3<sup>rd</sup> International Technical Meeting on Small Reactors

#### Direct Conversion of Fission Energy into Electricity in Liquid Gallium Contact Potential Cell

**T. Soh**<sup>1, 2</sup>

<sup>1</sup> Department of Mechanical Engineering, University of Saskatchewan. (Saskatoon, SK, Canada) (tes211@mail.usask.ca) <sup>2</sup> Sohtech R&D Inc., Saskatoon, SK, Canada

#### Abstract

Nuclear fission of uranium releases about 93% of its energy in the form of highly charged (up to 20+) and highly energetic fission fragment (83.5MeV average) and other ionizing radiations, including beta and gamma radiations. Liquid gallium is a semimetal that had been previously explored as a self recovering ionization media for alphavoltiac contact potential cell (CPC), and had been evaluated as a suitable replacement coolant for the next generation of fast reactor. By improving the neutronic aspect of a Liquid Gallium CPC (LGaCPC) with low neutron absorption construction materials, and by using a heterogeneous mixture of CANDU fuel grade uranium oxide powder (provided by CAMECO Inc.) and liquid gallium as its junction material, the direct conversion of fission energy to electricity has been observed when irradiated by the thermal neutron flux of the SLOWPOKE-II Research Reactor at the Saskatchewan Research Council. To further explore the effect of temperature on the operation of the LGaCPC, a High Temperature LGaCPC (HTLGaCPC), and a 6 meter high monolithic Large Volume Submersible Neutron Irradiation Chamber (LVSNIC) have been designed and constructed, which allow high temperature fission experiment up to 623K with a thermal neutron flux of  $1.18 \times 10^{10} \text{ n/cm}^2/\text{s}$ and the result is presented here, along with discussions on the operating principle of the LGaCPC, and on the construction and measurement techniques used in this study.

#### 1. Introduction

#### 1.1 Fission or fusion if it has to be small?

In the movie *Ironman*, a hypothetical portable nuclear fusion based 'Arc Reactor' provides gigawatt (GW) pulse to power myriad of propulsion and weapon systems. After 70 years with billions of dollars poured into a fusion reactor research, it is fair to suggest from a practical standpoint that a fusion based Arc Reactor will remain elusive, unless significantly practical progress, by order of magnitude, is made from the current research status.

A GW nuclear fission pulse is however readily available in a nuclear fission pulse research reactor (ex. General Atomics TRIGA). If there is a mean to directly convert this fission energy into electricity, at sufficiently high efficiency, a fission based 'Arc Reactor' is theoretical possible. While requirement in radiation safety and shielding will render such device not portable unless in unmanned applications, at even 1% conversion efficiency, the output from a 1GW

nuclear fission pulse will be 10MWe, a very respectable amount suitable for various devices. At 10% conversion efficiency, the 100MWe output will open an entire new vista in nuclear energy.

Such direct conversion technology is also directly applicable to power generating nuclear reactors to allow their miniaturization and better capturing a larger portion of fission energy as electricity.

This study uses a High Temperature Liquid Gallium Contact Potential Cell (HTLGaCPC) by subjecting it to the gamma flux and the thermal neutron flux of the SLOWPOKE-II Research Reactor at the Saskatchewan Research Council (SRC). A heterogeneous mixture of fuel grade uranium dioxide powder (provided by CAMECO Inc.) and liquid gallium is loaded into the HTLGaCPC, which is housed in a 6 meter high monolithic Large Volume Submersible Neutron Irradiation Chamber (LVSNIC) and is irradiated by a thermal neutron flux of 1.18 x 10<sup>10</sup> n/cm<sup>2</sup>/s. Ionization of the liquid gallium by both the reactor gamma flux and the fission fragments released by the uranium mixed with the liquid gallium generate Electron-Hole Pairs (EHPs) that are drifted by the electric field created by the contact potential difference between the zirconium and graphite couple and lead to an external measureable current.

The minimum critical mass of uranium-235 to sustain a chain reaction is known to be about 250g in polyethylene core reflected by beryllium.<sup>1</sup> The SLOWPOKE-II Research Reactor of Atomic Energy of Canada Limited (AECL) is designed based on this minimum and is optimized by a team lead by Dr. Hilborn and to this day still provides the highest stable thermal neutron flux  $(10^{12}n/cm^2/s)$  at the Inner Irradiation Site) available per mass of U-235 fuel used in all research reactors. By using the SLOWPOKE-II Research Reactor, the research reported here uses the smallest practical critical assembly available in the direct conversion of fission energy into electricity.

#### **1.2 Energy released by fission**

All power generating nuclear reactor converts the heat from nuclear fission to electricity, with the conversion efficiency limited by the thermodynamic Carnot Cycle. It is interesting to note that of the 210 MeV total instantaneous and delayed energies first released by the thermal neutron fission of U-235, 167 MeV (79.5%) is carried off by fission fragments, 21 MeV (10%) by gamma ray, 7 MeV (3.3%) by beta radiations from the fission products and these are all ionizing radiations<sup>2</sup>.

As these fission released ionizing radiations scatter and impinge on matter, they lose energy by ionization and generate EHPs in a non perfect process that produce heat as by-products. When the resultant EHPs recombine, the energy absorbed during their formation is also released as heat. The overall conversion process is from fission energy into heat but the energy of the intermediate EHPs can actually be harnessed with a non thermal conversion process, theoretically with above Carnot efficiency, to power an external load as in a photovoltaic battery.

# 1.3 Review of existing methods to directly convert nuclear radiation into electricity

The key to convert the cascading chain of ionization inside the reactor at high efficiency is to organize the myriad of random microscopic current element to line up their vectors so a macroscopic current can be measured. This is the operating principle of several types of nuclear batteries used to harness the electrical and ionizing nature of nuclear radiation without going through the heat cycle to directly convert radiation into electricity, although most of them are plagued by low efficiency or low working lifetime. New discoveries in material science and advancements made in the nuclear batteries technology in recent years overcome some of these hurdles and we are now closer than ever toward the direct conversion of nuclear fission energy to electricity.

### 1.3.1 Direct charge device - vacuum media

In 1957, G. Safanov recognized that the charged nuclear fragments released by the fission of uranium-235 or plutonium-239 carry can be converted directly into electricity.<sup>3</sup> For a fission fragment particle carrying 80 MeV of kinetic energy with a charge of 20<sup>+</sup>, a maximum potential of 4 MV can be attained. Direct charge device with a vacuum gap between the source and the collector is an electrical source of very high impedance (very high output voltage with very low output current) and is unsuitable to power conventional electrical device. Recent advance in this area includes magnetically insulated fission electric cells.<sup>4</sup>

# 1.3.2 p-n junction device - solid media

A p-n junction type nuclear battery has the advantage of lower impedance output. Similar to a hot carrier photovoltaic cell, inside a betavoltaic cell, a rectifying junction is created inside the device by the same principle and the electric field gradient across the junction sweep away all mobile charge carriers creating a depleted region across the device.

When a p-n junction is exposed to beta radiation<sup>5</sup>, a single beta particle creates many electronhole pairs (EHPs). If these EHPs are created within the depleted zone, under the influence of the electric field gradient, the electrons drift to the cathode and the ions drift to the anode and this gives out an amplified electrical current directly at lower output voltage. The reason that the current is amplified is because a single beta particle can ionize many atoms and in the process creates many EHPs, since the number of charge carrier increases, the current increases.

However, such betavoltaic p-n junctions devices have low working lifetime since the periodicity of crystal lattice of the junction material is destroyed by the energetic ionizing radiation and the semiconductor lose its desirable properties, which leads to the ultimate failure of the device in two ways. First, the charge carriers are increasing scattered by the atomic defects and recombines before providing useful work to the external load. Second, the depleted zone wherein the electron-hole pairs separates, lose its field as the lattice is destroyed and the electron-hole pairs again recombine without perform useful work.

# <u>1.3.3 Improvement of p-n junction using solid state radiation resistant and self-healing ionization</u> <u>media</u>

It was discovered that by using semiconductors with higher energy band gap like gallium arsenide or gallium phosphide, the resultant betavoltaic device will be more radiation resistance and perform for longer period. One of major breakthrough in material science recently for betavoltaic battery is the discovery and study of icosahedral boride<sup>6</sup>, a new type of radiation hard semiconductor possessing the unique properties of self healing.

For our purpose of directly converting fission energy into electricity, however, the major drawback of icosahedral boride is the very high thermal neutron capture cross section of boron (3800 barn) and being a compound semiconductor, its properties invariably will change with time when its components undergoes transmutation after capturing thermal neutrons. If there is a monoelemental self healing ionization media available with a much lower thermal neutron capture cross section area, it may be possible to harness it for our purpose (to be described further in this paper).

# 1.3.4. Contact Potential (CP) device - solid and liquid media

# 1.3.4.1. Formation of contact potential

The p-n junction is a popular but not the only method to form a rectifying junction that exerts a frozen electric field across a piece of semiconductor to give the radiation induced EHPs a fixed direction to travel. When two metals possessing different work functions,  $\phi_A$  and  $\phi_B$ , are placed in electrical contact, their Fermi energies equalize and create a contact potential,  $V_c$ , across the space between the two metals:

$$V_{c} = \phi_{B} - \phi_{A} \qquad (Eq. 1.3-1)$$

By applying this CP across an ionization medium such as silicon, the radiation induced electronhole pair can also be drifted and collected to do external work before recombination, as in the p-n junction.

# 1.3.4.2. Contact potential as radiation resistant depletion zone creation mechanism

The advantage of CP cell over p-n junction cell is that the CP generation mechanism is not degraded by energetic radiation to the same extent as in the p-n junction, and therefore it is more radiation resistant for generating and maintaining a constant electric field that guide the radiation induced EHPs. Dr. Paul Maurice Brown of Isogen Inc. has done extensive study in this field<sup>7, 8</sup>, and his work on  $\alpha$ -silicon co-doposited with tritum is followed by several studies done in Canada<sup>9, 10</sup> using p-i-n (p-type, intrinsic, n-type) betavoltaic configuration.

# <u>1.3.4.3. Contact Potential Cell using Liquid State Radiation Resistant Self Healing Ionization</u> <u>Media</u>

National Aeronautics and Space Administration (NASA) discloses a highly efficient alphavoltaic device<sup>11</sup>, consists of a radiation resistant Contact Potential Cell (CPC) loaded with semimetallic liquid gallium as a radiation resistant self healing ionization media. Unlike metals, semimetals have lower electrical conductivity and have both holes and electron as charge carrier. Semimetals have a *negative* band gap with their slightly empty energy band. This is in contrast to a *positive* band gap in semiconductor due to a slightly filled energy band.

NASA has claimed an energy conversion efficiency of 70% - 90%<sup>12</sup>. Unlike conventional solid state semiconductor, liquid gallium is *amorphous* and has no grain structure. When liquid gallium is bombarded by high energy ionizing radiation, the atomic displacement in gallium is not retained by the rigidity of a solid material and therefore leads to its self healing properties.

The use of self-healing liquid ionization media in nuclear battery has been further explored by University of Missouri-Columbia in Year 2009 using liquid semiconductive selenium-sulfur Schottky diode betavoltaic device<sup>13</sup>.

# 1.4 Nuclear applications of gallium

Gallium with its lower melting point, wide liquidus range, low fast neutron absorption properties, low toxicity and non flammability makes it attractive as an alternative heat transfer agent to sodium or lead for fast reactors<sup>14,15</sup>. These is important because if gallium can be used as a heat transfer agent in close contact with nuclear fuel without absorbing too much neutron, it should also be suitable as a carrier to allow nuclear fuel be mixed into it<sup>16</sup> and if placed in a CPC, the liquid gallium can be ionized by the nuclear fragment directly while sustaining a chain reaction.

Gallium remains liquid for the widest temperature range (302.92 K - 2478 K) of any metal and this allow unpressurized operation of nuclear reactor, which can lead to a major benefit in lowering the cost of nuclear reactor construction.

The alloys of uranium-gallium and plutonium-gallium are well studied<sup>17</sup>. In fact, gallium is used as an alloying agent to stabilize the  $\delta$  phase plutonium alloy in nuclear weapons<sup>18</sup>. At high temperature and low uranium concentration, a liquid Ga-U alloy is obtained. At higher concentration and lower temperature, a slurry of liquid Ga-U alloy and Ga<sub>3</sub>U is obtained.

# 1.5. Direct conversion of fission energy into electricity with LGaCPC

The use of LGaCPC to directly convert fission energy into electricity is an extension to nuclear battery technology discovered by NASA. Referring to Figure 1, when compare to the NASA device, inside the Direct Conversion Module (DCM), the alpha emitting radioisotope is replaced with the nuclear fission fragment released by the fissile element as the main energy source to ionize the gallium into EHPs, which are separated by the electric field applied across the gallium by the CP of the zirconium-carbon couple, which are chosen for their transparency to thermal neutron, and thereby allowing chain reaction to occur if many such elements are arranged into a lattice of suitable geometry. For this study which uses a single uranium oxide loaded LGaCPC, the liquid gallium is ionized by the external reactor gamma and internal fission fragment induced by the external hermal neutrons from core of the SLOWPOKE-II Research Reactor.

While there were similar fission studies done by Global Technologies Inc. sponsored by Defense Advanced Research Projects Agency (DARPA) using other self recovery ionization media<sup>19, 20</sup>, the details of those experimental method and results have not been made available to the public and the information provided in this paper should be of interest to researchers interested in the direct conversion of nuclear fission energy into electricity using liquid self healing junction materials.



**Figure 1:** Comparison of NASA US Patent# 6700298 (left) with the Direct Conversion Module (right). In DCM, both anode and cathode are thermal neutron transparent materials allowing free passage of thermal neutron through the DCM to sustain a chain reaction inside it with the fissile materials that has been incorporated into liquid gallium via alloying or intimate mixing (denoted by yellow spheres).

#### 2. Experimental

The experimental setup consists of 3 major components that will be described. The temperature range used is 323K-623K. The SLOWPOKE-II Research Reactor operates at full power at 16kW and the thermal neutron flux at the experimental site is determined to be  $1.18 \times 10^{10} \text{ n/cm}^2/\text{s}$  by activation method using a High Purity Germanium (HPGe) detector.

### 2.1 High Temperature Liquid Gallium Contact Potential Cell (HTLGaCPC)

To ensure identical thermal, electrical, and neutronic experimental conditions between the Measurement Cell (MC) and the Reference Cell (RC), the measurement assembly consists of two identical HTLGaCPCs built on a single block of nuclear grade graphite (MWI Inc.) and two 0.127mm thick 99.5% (metal basis) zirconium sheets (Alfa Aesar Inc.). As this experiment involve high temperature fission experiment, for safety reason, the internal volume of each cell is kept deliberately small to 0.34cm<sup>3</sup>.

The reason two HTLGaCPCs are needed is because in the MC, the liquid gallium can be ionized (and lead to an external current) by both the *internal* fission fragment within the UO<sub>2</sub>-Ga loaded MC induced by the external thermal neutron and the *external* reactor gamma field. To determine the net contribution of internal fission fragment generated current, a gamma ray compensation RC loaded only with liquid gallium is needed.

The MC is loaded with the  $UO_2$ -Ga composite consists of 0.311g of unenriched  $UO_2$  nuclear fuel powder (CAMECO Inc. Port Hope conversion facility) and molten gallium (0.989g Ga, 99.9999%, EMS Corporation) made by ultrasonic mixing. The RC is loaded with only gallium (1.981g Ga).

The graphite block with its two vertical slots acts as two crucibles connected in one body and contains the molten Ga in RC and the UO<sub>2</sub>-Ga composite in MC by gravitation mean. In each HTLGaCPC, a long zirconium foil supported by a silica insulated feedthrough is loaded from the top and the feedthrough is secured to the graphite block with aluminum screws with the joint sealed by graphite foil gaskets. The advantage of this arrangement is a gallium wetted joint is avoided as location of silica feedthough, by virtue of using a long zirconium foil, is high above the gallium wetted region. The MC and RC are connected to ground level instrumentation with colored coded insulated aluminum wires secured by aluminum fasteners.

### 2.2. Large Volume Submersible Neutron Irradiation Chamber (LVSNIC)

In the SLOWPOKE-II Research Reactor, the tubing leading to the various irradiation sites are constructed of aluminum alloy, which are prone to gallium corrosion. To mitigate all risks to these aluminum tubings, the HTLGaCPCs are mounted in a dedicated submersible thermal neutron transparent housing and inserted into the reactor pool to access the thermal neutrons and Referring to Figure 2a, a water tight LVSNIC is designed and built for this purpose. It consists of a 6 m high one piece assembly with a detachable cover both made of Al-T6061 alloy that places the HTLGaCPCs at the mid core position inside the Saskatchewan Research Council (SRC) SLOWPOKE-II Research Reactor pool to receive maximum thermal neutron flux while withstanding the hydrostatic pressure of a 5.25 m high water column.

Referring to Figure 2 b & 2c, the LVSNIC has a non load bearing gasketed flange and through it allow mounting of the HTLGaCPCs, which are thermally insulated to the aluminum body with ceramic standoffs. The aluminum mounting bracket is slotted to minimize thermal conduction and the neutron activation. The buoyancy of LVSNIC is carefully monitored by using *iProperties* function in the *Autodesk Inventor* software. As the LVSNIC is machined from 8" OD aluminum billet, the necessary sinking mass is made by preserving mass in the rear end of the LVSNIC, away from the reactor core to minimize neutron activation. LVSNIC has a foldable and detachable leg, which facilitate its insertion into the SLOWPOKE-II Research Reactor pool at Saskatchewan Research Council (SRC) Environmental Analytical Laboratories for thermal neutron irradiation (2d).

The thickness of Al-T6061 thermal neutron window on the cover is 5mm.

# 2.3. Temperature control and current measurement

The maximum working temperature of the Zr/C LGaCPC in a fission test is limited to about 673K primarily by the oxidation of zirconium without an inert atmosphere. Incidentally, this is about the operating temperature of CANDU and other reactors and any data in this temperature range will be useful in determining the merit of the Zr/C HTLGaCPC that is relevant to the nuclear industry. The experimental temperature is chosen to be 323K-623K to limit Zr oxidation and measurements are taken at 20K intervals.

A single ¼"OD x 2.5" long heater (McMaster-Carr Inc.) with internal K-type thermocouple is inserted laterally into the graphite block behind the MC and the RC to ensure identical temperature between them. The temperature is controlled by a variable transformer, which is found to provide better temperature control (+/- 0.3°C maximum) than PID method (+/- 3°C minimum) once dynamic thermal equilibrium is reached. The temperature is monitored by a J-KEM Model 210 Temperature Controller.

The current output of the HTLGaCPCs with and without thermal neutron irradiation are monitored by a Keithley 6485 Electrometer with a current measurement resolution of 10fA,

which is controlled and its data recorded by a computer via GPIB link. As the massive LVSNIC is activated by thermal neutron, the resultant Al-28 (half-life = 2.24 mins) will be the major neutron activation product and generate sufficient gamma ray and bremsstrahlung radiation field to ionize the liquid gallium. To minimize the contribution of Al-28 gamma to the measurements, Reactor-On and Reactor-Off measurements are only taken after an equilibrium time of 12 minutes (>5 half-lives) once the reactor has reach full power or turned off for the Al-28 to grow and decay to a more stable level of Al-28.

2014 November 5-7 Ottawa Marriott Hotel



**Figure 2** – 3D CAD representation of the completed LVSNIC with HTLGaCPCs installed (2a, left), and close up view (2b, left 2); actual device (2c, left 3); and the completed setup in the pool of the SRC SLOWPOKE-II Research Reactor (2d, right).

#### 3. Result & discussion

The sample size N is 100 for each current measurement  $I_{x,Reactor-y,T}$  with accuracy of 0.1% of the reading + 10nA. The area normalized current output results,  $i_{MC,gamma,T}$  and  $i_{MC,fission fragment,T}$  are obtained with following equations:

 $i_{\text{MC, gamma, T}} = i_{\text{RC, gamma, T}} = \frac{(I_{\text{RC, Reactor-on, T}} - I_{\text{RC, Reactor-off, T}})}{I_{\text{cell}} w_{\text{cell}} + 2(h_{\text{RC}} I_{\text{RC}} + h_{\text{RC, T}} w_{\text{cell}})}$ (Eq. 3-1)

$$i_{\text{MC,fission fragment, T}} = \frac{(I_{\text{MC, Reactor-on, T}} - I_{\text{MC, Reactor-off, T}})}{l_{\text{cell}} w_{\text{cell}} + 2(h_{\text{MC,T}} l_{\text{cell}} + h_{\text{MC,T}} w_{\text{cell}})} - \frac{(I_{\text{RC, Reactor-on, T}} - I_{\text{RC, Reactor-off, T}})}{l_{\text{cell}} w_{\text{cell}} + 2(h_{\text{RC,T}} l_{\text{cell}} + h_{\text{RC,T}} w_{\text{Cell}})}$$
(Eq. 3-2)

where T is temperature in K

 $i_{x,gamma,T}$  is the area (wetted by Ga) normalized current of the cell x (RC or MC) from external gamma at T, A/cm<sup>2</sup>  $I_{x,Reactor-y,T}$  is the current measured of the cell x (RC or MC) at Reactor condition y at T, A  $I_{cell}$  and  $w_{cell}$  are the length and width of the wetted graphite electrode in RC and MC, cm

 $h_{RC,T}$  is the height of Ga in RC in cm at temperature T,  $h_{RC,T} = \left[\frac{m_{Ga,RC}}{\rho_{Ga,T} * w_{cell} * l_{cell}}\right]$ , cm

 $m_{Ga,RC}$  is the mass of gallium in RC, g;  $\rho_{Ga,T}$  is the density of gallium at T,  $\frac{g}{cm^3}$ 

 $\mathbf{h}_{\mathrm{MC,T}} \quad \text{is the height of UO}_{2} + \text{Ga in RC in cm at temperature T, } \mathbf{h}_{\mathrm{MC,T}} = \left[\frac{\mathbf{m}_{\mathrm{Ga,MC}}}{\rho_{\mathrm{Ga,T}} * \mathbf{w}_{\mathrm{cell}} * \mathbf{l}_{\mathrm{cell}}} + \frac{\mathbf{m}_{\mathrm{UO}_{2},\mathrm{MC}}}{\rho_{\mathrm{UO}_{2},\mathrm{T}} * \mathbf{w}_{\mathrm{cell}} * \mathbf{l}_{\mathrm{cell}}}\right], \text{ cm}$ 

 $m_{Ga,MC}$  is the mass of gallium in MC, g;  $m_{UO2,MC}$  is the mass of UO<sub>2</sub> in MC;  $\rho_{UO2,T}$  is the density of UO<sub>2</sub> at T,  $\frac{g}{cm^3}$  i<sub>MC,fission fragment,T</sub> is the area (wetted by Ga) normalized current in of the MC from internal fission fragment at T, A/cm<sup>2</sup>

 $l_{cell}$ ,  $w_{cell}$  and have random error of 0.013mm.  $m_{Ga}$  and  $m_{UO2}$  has random error of 0.001g, The densities of gallium at T,  $\rho_{Ga,T}$ , are calculated with a 0.4% uncertainty at 95% confidence level<sup>21</sup>. Similarly, the densities of UO<sub>2</sub> are calculated with 1% uncertainty at 95% confidence level<sup>22</sup>. Uncertainty analysis on the Eq. 3.2 using propagating method leads to an uncertainty of almost 2 order of magnitude of the experimental results due to the correlated systematic uncertainties in  $w_{cell}$ , which dominates the sensitivity.

A more reasonable analysis replaces the variable  $h_{RC,T}$  and  $h_{MC,T}$  to lessen the exaggerated effect of  $w_{cell}$  on sensitivity with fixed values  $h_{RC,463K}$ ,  $h_{MC,463K}$  (mid experimental temperature) for the entire experimental temperature range and the resultant error is contributed by the volumetric thermal expansion of gallium and uranium oxide inside the HTLGaCPC, which is less than 2% with 1% uncertainty. Another source of error is the transient growth of Al-28 between reactor-on measurements of the RC and of the MC, and it is +1.8% maximum on the  $i_{MC,gamma,T}$  for a 4 minutes interval after the initial 12 minutes equilibrium time. Referring to Figure 3, the current collected by the HTLGaCPC has two distinct regions. In the first low temperature region, from 323K - 503K, the current collected is in the range of  $10^{-9} - 10^{-7}$   $\mu$ A/cm<sup>2</sup>. In the second high temperature region from 523K - 623K, the current collected increases noticeable to  $10^{-7} - 10^{-6} \mu$ A/cm<sup>2</sup>, peaking at 603K. Our observation that the current collection efficiency changes with temperature agrees with the information disclosed in the patent by GTI Inc. that there is an optimum operating temperature range for each ionization media employed (ex.: 503K-523K for liquid Se).

At temperature below 503K, liquid gallium has high surface tension and high contact angle with graphite and does not wet the graphite electrode<sup>23</sup>. As temperature increases, the viscosity and surface tension is reduced and the wetting of graphite occurs, which leads to the jump in output current observed. As temperature increases further from 523K, the density of gallium decreases, reducing the charge carrier density of gallium which leads to broadening of the collection volume (space charge region) around the electrodes and increases the amount of EHPs collected and therefore increases the output current. As the temperature increase from 603K, increased thermal scattering of charge carrier in gallium reduced the diffusion length of the minority charge carrier and the amount of EHPs collected, leading to a decrease of output current. This decrease of output current at high temperature is not believed to be due to oxidation of zirconium as the portion wetted by gallium remained submerged and protected by gallium. The output current recovers at lower temperature between 523K-583K, which will not occur had the zirconium oxide formed on the surface of the submerged zirconium, as any zirconium oxide film once formed will not decompose and remain intact at low temperature.



Figure 3: Contribution of internal fission fragment and external reactor gamma to the HTLGaCPC output from 323K-623K under thermal neutron flux of  $1.18 \times 10^{10}$  n/cm<sup>2</sup>/s.

While our result points to the ability of LGaCPC to directly convert fission energy into electricity and has acceptable current output per area, the output voltage is low, in the range of millivolt. Unlike conventional p-n junction device or contact potential cell employing semiconductor, the electrical resistance across Zr/C LGaCPC is low. The series resistance across the HTLGaCPC is less than 10  $\Omega$  and this contributes to low output voltage observed, in the range of milliVolt, which is in contrast to the contact potential difference of 0.95V expected based on the work function of the Zr ( $\phi = 4.05V$ ) and C ( $\phi = 5.0V$ ). As liquid gallium has high density of charge carrier, the electric field distribution in the gallium can be explained by the formation of a rectifying Schottky junction. The relatively high electrical conductivity of liquid gallium limits the depletion region thickness to micrometer range from the electrodes. Only the EHPs created in the vicinity of the depletion regions contribute to an external current.

Our data does not support the over 70% conversion efficiency (CE) claimed by NASA but is in line with ~ 1% figure in DARPA supported studies. It should be pointed out that CE of the earliest photovoltaic cells made by Bell Laboratories was also ~1% and inferring to the recent advances made in the large scale practical implementation of photovoltaic system, and that there is no theoretical limit that bound the direct fission-to-electricity CE to the current results obtained, we are confident that if more resources is allocated to this area, more favourable CE should be attainable.

#### 4. Conclusion

Nuclear fission of uranium releases majority of its energy in the form of energetic ionizing radiation, which can be converted to electricity by several non thermal means but their working life are limited by radiation damage. Liquid gallium has several desirable thermal, chemical and nuclear properties and is a monoelemental semimetallic self healing ionization media immune to fission fragments and gamma ray damage. We have constructed a HTLGaCPC with Zr/C couple using a heterogeneous mixture of CANDU fuel grade uranium oxide powder and liquid gallium as its junction material, housed in the LVSNIC, the direct conversion of fission energy to electricity has been observed when irradiated by the thermal neutron flux  $1.18 \times 10^{10}$  n/cm<sup>2</sup>/s at the SRC SLOWPOKE-II Research Reactor. The result indicates the current collection efficiency improves at higher temperature and for the experimental range studied, the optimum temperature range is from 543K- 623K. The conversion efficient is ~1% mainly due to the low electrical resistivity of liquid gallium.

### 5. Acknowledgement

It is only with the generous help and support of many people and organizations that this project is possible. The author would like to thank his supervisor Prof. Szpunar and all members of his Ph.D Advisory Committee (Prof. Bugg, Prof. Yuen, Prof. Pywell & Prof. Fotouhi), Debbie Frattinger and Dean Yurkowski of Radiation Safety Office, Prof Kasap, Tom Meyer and George Belev, Ken Jodrey and Dan Vessey of Machine Shop, of the University of Saskatchewan; Brenda Stanek, Jeff Zimmer, Dave Chorney, Jenna Smith-Windsor of Saskatchewan Research Council Environmental Analytical Laboratories for reactor access; Engin Ozberk and CAMECO Inc. for providing the uranium dioxide fuel. This project is supported by the Scientific Research & Experimental Development Program.

### 6. References

[1] George A. Jarvis and Carroll B. Mills, "Critical mass reduction", *Los Alamos Scientific Laboratory of the University of California Report LA-3651*, Los Alamos, New Mexico, 1966 [2] P.61, "Department of Energy Fundamentals handbooks-Nuclear physics and reactor theory-atomic and nuclear physics (Volume 1 of 2)", DOE-HDBK-1019/1-93

[3] G. Safanov, "Direct Conversion of fission to electric energy in low temperature reactors", Rand Research Report, No. RM-1870, 1957.

[4] S. A. Slutz, L. C. Brown, "Magnetically insulated fission electric cells for direct energy conversion", *Physics of Plasma*, Vol. 10, #7, 2003, p.2983-2993.

[5] L. C. Olsen, "Betavoltaic energy conversion", *Energy Conversion*. Vol. 13, Pergamon Press, Great Britain, 1973, p. 117-127.

[6] Terrence L. Aselage, "Beta cell device using icosahedral boride compounds", United State of American Patent Number: 6479919 B1, 2002.

[7] Paul Brown, "Radioisotopic energy conversion utilizing a solid-state contact potential difference cell," 12<sup>th</sup> <u>Symposium Space Nuclear Power and Propulsion-Section A6</u>, American Institute of Physics, Jan 8-12, 1995, New Mexico, USA, pp. 439-450.

[8] Paul Brown, "Contact potential difference celle", United State of American Patent Number: 5087533, 1992.

[9] T. Kosteski et al., "Tritiated amorphous silicon films and devices", *J. Vac. Sci. Technol. A 16.2*, American Vacuum Society, Mar/Apr 1998, p. 893-896.

[10] Liu et al., "Betavoltaics using Scandium Tritide and Contact Potential Difference", *Applied Physics Letter*, Vol. 92, American Institute of Physics, 2008, p.083511

[11] Snyder et al., "Extremely efficient, miniaturized, long-lived alpha-voltaic power source using liquid gallium", US Patent Number: 6700298 B1, 2004.

[12] "Alpha-Voltaic Sources Using Liquid Ga as Conversion Medium", *Tech Brief*, NASA Jet Propulsion Laboratory, Pasadena, California, USA, July 2006.

[13] Wacharasindhu et al., "Radioisotope microbattery based on liquid semiconductor", *Applied Physics Letters*, Vol 95, 014103, 2009.

[14] V. Ya. Prokhorenko et al., "Liquid gallium: Potential uses as a heat-transfer agent", *High Temperature*, Vol. 38, No. 6, 2000, p. 954–968. Translated from Teplofizika Vysokikh Temperatur, Vol. 38, No. 6, 2000, p. 991–1005.

[15] T. Sawada et al., "Gallium-cooled liquid Metallic-fueled fast reactor", *Progress in Nuclear Energy*, Vol. 37, No. 1-4, p. 313-319, 2000.

[16] Walter D. Wilkinson, "Uranium gallium alloys and method of preparation", US Patent #3193380, 1965.

[17] Massaski (Editor) et al., *Binary alloy phase diagrams*, ASM International, Material Park, Ohio, USA, 1990, p.1872.

[18] Kaye Allan Johnson, "Homogenization of gallium-stablized delta-phase plutonium", *Los Alamos Scientific Laboratory of the University of California Report LA-2889*, Los Alamos, New Mexico, 1964.

[19] Samuel K. Moore, "Cold Fission", IEEE Spectrum, Issue February 2007., p.16

[20] Francis Yu-Hei Tsang, et al., "Nuclear voltaic cell", US Patent Number: 8073097 B2, 2011 [21] V. P. Chentsov, et al., "Density and surface tension of heavy liquid-metal coolants: gallium and indium", Materials of Power Engineering and Radiation-Resistant Materials, Vol. 2. No. 5, 2011, p.46-52.

[22] S. G. Popov, et al., Thermophysical properties of MOX and UO2 fuel including the effects of irradiation, ORNL/TM-2000/351, 2000, p.9

[23] Hideto Kamiyabu, et al., "Wettability and supercooling phoenomena of Ga", Memoir of the Faculty of Engineering, Okayama University, Vol. 23, No., 2, March 1989, p.5