Supercapacitor like structure for micro-battery and radiation energy harvesting tile

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ABSTRACT

The usage of hetero-nano-layers to harvest the energy deposited in ionization by the stopping particles and radiation passing through the layers produces very efficient and compact solid-state battery devices. The structure in its simplest forms looks like a planar capacitor made from a plurality of nano-strata composed from minimum 4 components in a repeated periodically. The first external element is the radiation generator that can be a alpha particle emitter such as ^{239}Pu, ^{241}Am or a beta emitter as ^{67}Ga, ^{3}T, or gamma source. The first layer from the source has high electronic density generating a shower of knock-on electrons leaving the layer through a thin dielectric leaving it polarized positively. The electron shower is stopped in a second low electronic density conductor nano-layer that polarizes negatively and separated by a low emittance dielectric from the next structure. This structure of conductor dielectric is repeated all along the radiation stopping range, converting almost all the radiation energy into electricity. For ^{238}Pu battery it may have a module thickness of 50 microns possible to deliver up to 50mW/cm^2. In other configurations the super-capacitor like harvesting structure may be used in a multi-radiation energy-harvesting tile for space applications.

1. INTRODUCTION

Miniature, high power density, long life batteries are needed in many fields, such as consumer electronics, medical instruments, field distributed sensors (SCADA), space electronics, etc. These batteries should have a volume on the order of a few mm^3,up to several cm^3 power density depending on material in the range of hundred of W/cm^3 to several mW/cm^3, lifetime extending to several months, or even tens of years, and the potential of being able to morph into micro-systems. These represent a new kind of miniature, high power, long lifetime, low radiation battery that directly converts the energy of alpha, beta even gamma particles into electric power. These batteries have to be very small (on the order of few tens of cm^2 by few square micrometers), produce minimal external radiation dose, and have high power density and long lifetime and safe operation.

1.1 State-of Art of the Existing Approaches

The interest in direct conversion of nuclear energy into electricity appeared in early 1940s .[1], from the invention of a device for measuring the intensity of radiation of slow neutrons by means of an ionization chamber. In 1946 Linder invented a thermo-ionic fission device [2] that, was in fact a modified ionization chamber. In 1948 a differential ion chamber was invented [3], which improves the Linder's chamber performances. A better connection of such assemblies was developed by 1953 [4] as a set of so called nuclear batteries. In 1955 [5] Wilson observed that in nuclear reactors a neutron fissionable isotope such as ^{233}U, ^{235}U, ^{239}Pu or mixtures thereof is subjected to fission by absorption of neutrons into a self-sustained chain reaction established by the neutrons evolved by the fission if the reactor is sufficiently large. Wilson also noticed that energies of the order of 200 MeV per fission are common. Of this energy perhaps 170 MeV represents kinetic energy, and about 30 MeV represents the energy of beta and gamma rays resulting from fission and fission products. About 7.5% or possibly about 10% of power is developed in neutron reactor in the form of beta rays. He developed more specific means to provide beta ray collection in close proximity to the fissionable bodies. The era of beta-voltaic batteries was initiated by this specific invention and that of Schwartz [6] which relates to nuclear batteries and more particularly to batteries utilizing fission products as well as neutron activated isotopes of a reasonable half-life which deliver negative beta-particles that is free electrons of high speed. A nuclear battery of this type comprises within a highly evacuated container, a solid beta emitter emitting negatively charged electrons and thereby acquiring a positive charge itself and a collector which is hit by the electron thereby charging the collector negatively. An electric field is thus being build up between emitter and collector. By 1958 the era of semiconductor usage in energy harvesting begins by the Schuyler's invention [7], who observed that in the previous arrangements only the primary charged particles are collected and the apparatus does not permit the use of neutral emission for voltage charging. According to his invention the neutral and/or charged particles high energy may be utilized to achieve a more efficient voltage charging. It was found that when two electrodes are brought into intimate contact which each other a potential barrier is established there between. Preferably one electrode is a metal and the secondary is a semiconductor. Because of the surface state of the semiconductor the potential barrier between the electrodes is thus enhanced. When this junction is exposed to radiation the potential created may be utilized to supply energy and current to a load circuit. By the 1970s the transistor era was at its apogee, and a new development of the pn junction was brought into direct conversion devices by the research of Hoff [8] Adler [9] and Yasuro [10]. Hoff disclosed an electron-voltaic semiconductor power source comprising a semiconductor body with a P-N junction terminating in a passivated channel on one surface of the device. A radioactive source with energy less than the radiation damage threshold of the semiconductor is used to generate carriers within the semiconductor body and the entire device is shielded with a metal casing formed on the device surface which also serves as electrical contacts for the device. Yasuro converted radioactive energy to electric energy by irradiating a converter body of semiconductor material etc. with radioactive rays to produce a number of electron-hole pairs in the converter, applying a magnetic field to the converter in a direction perpendicular to the direction of diffusion of the electron-hole pairs to separate the electrons and the holes in a direction perpendicular to the direction of diffusion of the electron-hole pairs and to the direction of application of the magnetic field and deriving the electrons and the holes from electrodes provided on the respective end faces of the converter body as electric energy. This is the period when use of magnetic fields to enhance the breakdown values of materials appears correlated with the Tokamak studies, which was further used in the DOE funded study [11]. Schenectady [12] discovered deep diode atomic battery made from a bulk semiconductor crystal containing three-dimensional arrays of columnar and lamellar P-N junctions. The battery is powered by gamma rays and x-ray emission from a radioactive source embedded in the interior of the semiconductor crystal. Van Dine develops a fabrication method [13] for directed energy conversion of semiconductor by the directed energy fusion of a selective region of semiconductor layer to provide a conductive path through the layer. Another approach is based on photoelectric effect and was introduced by Ritter [14]. He proposed a radioisotope photoelectric generator for use as high voltage source - comprising alternating electrodes of high and low atomic number materials activated by photon-emitting radioisotope This disclosure is directed to a radioisotope photoelectric generator for producing electrical energy. The construction of the generator is similar to that of a well-known storage battery. The generator is composed of alternate layers of high-Z, (high atomic-number) and low-Z (low atomic number) material that are insulated by vacuum or other insulating material. Low-energy photons from a radioactive source interact predominantly with the high-Z material by the photoelectric process, ejecting photoelectrons whose energy extends up to the incident gamma-ray energy “E”. By selecting the high-Z material thickness to be less than one electron range (at energy E) and the low-Z material thickness to be more than one electron range, there is a net electron transfer from the high-Z plates to the low-Z plates because electrons are emitted predominantly from the high-Z plates and stop in the low-Z plates. After start-up, a potential difference will build up between the high-Z and low-Z plates. An upper limit for this potential difference in kilovolts is the energy E in keV. The high-Z plates are connected together electrically and the low-Z plates are connected together electrically thus forming a "battery.

There is also a new trend in nuclear technology development stressed by Prof. Blanchard [15]: “Nuclear power may not be going away, as some activists might hope, but it is scaling down. Scientists are developing miniature devices that might soon be used in cars and to monitor health instead of fueling submarines and cities”.

Another approach [16] is based on a multi-layered graded band-gap heterostructure the composition of the continuous solid solution from SiO_2 up to Si changes very smoothly to reach a high degree of the scattering radiation coherence. Thus the radiation of any center of scattering is in a phase with external scattered radiation. Therefore external scattered radiation is included into resonant interaction with atoms and molecules of heterostructure. The authors sustain that the effective conversion of the radiation frequency and effective conversion of radioactive radiation to an electric current takes place.

The properties [16] of the graded band gap quasi-epitaxial hetero-structures are the following: – they possess a great range of the band gap width meanings;– the crystal lattice has smooth enlargements; – in the direction to the growing layer surface the concentration of the compound (binary or triple, having the higher melting point) which is a part of a multi-componential solid solution increases.

2. RESULTS AND DISCUSSION

The typical harvesting structure resembles a capacitor having the metal armatures made from different materials with different work functions such as the electrons released by the ionization process to produce polarization between the armatures. Fig. 1 shows the ionization energy deposited by a 5MeV alpha particle, specific to actinides decay, into a alternate structure formed by a high electron density conductor as gold, a silica insulator, a aluminum low electron density conductor armature and insulator, repeated all along the main radiation stopping range that is about 15 to 25 microns in this layers sequence. The exact value is depending on the specific dimensions of the layers that may be varied to balance the energy-harvesting rate. This is the case of the capacitor armatures connected in parallel to the external plots harvesting a voltage smaller than the smallest

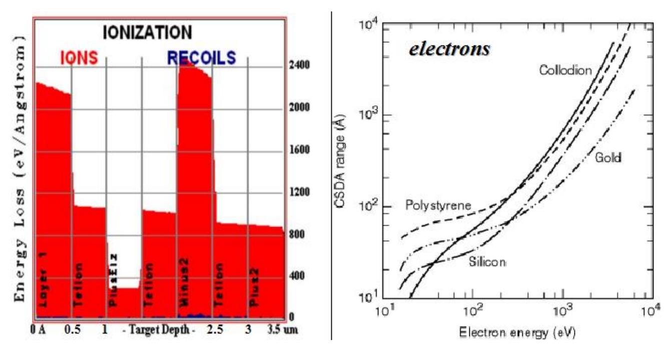

Fig. 1 – Principle of operation of the harvesting structure
Left – *The Radiation ionization energy in various materials*
Right –*The electrons path in various materials versus energy*

breakdown voltage of the insulators being in miliVolt range. The most intuitive direct conversion device looks mainly like a super mirror- or a heterogeneous super-capacitor. The issues associated to these devices are related to global conversion efficiencies and the stable operation lifetime in high radiation field.

2.1 Parallel versus Serial Internal Connection of the Capacitor's Armatures

Another alternative is to make an internal serial connection between successive armatures such as the polarization voltage to sum over the entire structure as Fig. 2 shows.
Following the morph of the serial capacitor structure Fig. 2a by eliminating the unnecessary insulator layer Fig.2b in

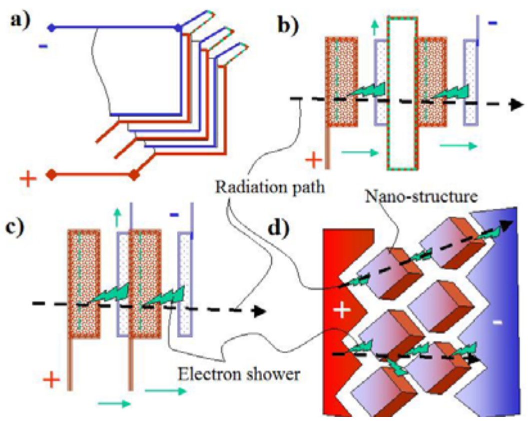

Fig. 2 – The serial connection evolution

shortcut by the electric connection of armatures drives to the simplified structure Fig. 2c having a bi-material armature separated by insulator. The optimal thickness as Fig.1-Right shows is in tenth of nm domain where the large planar nano-layers have increased instability in nuclear radiation, thermal and mechanical stress and naturally breaks into smaller pads. It is then recommended to change the fabrication design by making the final armatures robust and the internal bi-material armatures in stable pads with the dimensions smaller than 1 μm as in Fig. 4d. In this way the structure looks like a capacitor having a special dielectric, with metal-insulator junctions conferring special properties under nuclear irradiation.

2.2 Insulator optimization

Compared to the last patents in the field this structure offers a higher voltage less sensitive to production thickness fluctuations and voltage accumulations. The structure is able to deliver the power in the Volts domain compared to the flat layer approach that may generate mili-Volts only being extremely sensitive to voltage and thickness fluctuation driving to breakdown failures.

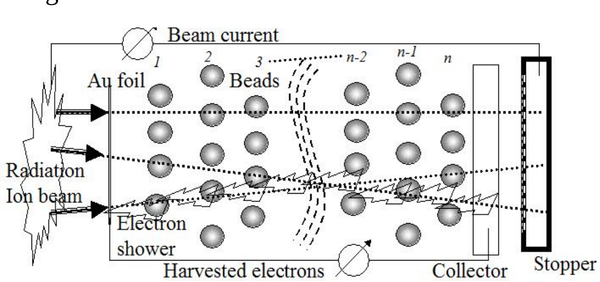

Fig. 3 Section through a supercapacitor with plasmonic insulator cell

The theoretical and simulation development drove to the usage of electron beam evaporation of gold and silica to form a suspension of nano-gold beads separated by several bead's radius from each other in amorphous silica insulator on an aluminum or gold thicker substrate covered by a gold thin layer to prepare a set of samples all having the structure, as shown in the figure below.
The ion beam generated by a radioactive source or accelerator passes through the suspension of beads that polarizes and is stopped in a stopper material. Silica will act as a low-Z dielectric separating the two armatures. The particle radiation will impact the structure from the gold side (high-Z, high electron density medium). While the gold recoils will be stopped mostly within the gold bead, with a very small fraction impacting the silica layer, most of the knock-off electrons will completely penetrate the silica layer, stopping almost entirely in the next beads. The result will be a positively charged gold foil armature opposite a negatively (or otherwise grounded) collector foil, separated by the polarized silica layers delivering the voltage into an external load.
Usual layered structures deteriorate and mix together as the fluence increases in the 10^{15} ions/cm^2 range, while this already annealed and nano-clustered metal-insulator is expected to have increased stability. The structure being annealed at high temperature and high ion beam dose is already stabilized, and less sensitive to thermal spike, a crossing nuclear particle leaves behind. As a detail, the thermal spike is strongly dimmed by the electron harvesting in the resonant plasmon structure created by the nano-grains of high electron density metals.
The amorphous silica insulator having nano-clusters of metals become a special material due to the increase in the weight of quantum and plasmon effects.
One interesting effect is the thermo-electric enhanced properties that may drive to electronic heat transport effects associated with nuclear radiation energy harvesting.
As a disadvantage such structure is sensitive to radiation displacement direction requiring the radioisotope deposition over the plus armature only limiting its mass, being integrated and encapsulated in the armature material.

2.3 From super-capacitors to ultra capacitors

A new enhanced structure shown in Fig. 4 is proposed in order to improve the radiation harvesting geometry and to allow less sensitivity to the radioactive material positioning. Gold loaded carbon nanotubes immersed in a LiH electrolyte is looking similar to the last developments in ultra-capacitors. The electricity is produced between the electrolyte pole and the nanotube center that also exhibits an increased capacitance in the domain of ultra-capacitors. These may be developed in encapsulated layers, or mili-tiles assembled in various thicknesses matching the dominant radiation stopping power. The specific ionization energy difference is increased between the nano-tube center of gold alloy exhibiting high ionization stopping power, similar to what Fig. 1 –left shows, while LiH is among the materials with the lowest stopping power. It also interacts with the carbon nano-tube structure behaving as a special semiconductor and offers an increased capacitance in the ultra-capacitors domain. The Monte-Carlo simulations predict harvesting energy conversion efficiency greater than 80%, with increased voltage stability due to high specific capacitance.

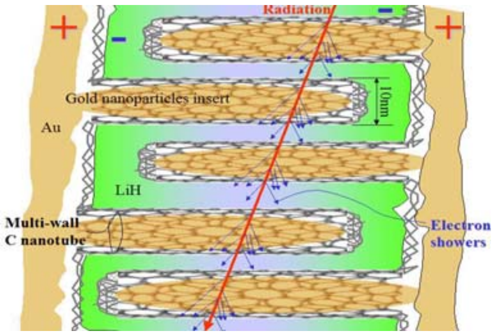

Fig. 4 – carbon nano-tube direct conversion advanced ultra-capacitor structure

Fig. 4 shows the ultra-capacitor designed structure achieved by connecting together the two substrates on which carbon nanotubes trapping gold nano-clusters or nanowires inside are connected together encapsulating the LiH electrolyte representing the negative electrode. To increase the effectiveness of current harvesting a conductive mesh may be added with the role of reducing the internal resistance and the inductance of the harvesting structure. This structure allows the increase of the weight of radioisotope uniformly distributed all around the harvesting cell growing its power.

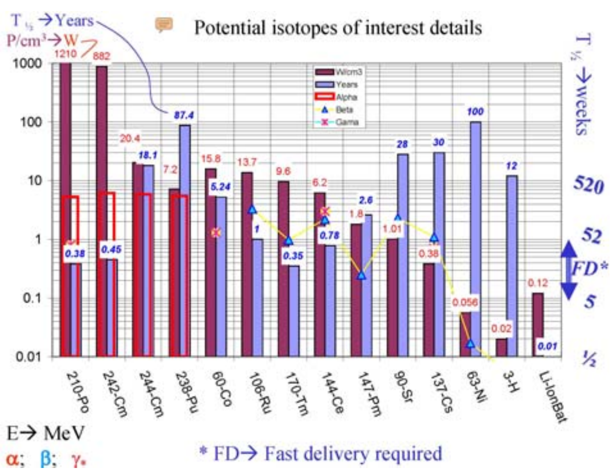

Fig. 5 – Potential powering isotopes performances

The chart in Fig. 5 shows the tradeoff between the radioisotopes lifetime and the specific power. If the ultra-capacitor harvesting structure is used up to 20% of the isotope specific power may be obtained driving to exceptionally high power densities and total energies delivered of about more than 10,000 times higher then the Li-Ion batteries can deliver. The ^{210}Po powered ultra-capacitor may rich up to 200 W/cm3 decreasing at 10% after 1.1 years, while if powered by ^{238}Pu delivers up to 4 W/cm3 decreasing at 50% after 87 years. The dimensions of these novel power sources span from several cubic microns to tenth of cubic centimeters. Special attention is given to operation safety the power supplies exhibiting minimal collateral radiation due to the specific features of the powering radioactive isotopes. The new miniaturized power sources are susceptible of modifying the future electronic design by introducing long life ripple free distributed power.

3. CONCLUSIONS

The nanotube ultra-capacitor structures may be successfully used to harvest radiation energy exhibiting high polarization independently of the radiation path and direction, susceptible of ultra high efficiency direct radiation conversion.

The high-efficiency nuclear radiation harvesting tiles have multiple potential applications as beam dumpers, radioactive high power micro-batteries, fission reactors based on direct conversion, fusion devices blankets, space beam propulsion and remote power applications.

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