oxygen molecule during the photolysis or during the radiosynthesis requires the simultaneous loss of four electrons from two water molecules according to the reaction:



A reaction center can only generate a positive charge or its oxidizing equivalent at the same time. This problem is solved hypothetically by the presence of four nitrogen atoms in the reaction center of the melanin molecule, each one of them transferring only one electron. This nitrogen concentration, adds may be four positive charges upon transferring four electrons (one each time) to the closest quinone⁺ molecule.

The transfer of electrons from the nitrogens of the reaction centers to the quinone⁺ is obtained by means of the passage through a positively charged tyrosine moiety. After each electron is transferred to quinone⁺, regenerating quinone, the pigment is reoxidized (again a

quinone+) after the absorption of another electron from the radiation emitting source that is similarly to a photon to the reaction center. So the accumulation of four positive charges (oxidizing equivalents) by the nitrogen atoms of the reaction center is modified by the successive absorption of four electrons by the melanin reaction center during irradiation of Melanin. Once the four charges have been accumulated the oxygen releasing quinone complex can catalyze the $4e^-$ removal from $2H_2O$ forming an O_2 molecule (diatomic oxygen), and regenerating the totally reduced quantity of nitrogens in the reaction center.

The protons produced in the radiosynthesis are released in the medium where they contribute to the protons gradient. The reaction center must be irradiated several times before the occurrence of O_2 release and thus hydrogen (diatomic hydrogen) can be measured; this indicates that the effects of the individual irradiation reactions must accumulate before O_2 and hydrogen are released. The gases released during the radiosynthesis, wherein the gases are mainly diatomic hydrogen and diatomic oxygen can be extracted during the process, preferably. The gases are very stable because it needs no spark to perform the radiosynthesis. In this case water is consumed slowly and must be replaced at least temporarily to disable the output of diatomic oxygen and diatomic hydrogen. Therefore an advantageous aspect of the present invention is the use of the diatomic oxygen and the diatomic hydrogen which can be extracted as gas formed during the radiosynthesis as useable energy, for example in fuel cells.

The quinones are considered carriers of mobile electrons. It is to be kept in mind that all electron transfers are exergonics and occur as the electrons are successively taken to carriers with an increasing affinity for the electrons (more positive redox potentials). The need of having electron moving carriers is obvious. The electrons generated by the radiosynthesis can pass to several inorganic receivers, which are thus reduced. These ways for electrons can lead (depending on the composition of the used mix) to the eventual reduction of nitrate molecule (NO_3) into ammoniac molecule (NH_3) or the sulphates in sulphhydrides (SH^-) reductions that change the inorganic wastes into compounds necessary for life. So the electromagnetic radiation can be used not only to reduce the most oxidized form of a carbon atom (CO_2) but also to reduce the most oxidized forms of nitrogen and sulphur.

The production of one O_2 molecule requires the removal of four electrons from two molecules of water, the removal of four electrons from water requires the absorption of four photons, one for each electron.

The design of the cell is an important parameter for the optimization in obtaining the product of the reaction in which we have a particular interest, because the addition of electrons, the nature of them, the use of magnetic fields, the addition of several compounds (organic or inorganic, ions, metals, drugs or medications) to the system that at the beginning was only melanin and water, plus the addition of electrolytes, plus the addition of medicines, and temperature management, the control of partial pressures of gases, the management of the electrical current generated, the application of magnetic fields, the level of pH, the material used in making the cells and the shape and disposition of its internal divisions, etc. Apart from other variables, which are able to be controlled in such a way that the final design can recover electrons, or protons, or oxygen, and the resulting compounds according to the formulation of the medium in the melanin is dissolved. Thus, the melanins (pure or combined with organic compounds and inorganic compounds, metals) allow a notable flexibility of the design according to the goals to reach.

The optimization of radiation-electrochemical design relates to the objectives, for example: for a higher generation of protons and oxygen or generation and conserving of electricity; the largest possible area of exposition of the liquid compound to the emitter emitting electromagnetic radiation in an extended container, apart from other procedures such as the addition of electrons carrier compounds, melanin doping, etc., as also positive microlens to concentrate the light to support photolysis beside the radiosynthesis.

The design of the container is not limited and can have a spherical, cubic, rhomboidal, polyhedric, plain concave, plain convex, biconvex, biconcave shape with microlens in a side (the side exposed to light to concentrate it) and flat on the other side cylindrical, circular cylindrical, hollow cylindrical, circular cone (straight) truncated cone, rectangular prism, oblique prism, rectangular pyramid, straight truncated pyramid, truncated spherical segment, spherical segmented, spherical sector, spherical with cylindrical perforation, sphere with conic perforations, torus (circular section ring), cylinder with slanted cut, cylindrical wedge, semi prism barrel, and combinations of them, etc, because the liquid assumes any shape,

depending of the kind of melanin used (doped or not, for example), it will be convenient to select a specific irradiation, preferably gamma irradiation to the soluble melanin, but until this moment one of the big virtues of soluble synthetic melanin is that it absorbs the majority of the radiation emitted by the emitting source. The control of the partial pressures of the gases in the interior of the cell is an important variable, and depending on the cell shape and the use given to it, these pressures can go from 0.1 mm Hg until 3 or 4 atmospheres; another variable that must be taken into account is the concentration of different substances dissolved in the liquid, where the critical concentration is mainly of melanin and can go from 0.1% to 100%; other variable that can be modified is the ratio among the different components of the formula (depending on the use), because potassium can be added in a concentration from 0.1 to 10%; sodium in a concentration from 0.1 to 10%; chlorine in a concentration from 0.1 to 10%; calcium in a concentration from 0.1 to 10%; iron in a concentration from 0.1 to 8%, copper in a concentration from 0.1 to 5%, arsenic in a concentration from 0.1 to 8 or 9%, gold in a concentration 0.1 to 8 or 9%, silver in a concentration similar to gold, nickel in a concentration from 0.1 to 8%, gadolinium, europium, erbium, etc.

The final volume can range from 1 microliter to 10 or 20 liters depending on the size of the container and the available space; the temperature can fluctuate from 2 to 45° C, especially from 45°C to 150°C and in most advantage from 200°C to 500°C. The frequency of change of solution can be from every 15 minutes to several months or 2 or 3 years; the formation of compartments inside the little cell, in the interior of the cell shapes ranging from small spheres (microspheres, there can be several dozens of them) to spheres the size of which could be included 3 or 4 times inside the whole design, and in the shape of the interior of the little cell cubic rhombic, polyhedral, concave plane, convex plane, biconvex, biconcave with microcells, biconvex on one side (the side exposed to light to concentrate it) and flat on the other side, cylindrical, circular cylindrical, hollow cylindrical, circular cone (straight), truncated cone, rectangular prism (straight), oblique prism, rectangular pyramid (straight), truncated pyramid, truncated spherical segment, spherical segment, spherical sector, spherical with cylindrical perforation, sphere with conic perforations, toro (circular section ring), cylinder with slanted cut, cylindrical wedge, barrel, semiprism, can be used including combinations of these, the power of the optionally microlens can range from 0.1 to 100 diopters, the redox properties of the materials used in the formation of the compartments (iron, silver, copper,

nickel, gold, platinum, gallium arsenide, silicon, gadolinium, europium, erbium, praseodymium, dysprosium, holmium, chromium, magnesium, lead selenide and alloys of them, etc).

The use or not of cathodes and anodes, their material (for example platinum, iron, silver, gold, steel, aluminum, nickel, arsenium, gadolinium, europium, erbium, praseodymium, dysprosium, holmium, chromo, magnesium; gallium), depending on the optimal characteristics to recover electrons or hydrogen, but it has to be kept in mind that in presence of metal or borium, the hydrogen works with -1; another variable is initial pH of the solution that can range from 2 or 3 to 8 or 9 units of pH, being the most used about 7, the above mentioned variables that can be handled in order to control the photoelectrolysis process depending on the needs of the project in question.

The core of any efficient radiation-electrochemical designs are the melanins, melanin variants and analogues, water soluble, where they catalyze the radiosynthesis process, without undergoing significant changes except the presence of elements such as magnesium, iron, copper, lead, and others, the resulting products of which together with the resulting products of the partial reduction of the oxygen atom (superoxide anion, hydroxyl radical, hydrogen peroxide, quinones and orthoquinones), can fast or slowly damage the effectiveness of melanin, but in the case of pure melanin, at a 10% concentration, for example, the duration of the compound is long enough to be economically convenient (years), and the synthesis of melanin is a very efficient process. Thus, from an economic and ecological point of view it is very viable, because pure melanin is fully biodegradable. Thus, the little cell only requires beside the solid electromagnetic radiation source a periodic supply of distilled water, as well as a periodic replacement of soluble melanin, or eventually, the renewal of substances added to the design to optimize or potentiate some of the processes occurring as a result of exposing the radiation-electrochemical design to the irradiation. The ecological advantage of the final products of the reaction being water molecules, oxygen molecules or atoms, hydrogen, high energy electrons, and electrical current can be easily realized. There is little generation of greenhouse effect CO₂ molecules. The transfer of electrons releases energy, which is used to establish a proton gradient.

The proton movement during the electrons transportation can be compensated by the movement of other ions, advantageously using membrane and a solvent with adequate solutes, membrane potential can be formed from photons capture by mean of melanin.

Melanins, melanin precursors, melanin derivatives, variants and analogues, oxidize the water molecule to O, O₂, and H₂, absorbing energy obtained by the electromagnetic/ionizing radiation (electrons), and reduce oxygen atom with hydrogen atoms to H₂O, liberating energy (electricity, although it can “keep” or “conserve” the electricity, i.e. it can function as a battery or accumulator, i.e. not only generating energy but also keeping it for a while and within some limits). That is why the cell design can be adapted to the requirements.

H₂ and O₂ atoms are produced with irradiation by electromagnetic radiation, but the generation of these elements can be increased by melanin doping (melanin) with metals or adding organic and inorganic molecules, also modifying the electrolyte concentrations, adding drugs or controlling the characteristics of irradiation, over the liquid containing water and melanins (melanin).

The radiation-electrochemical reactions happen in two ways, i.e. the water molecule is separated but also formed, so it can recover electric current of the design and it can also be optimized through melanin doping with different substances (drugs, metals, electrolytes, organic and inorganic molecules, and others) or by light concentration by mean of lens, among others.

The box containing the liquid can have different shapes that adapt to different needs, in the house roofs, car roofs, plants buildings, industrial processes, etc. cells connected among them, but the central component of the design is melanin (melanins, water soluble), that induces and carries out the radiosynthesis in the manner as the photolysis of the water molecule, but in presence of electromagnetic radiation instead of light.

The melanins remove electrons from water and generate a gradient of protons.

The electromagnetic radiation depending reactions can also generate energy to reduce CO₂ to CH₂O, nitrates to ammonia and sulphates to sulphydriles.

As one aspect of the invention the electrochemical response of the pigment eumelanin to ionizing radiation with a carbon paste/melanin electrode was investigated. The results shown in the figures establish that gamma radiation can interact with melanin thus providing key supportive evidence for the initial interactions of this pigment with electromagnetic/ionizing radiation in such processes as radiosynthesis.

Advantageously, the inner energy source emitting electromagnetic/ionizing radiation is an emitter that is within the aqueous solution or that is separated within the container from the aqueous solution, maybe by incorporating the inner energy source in a container that is surrounded by the aqueous solution, so that the emitter is not in contact with the aqueous solution, directly. But in another preferred embodiment of the invention, the emitter could be within the aqueous solution, preferably as particles. This embodiment of emitter particles in the aqueous solution has the advantage, that the emitter is fairly equally distributed in the aqueous solution.

The substance that can be used as the emitter or as the emitter particles are from, but are not limited to the substance: titanium-44, Uranium 232, plutonium-238, nickel-63, silicon-32, argon-39, californium-249, silver-108, americium-241. Niobium-91, carbon-14, curium-245, niobium-94, plutonium-239, nickel-59, neptunium-236, uranium-233, technetium-99, uranium-234, chlorine-36, curium-248, aluminum-26, beryllium-10, zirconium-93, technetium-97, manganese-53, technetium-98, palladium-107, curium-247, uranium-236, niobium-92, plutonium-244, samarium-235, potassium-40, uranium-238, thorium-232, lutetium-176, rhenium-187, rubidium-87, lanthanum-138, samarium-147, platinum-190, barium-130, gadolinium-152, indium-115, hafnium-174, osmium-186, neodymium-144, samarium-148, cadmium-113, vanadium-50, tungsten-180, molybdenum-100, bismuth-209, zirconium-96, cadmium-116, selenium-82, tellurium-130, germanium-76, xenon-136, neodymium-150, calcium-48, cobalt-60 or tellurium 128. The emitter and/or the emitter particles can also be a combination of at least two of the aforementioned substances, and could be preferably in form of mixed oxide.

In one preferred embodiment of the invention, the substances as for the emitter and/or as for the particles are preferably from enriched reprocessed substances, for example enriched reprocessed uranium (ERU) and mixed oxide (MOX). Thus, this invention can be seen as a

development of a new beneficiation method or as a recycling concept for sustainable utilisation of secondary raw materials, which are difficult to dispose.

As already mentioned above the temperature of the aqueous solution within the container is from 2°C to 500°C. But in one particularly preferably embodiment of the invented process the temperature of the aqueous solution within the container is from 350°C to 500°C, preferably close to 500°C. Therefore, it needs to design the container with a material that is heat resistant, also.

One special embodiment of the invention is a cell, for example a battery or an accumulator, for generating an electric current. For using the cell as for the invented process, especially to incorporate the emitter as an inner energy source emitting electromagnetic radiation and the aqueous solution, the cell comprises a container that is preferably made of a material permitting at least electromagnetic radiations to pass through. Moreover the material as for the container is preferably heat resistant especially at least until 500°C, so that the invented process may be performed with temperatures until 500°C or preferably close to 500°C. In the aqueous solution within the container of the Cell melanin, its precursors, variants, derivatives, or synthetic or natural analogues, are preferably dissolved. As for the connection of the cell to a consumer the cell preferably comprises a cathode and an anode as already mentioned above.

In one advantageous embodiment of the invented cell the aqueous solution including melanin, melanin precursors, melanin derivatives, or analogues is able to absorb the electromagnetic radiation. But in a particularly preferably embodiment of the invention, the aqueous solution including melanin, melanin precursors, melanin derivatives, or analogues is able to fully absorb the electromagnetic radiation. This design of the cell with an aqueous solution including melanins that is able to fully absorb the electromagnetic radiation in addition with a material that permits to pass through electromagnetic radiation increases the safety of the invented cell to a maximum.

The present invention is further described in detail with respect to the accompanying material and methods and the drawings. Features discussed within the material and methods and in the description of the drawings can be combined with each other freely. The features

mentioned in the claims and the specification are essential to the invention, either in themselves or in any given combination.

EXAMPLES

Materials and Methods

Eumelanin produced by the fungus *Cryptococcus neoformans* grown in presence of L-DOPA melanin precursor was used. The growth of melanized cells was followed by acid hydrolysis resulting in production of hollow melanin shells dubbed "ghosts" (as they preserve the shape of the cell) was performed as in [2].

A carbon paste (CP) electrode was utilized, which was previously used for studying melanin electrochemistry [6, 14]. Dry melanin ghosts were crushed lightly to a powder with a clean glass rod and mixed by weight with CP (Bioanalytical Systems, BAS, West Lafayette, IN) at melanin /CP ratios of 10:90, 20:80 and 30:70. The melanin/CP mixtures were packed into electrode housings (BAS) as described in [6, 14]. The packed electrodes were stored in sterile, pH 7, 50 mM/L, phosphate buffered saline (PBS) at 25°C for at least one week to allow the melanin to hydrate. Since fungi are known to produce extracellular reductants [13, 14] the method was performed in the presence and absence of a reductant, in order to gain a better understanding of the potential role of extracellular reductants on the long-term radioprotective properties of melanin. When used as a reductant, 1 mM/L ascorbate (final concentration) was incorporated with PBS and electrodes were exposed to this solution for at least 24 hrs to allow ascorbate to diffuse into the electrode material. Cyclic voltammetry was used to confirm that sufficient ascorbate was in contact with the electrode material by demonstrating reduced conditions.

Electrochemical measurements were performed in a three-electrode geometry with a CP or melanin/CP working electrode, a Pt counter electrode (BAS) and a Ag/AgCl, 3M NaCl reference (BAS), all immersed in 50 mM/L, pH7.0, PBS with or without 1mM/L ascorbate. Electrodes were contained in glass vials fitted with screw on caps and teflon septa at either end of the vial. Electrodes were positioned via holes in the septa. The working electrodes were positioned facing up to prevent gas bubble accumulation on their surface (see Fig. 1).

Electrochemical analyses were conducted on a VersaSTAT MC Multichannel Potentiostat/Galvanostat (Princeton Applied Research) employing 2 channels simultaneously. For gamma irradiation studies, the cables from the potentiostat were run through access ports of a 25 cm x 25 cm x 100 cm chamber (J. L. Shepherd Model 484). Vials with electrodes were positioned in a stainless steel rack and attached to leads from the potentiostat. Dose rate was determined by distance of the electrodes from a ^{60}Co gamma source inside the chamber for each experiment. For some experiments that required a high current response to aid in quantification, racks were designed to fit as close as possible to the gamma source for maximum radiation dose.

Chronoamperometry was performed with electrodes poised at -700 mV and irradiation began after electrode current stabilized. Current production as a result of gamma irradiation was determined by the difference in current from the start of irradiation and at 60 and 90 min. Chronopotentiometry was conducted with electrodes poised at 1 mA. Cyclic voltammetry was conducted at a sweep rate of 100 mV /s from -1 V to 1 V for multiple cycles. Cyclic voltammograms were recorded prior to and throughout irradiation. Selected voltammograms were used to illustrate changes as a result of ionizing radiation exposure. All experiments were repeated 2 to 3 times with representative data illustrated here.

There are shown:

Fig. 1 configuration of an electrochemical cell for electrochemical measurements in radiation studies,

Fig. 2 a diagram showing gamma radiation-induced melanin oxidation with increased current production of a 20% melanin/80% CP electrode (upper line), poised at -700 mV (vs. Ag/AgCl) during irradiation with 300 Gy/h ^{60}Co gamma radiation. The CP (control) electrode response is shown in the lower line. Both electrodes were submerged in pH 7.0 PBS,

Fig. 3 a diagram showing changes in melanin oxidation-reduction potential upon ^{60}Co exposure; CP with 10% melanin electrodes (upper line) and CP controls

(lower line) in pH 7 PBS were poised at a constant current of 1 mA during 600 Gy/h ^{60}Co gamma irradiation,

- Fig. 4 a diagram showing gamma radiation-induced melanin oxidation with and without 1 mM/L ascorbate; 20% melanin/80% CP electrode was submerged in pH 7.0 PBS with the reductant ascorbate (upper line) relative to pH 7.0 PBS alone (lower line) during exposure to 300 Gy/h ^{60}Co gamma radiation; both electrodes were poised at -700 mV (vs. Ag/AgCl) (background current from CP electrode exposed to identical conditions was subtracted),
- Fig. 5a a diagram showing cyclic voltametry studies during gamma irradiation without a reducing agent,
- Fig. 5b a diagram showing a diagram showing cyclic voltametry studies during gamma irradiation in the presence of a reducing agent,
- Fig. 6 a schematic drawing of a sample embodiment of the invented cell, and
- Fig. 7 table 1 showing results of change in current production as a function of melanin concentration and time of exposure to 4000 Gy/h ^{60}Co gamma radiation; CP electrode (control) and 10-30% melanin/CP electrode poised at -700 mV in pH 7 PBS.

In the different figures same features always correspond to the same reference signs, therefore generally the features are only described once.

In the sample embodiment of an electrochemical cell 1 for electrochemical measurements depicted in Fig. 1 the electrochemical cell 1 comprises a three-electrode geometry with a working electrode 2, that can be a CP or melanin/CP working electrode 2, a Pt counter electrode (BAS) 3 and a Ag/AgCl, 3M NaCl reference electrode (BAS) 4. The electrodes 2, 3 and 4 are immersed in an aqueous solution 6 (50 mM/L, pH7.0, PBS with or without 1mM/L ascorbate as reducing agent). The Electrodes 2, 3 and 4 are contained in a glass vial 8 fitted with screws 6 on caps 7 and teflon septa (not shown) at either end of the vial 8. The

electrodes 2, 3 and 4 are positioned via holes in the septa. The working electrode 2 is positioned facing up to prevent gas bubble accumulation on its surface.

Fig. 2 shows in a diagram results from gamma radiation-induced melanin oxidation made in the electrochemical cell 1. Increased current production of a 20% melanin/80% CP electrode 2 (upper line), poised at -700 mV (vs. Ag/AgCl) during irradiation with 300 Gy/h ^{60}Co gamma radiation. The CP (control) electrode 2 response is shown in the lower line. Both electrodes 2 were submerged in pH 7.0 PBS. With the electrochemical cell 1 it could be shown that CP mixtures packed into working electrode 2 housing resulted in greater current production from stable electrodes 2 consisting of CP (80% w:w) plus melanin (20%) compared to only CP electrodes 2. The increased current from electrodes 2 poised at -700 mV was a result of oxidation of the electrode 2 materials during irradiation. The sustained current of the working electrode 2 with melanin relative to the working electrode 2 with CP alone indicated that the melanin remained oxidized for an extended time after irradiation ceased (radiation off). This is likely due to the presence of stable free radicals in melanin [11]. Current production from the working electrode 2 is directly proportional to melanin concentration (10-30%) and irradiation time (see Table 1) and corroborates previous studies with eumelanin [7].

Fig. 3 shows a diagram showing changes in melanin oxidation-reduction potential upon ^{60}Co exposure. CP with 10% melanin electrodes 2 is indicated with the upper line and CP controls electrode 2 is indicated with the lower line. Both electrodes were submerged in pH 7 PBS and were poised at a constant current of 1 mA during 600 Gy/h ^{60}Co gamma irradiation. During gamma irradiation, oxidation of 10% melanin/90% CP continued to increase under a continuous 1 mA current supplied to the electrode 2. In contrast, the potential of the control CP electrode 2, without melanin, decreased. These results demonstrated that with a 1 mA input, melanin demonstrated a significant capacity for sustained oxidation in radiation fields.

Fig. 4 shows a diagram showing gamma radiation-induced melanin oxidation with and without 1 mM/L ascorbate as a reducing agent. The upper line shows the results of the 20% melanin/80% CP electrode 2 that was submerged in pH 7.0 PBS with the reductant ascorbate. The lower line shows the results of the electrode 2 with pH 7.0 PBS alone during exposure to 300 Gy/h ^{60}Co gamma radiation. Both electrodes 2 were poised at -700 mV (vs. Ag/AgCl) (background current from CP electrode exposed to identical conditions is

subtracted). In the presence of the reducing agent ascorbate, current production is greater than in PBS alone. The reductive properties of ascorbate likely contributed to the reductive behavior of melanin by providing electrons supplemental to that of the electrode 2. Although the ascorbate, which is a known free radical scavenger, was present in much higher concentrations than estimates of free radicals formed as a result of water radiolysis, its presence did not abrogate the oxidation of melanin during gamma irradiation. This observation could suggest that other free radicals from radiolysis were involved in the oxidation of melanin. Similarly, melanin oxidation resulting from a loss of Compton electrons during Compton scattering could not be discounted. Current production by the electrode 2 in ascorbate is monitored well after irradiation ceased (radiation off) in order to track this phenomenon. Current production was maintained for over 2.75 h and was likely due to the presence of stable free radicals in melanin, especially after exposure to ionizing radiation.

Fig. 5a and Fig. 5b illustrates diagrams showing results cyclic voltammetry studies during gamma irradiation of electrodes 2 submerged in pH 7.0 PBS without a reducing agent (Fig. 5a) and cyclic voltammetry studies during gamma irradiation of electrodes 2 submerged in pH 7.0 PBS in the presence of 1 mM/L ascorbate as the reducing agent (Fig. 5b) of 20% melanin/80% CP electrodes 2 exposed to 4000 Gy/h of ^{60}Co gamma radiation for; 0 min. (line a); 17 min. (dashed line); and 33 min. (line b). Background from CP electrode 2 (control) is subtracted. Scan rate was 100 mVs^{-1} . All scans began at -1 V.

Without irradiation, cyclic voltammetry demonstrated an oxidation peak at about 100 mV (vs Ag/AgCl) and a broad reduction peak (Fig. 5a). This peak is similar to those previously reported for oxidized melanin using synthetic melanin films. A slight reduction peak at about -100 mV (vs Ag/AgCl) can be also observed. Following 17 min of irradiation, the oxidation peak increased while the cathodic, or reduction response demonstrated diminished peak current. This trend continued into 33 min of irradiation with a slight increase in peak current at 100 mV and a more defined reduction response with two reduction peaks emerging, near -100 mV and -400 mV. Similar reduction peaks at various pH values were reported previously for synthetic L-DOPA melanin.

Cyclic voltammetry demonstrated a dramatically increased oxidation of melanin in ascorbate solution during gamma irradiation (Fig. 5b). In the presence of ascorbate, melanin was more

reduced (Fig. 5b), as evident in the oxidation peak at about 100 mV (vs Ag/AgCl) and the reduction peak at about -100 mV (vs Ag/AgCl). The addition of ascorbate also resulted in increased peak current at 100 mV (vs Ag/AgCl) from both electrodes with a substantial increase in oxidation (Fig. 5b). These data suggest a 2-step reduction from semiquinone to hydroquinone, followed by a 1-step oxidation to the quinone state.

The continued decrease in reduction peaks (Figs. 5a and 5b) indicated that reduced components of the melanin molecule were continuously oxidized during radiation and the increased peak current of the anodic peak corroborates this scenario.

Fig. 6 illustrates a schematic drawing of a sample embodiment of the invented cell 100 to generate an electric current, so that the cell 100 can be mentioned as a battery or an accumulator. The cell 100 comprises a container 110. The material of the container 110 is preferably a material that permits at least electromagnetic radiations to pass through. The container 110 is hermetic sealed. Moreover the container is designed in such a way that it is stable at boiling temperature until 500°C. The cell 100 also comprises melanin, its precursors, variants, derivatives, or synthetic or natural analogues (melanins), which are mainly dissolved in an aqueous solution 120 in the cell 100, especially within the container 110. Optionally the cell 100 comprises a cathode and an anode (not shown in the figure) for example to connect the cell 100 with a consumer. As for the radiosynthesis the cell 100 comprises within the container 110 an inner energy source 130. The inner energy source 130 is preferably a radioactive substance emitting electromagnetic/ionizing radiation. In the figure 6 the inner energy source 130 is a one piece emitter 140, but the inner energy source 130 could be particles in the aqueous solution 120, also (not shown). The concentration of the melanins can be 3-10 % by volume. With such a design the output of the cell 100 that is a battery or an accumulator increase significantly. Moreover, no radiation escapes because the aqueous solution 120, though, melanin, in the right proportion, is able to fully absorb the radiation emitted by the emitting inner energy source 130. On the other hand melanin emits energy absorbed and dissipated by dissociating water molecules, which to reform it, generates electric current.

Aforesaid discussion of the different embodiments is only carried out by the way of example and does not limit the scope of protection of the present invention. Moreover, the use of ⁶⁰Co

gamma radiation as an emitter emitting electromagnetic/ionizing radiation is depicted as an example only, but ^{60}Co gamma radiation could be replaced with any other energy source 130 disclosed in the present invention, also.

List of reference numbers

- | | |
|-----|--|
| 1 | cell |
| 2 | working electrode with melanin and without melanin |
| 3 | Pt counter electrode (BAS) |
| 4 | reference electrode (BAS) 4 |
| 5 | aqueous solution |
| 6 | screws for fitting electrodes |
| 7 | cap |
| 8 | vial |
| 110 | cell/battery/accumulator |
| 110 | container |
| 120 | aqueous solution |
| 130 | energy source |
| 140 | one piece emitter |

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P a t e n t c l a i m s

1. Process of generating an electric current, wherein melanin, melanin precursors, melanin derivatives or analogues in aqueous solution (120) within a container (110) absorb energy obtained by an energy source (130), whereby melanin, melanin precursors, melanin derivatives or analogues dissociate water molecules by dissipating the absorbed energy, whereby high energy electrons are transferred to a primary electron acceptor, and whereby melanin, melanin precursors, melanin derivatives or analogues are used the reverse reaction that comprises the union of atoms of hydrogen and oxygen generating water molecules and an electric current, characterized in that the energy source (130) is an inner energy source (130) within the container (110) and whereby the energy obtained from the energy source (130) is electromagnetic/ionizing radiation, preferably gamma radiation.

2. Process of claim 1 characterized in that the inner energy source (130) is an emitter (140) and/or emitter particles, in particular particles within the aqueous solution (120).

3. Process of claim 1 or 2 characterized in that the energy source (130) is from, but is not limited to the substance: titanium-44, Uranium 232, plutonium-238, nickel-63, silicon-32, argon-39, californium-249, silver-108, americium-241. Niobium-91, carbon-14, curium-245, niobium-94, plutonium-239, nickel-59, neptunium-236, uranium-233, technetium-99, uranium-234, chlorine-36, curium-248, aluminum-26, beryllium-10, zirconium-93, technetium-97, manganese-53, technetium-98, palladium-107, curium-247, uranium-236, niobium-92, plutonium-244, samarium-235, potassium-40, uranium-238, thorium-232, lutetium-176, rhenium-187, rubidium-87, lanthanum-138, samarium-147, platinum-190, barium-130, gadolinium-152, indium-115, hafnium-174, osmium-186, neodymium-144, samarium-148, cadmium-113, vanadium-50, tungsten-180, molybdenum-100, bismuth-209, zirconium-96, cadmium-116, selenium-82, tellurium-130, germanium-76, xenon-136, neodymium-150, calcium-48, cobalt-60 or tellurium 128 or a combination of at least two of the aforementioned substances, preferably in form of mixed oxide.

4. Process of any of the preceding claims characterized in that the energy source (130) is from enriched reprocessed substances, preferably substances of claim 3 that are enriched and reprocessed.
5. Process of any of the preceding claims characterized in that the temperature of the aqueous solution within the container is from 2°C to 45°C, preferably from 350°C to 500°C and particularly preferably close to 500°C.
6. Process of any of the preceding claims characterized in that the initial pH of the solution is variable from 2 or 3 to 8 or 9 units of pH, being the most used about 7.
7. Process of any of the preceding claims characterized in that the melanin conserve electricity like an accumulator or battery during a time and within certain limits.
8. Process of any of the preceding claims characterized in that during the process oxygen and hydrogen atoms and high energy electrons are built, and optionally OH, hydrogen peroxide or anion superoxide are generable.
9. Process of any of the preceding claims characterized in that the aqueous solution (120) is a suspension or a solution with water; natural or synthetic; with vegetal, animal or mineral origin; pure or mixed with organic or inorganic compounds, ions, drugs, metals like gadolinium, iron, nickel, copper, erbium, europium, praseodymium, dysprosium, holmium, chromium or magnesium, lead selenure; and with a concentration from 0.1% to 100%.
10. Process of any of the preceding claims characterized in that the melanin, melanin precursors, melanin derivatives, or analogues comprises polihydroxyindole, eumelanin, feomelanin, alomelanin, neuromelanin, humic acid, fullerens, graphite, polyindolequinones, acetylene black, pyrrole black, indole black, benzene black, thiophene black, aniline black, poliquinones in hydrated form sepiomelanins, dopa black, dopamine black, adrenalin black, catechol black, 4-amine catechol black, in simple linear chain, aliphatics or aromatics; or their precursors as phenoles, aminophenols, or diphenols, indole poliphenols, ciclodopa DHI Y DHICA1, quinones, semiquinones or

hydroquinones, L-tyrosine, L-dopamine, morpholin, ortho benzoquinone, dimorpholin, porphirin black, pterin black, ommochrome black, free nitrogen precursors, any of the above listed with any size or particles from 1 angstrom to 3 or 4 cm.

11. Cell (100) for generating an electric current comprising a container (110) in order to permit at least electromagnetic radiations to pass through, melanin, its precursors, variants, derivatives, or synthetic or natural analogues, which are mainly dissolved in an aqueous solution (120) within the container (110), an inner energy source (130) within the container (110), a cathode and an anode, characterized in that the inner energy source (130) emits electromagnetic radiation.

12. Cell (100) of claim 11 characterized in that the aqueous solution (120) including melanin, melanin precursors, melanin derivatives, or analogues is able to absorb the electromagnetic radiation.

13. Cell (100) of claim 11 or 12 characterized in that the inner energy source (130) is an emitter (140) and/or are particles from, but not limited to the substance: titanium-44, Uranium 232, plutonium-238, nickel-63, silicon-32, argon-39, californium-249, silver-108, americium-241, Niobium-91, carbon-14, curium-245, niobium-94, plutonium-239, nickel-59, neptunium-236, uranium-233, technetium-99, uranium-234, chlorine-36, curium-248, aluminum-26, beryllium-10, zirconium-93, technetium-97, manganese-53, technetium-98, palladium-107, curium-247, uranium-236, niobium-92, plutonium-244, samarium-235, potassium-40, uranium-238, thorium-232, lutetium-176, rhenium-187, rubidium-87, lanthanum-138, samarium-147, platinum-190, barium-130, gadolinium-152, indium-115, hafnium-174, osmium-186, neodymium-144, samarium-148, cadmium-113, vanadium-50, tungsten-180, molybdenum-100, bismuth-209, zirconium-96, cadmium-116, selenium-82, tellurium-130, germanium-76, xenon-136, neodymium-150, calcium-48, cobalt-60 or tellurium 128 or a combination of at least two of the aforementioned substances, preferably in form of mixed oxide.

14. Cell (100) of any of the preceding claims characterized in that the volume of the container is in a range from 1 microliter to 10 liters or to 20 liters.

15. Cell (100) of any of the preceding claims characterized in that a process of any of the claims 1 to 10 is operable in the cell (100).