

Energy-Recovery Linacs for Commercial Radioisotope Production - Phase I

Final Report for STTR Project Starting 03/15/2015, ending 12/15/2015

Small Business: Muplus, Inc.
45 Jonquil Lane, Newport News, VA 23606

Muplus, Inc. PI: Dr. Rolland P. Johnson

Research Institution: Thomas Jefferson National Accelerator Facility
12000 Jefferson Ave., Newport News, VA 23606

Research Inst. PI: Dr. Geoffrey Krafft, under CRADA JSA-2015S006

Subcontractor: Niowave, Inc., 1012 N. Walnut St., Lansing, MI 48906

Subcontractor PI: Dr. Chase Boulware

Approved for public release; further dissemination unlimited. (Unclassified Unlimited)

PREPARED FOR THE UNITED STATES DEPARTMENT OF ENERGY

Work Performed Under grant DE-SC0013123

SBIR/STTR RIGHTS NOTICE

These SBIR/SITR data are furnished with SBIR/SITR rights under Grant No DE-SC0013123 and subgrant JSA-2011S009. For a period of four (4) years after acceptance of all items to be delivered under this grant, the Government agrees to use these data for Government purposes only, and they shall not be disclosed outside the Government (including disclosure for procurement purposes) during such period without permission of the grantee, except that, subject to the foregoing use and disclosure prohibitions, such data may be disclosed for use by support contractors. After the aforesaid four-year period, the Government has a royalty-free license to use and to authorize others to use on its behalf, these data for Government purposes, but is relieved of all disclosure prohibitions and assumes no liability for unauthorized use of these data by third parties. This Notice shall be affixed to any reproductions of these data in whole or in part. (End of Notice)

ABSTRACT

Most radioisotopes are produced by nuclear reactors or positive ion accelerators, which are expensive to construct and to operate. Photonuclear reactions using bremsstrahlung photon beams from less-expensive electron linacs can generate isotopes of critical interest, but much of the beam energy in a conventional electron linac is dumped at high energy, making unwanted radioactivation. The largest part of this radioactivation may be completely eliminated by applying energy recovery linac technology to the problem with an additional benefit that the energy cost to produce a given amount of isotope is reduced. Consequently a Superconducting Radio Frequency (SRF) Energy Recovery Linac (ERL) is a path to a more diverse and reliable domestic supply of short-lived, high-value, high-demand isotopes at a cost lower than that of isotopes produced by reactors or positive-ion accelerators. A Jefferson Lab approach to this problem involves a thin photon production radiator, which allows the electron beam to recirculate through rf cavities so the beam energy can be recovered while the spent electrons are extracted and absorbed at a low enough energy to minimize unwanted radioactivation. The thicker isotope photoproduction target is not in the beam. MuPlus, with Jefferson Lab and Niowave, proposed to extend this ERL technology to the commercial world of radioisotope production. In Phase I we demonstrated that 1) the ERL advantage for producing radioisotopes is at high energies (~100 MeV), 2) the range of acceptable radiator thickness is narrow (too thin and there is no advantage relative to other methods and too thick means energy recovery is too difficult), 3) using optics techniques developed under an earlier STTR for collider low beta designs greatly improves the fraction of beam energy that can be recovered (patent pending), 4) many potentially useful radioisotopes can be made with this ERL technique that have never before been available in significant commercial quantities. We developed a plan for the Phase II project that started with a Conceptual Design Report (CDR) based on the results of the Phase I studies and concluded with a Technical Design Report (TDR) for a facility to make isotopes that are most attractive based on market analyses.

Phase I Final Technical Report

Energy-Recovery Linacs for Commercial Radioisotope Production

Table of Contents

Table of Contents	3
Project Overview	4
Identification and Significance of the Problem or Opportunity	4
Technical Approach	5
Concept	5
Available isotopes and radiator materials	7
Comparison to conventional linac	8
Interaction of beam with radiator	9
Beam parameters at radiator	10
Non-linear compensation after radiator	12
Beam collimation	14
Radiator design	15
Solid radiator	15
Liquid radiator	15
Jefferson Lab SRF Energy Recovery Linac Expertise	17
Niowave, Inc. Commercialization Expertise	17
MuPlus/Muons, Inc. Simulation Expertise and MuSim Commercialization	18
Anticipated Public Benefits	18
Presentation	19
Participants	19
Patent	19
Conclusions	20
References	20

Project Overview

Photonuclear reactions using bremsstrahlung photon beams from electron linacs can generate isotopes of critical interest. An SRF Energy Recovery Linac (ERL) is a path to a more diverse and reliable domestic supply of short-lived, high-value, high-demand isotopes at a cost potentially lower than that of isotopes produced by reactors or positive-ion accelerators. While the specific activity of a produced isotope for photonuclear reactions is not so high (≤ 1 Ci/g), the efficiency of production is considerably higher with gamma rays than for protons and neutrons. ERLs are increasingly the technology of choice for highly demanding high average beam power applications. In energy recovery, more than 90% of the beam power can be recycled and not deposited in a beam dump. Therefore, the energy of the waste beam can be lower than the threshold for neutron production and the activation of shield components, thereby reducing both complexity and cost. An Electron Linac combined with ERL technology has a number of advantages compared to reactors or other accelerator sources of isotopes as discussed below in the technical approach section.

MuPlus, in partnership with Jefferson Lab and Niowave, proposes to transfer ERL technology pioneered and developed at Jefferson Lab [1] to the commercial world of radioisotope production for medical diagnostics and therapy. A recent Jefferson Lab approach to this problem involves a thin photon production target, which allows the electron beam to recirculate through rf cavities so the beam energy can be recovered while the spent electrons are extracted and absorbed at a low enough energy to minimize unwanted radioactivation. In Phase I, we used our developed codes (MuSim-MCNP6, G4beamline-GEANT4) and others that we are very familiar with (ANSYS, ACE3P, CST, ...) to explore and optimize beam parameters of a radioisotope production facility based on an ERL with a photon radiator. This includes photon and electron beam parameters, absorbers for scattered electrons, target and radiator cooling, beam-radiator interactions, radiator optimization, thermal distributions and power handling, management of energy spread and angular acceptance for the recirculation arc, and optimization of isotope production versus energy recovery requirements. We will make choice of a particular isotope to be a first example of this new technology, and we will further explore this concept by a conceptual design report to be made in Phase II. The resulting design can be tested at the Jefferson Lab's FEL or, if the investment funds are available, built and operated by our Niowave partners at their facility in Lansing, MI. By the end of Phase II we will have a technical design report that includes engineering designs and cost estimates for a production facility. The project combines the design and simulation expertise of MuPlus, the ERL experience and capabilities of JLab, and the unique commercialization experience of Niowave to use superconducting rf for radioisotope production.

Identification and Significance of the Problem or Opportunity

Most radioisotopes are produced by nuclear reactors or positive ion accelerators, which are expensive to construct and to operate. Photonuclear reactions using bremsstrahlung photon beams from less-expensive electron linacs can generate isotopes of critical interest, but much of the beam energy is lost in making unwanted radioactivation. A Superconducting Radio Frequency (SRF) Energy Recovery Linac (ERL) is a path to a more diverse and reliable domestic supply of short-lived, high-value, high-demand isotopes at a cost lower than that of isotopes produced at reactors or positive-ion accelerators. A Jefferson Lab approach to this problem involves a thin photon production target, which allows the electron beam to recirculate

through rf cavities so the beam energy can be recovered while the spent electrons are extracted and absorbed at a low enough energy to minimize unwanted radioactivation. MuPlus, in partnership with Jefferson Lab and Niowave, proposes to extend the ERL technology pioneered and developed at Jefferson Lab to the commercial world of radioisotope production for medical diagnostics and therapy.

Technical Approach

Concept

Recent technological advances, especially in the fields of diagnostic and therapeutic medicine, require ever-increasing quantities of radioactive isotopes. These are produced by a limited number of large facilities yielding a small variety of isotopes. A direct consequence is that research and development in certain areas has stagnated because of the issues with production, transportation and economies of scale. Numerous industrial sectors would benefit from a compact, efficient, clean source of isotopes that is geographically close to the point of use, so as to take advantage of shorter half-life variants.

Most isotopes in use today are generated from reactors or cyclotron accelerators. Isotopes produced in reactors are mainly from the neutron, gamma (n, γ) reaction (radiative capture). By contrast, cyclotrons bombard a target with a stream of heavy, charged particles (commonly protons). Electron linacs, as being proposed here, use a radiator to produce bremsstrahlung photon beams which interact with a target which creates isotopes of interest (Figure 1). Bremsstrahlung or “braking radiation” is produced as charged particles (electrons) are slowed down in their interaction with matter (the radiator).

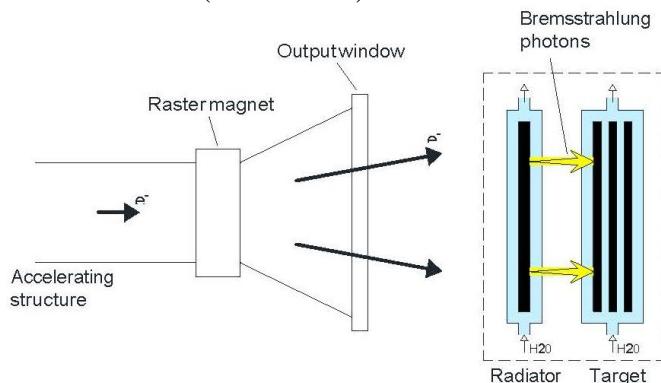


Figure 1: Schematic layout of a photonuclear reaction in a traditional electron accelerator.

Electron linacs offer unique advantages over traditional techniques. They are reasonably simple devices to operate and maintain. JLab has extensive experience built up over more than a quarter century in the design, fabrication and operation of SRF electron linacs. JLab is uniquely positioned to provide an alternative to more traditional production techniques using reactors or high-energy positive ion accelerators. This new and innovative approach using ERL technology has a number of advantages:

- Electron machines have the potential for higher production yields for certain isotopes compared to more traditional methods. For example, mCi quantities of ^{67}Cu may be generated by exposing Zn targets for several minutes.

- Separation of a desired isotope after production is simplified.
- Electron beam energies can be ‘tuned’ to optimize production of a certain isotope.
- Energy recovery means greater overall (wall plug) efficiency.
- Energy recovery allows a reduction in beam energies lost to beam dumps, to below the threshold of neutron production.
- Electron linacs are reasonably simple devices to operate and maintain. A production facility could be run by a relatively small group of technicians.
- Unlike a reactor, the machine could be powered down (instantly).
- End of life decommissioning is simpler, cleaner and less costly than other technologies.
- Smaller machine footprint allows flexibility in the placement and location of production facilities.

Figure 2 shows a potential ERL-based isotope production facility schematically. This proposal looks at consolidating a number of technologies to create a machine unrivalled in performance and flexibility. Currently, few options exist for getting isotopes to where they are needed. Isotope selection is also limited, because the most useful isotopes often have short half-lives, making transportation a problem. Envisage a facility that can be built in a hospital basement or in a distribution center within an hour or two of tens of hospitals and research establishments. The availability of new isotopes would open a frontier of innovative research and treatments and would help drive down production costs.

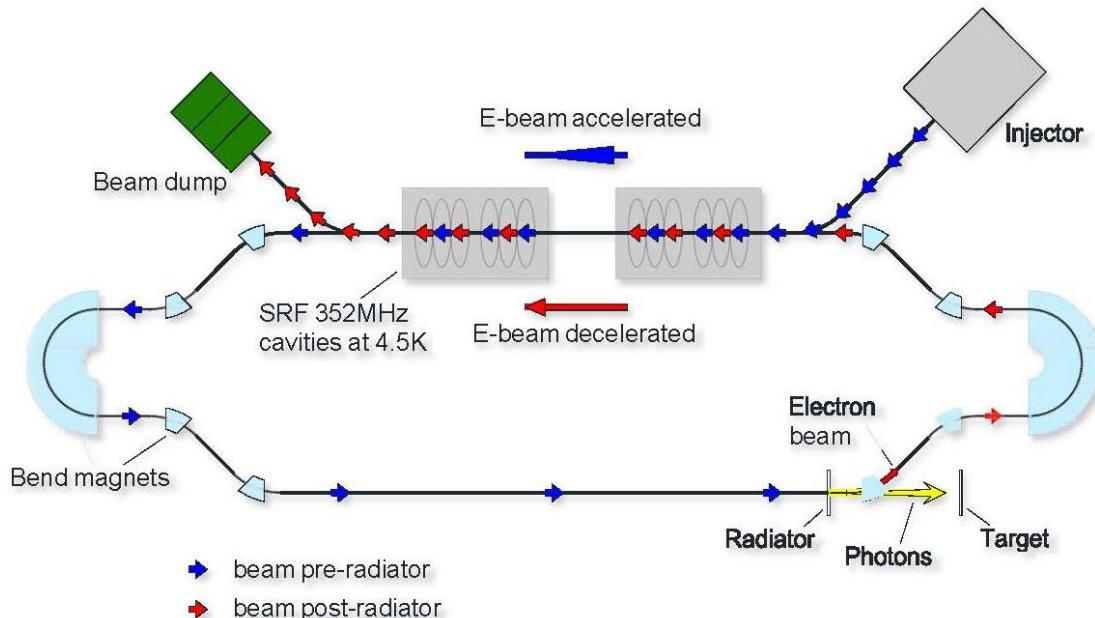


Figure 2: A schematic layout of a future ERL-based isotope production facility. The injector produces a CW electron beam which is accelerated in multiple 4.5 K SRF cavities. The beam passes through a radiator creating bremsstrahlung photons, which in turn hit a target for radioisotope production. The remaining electrons are sent back through the SRF cavities, 180° out of phase and their energy is recovered before being extracted to the beam dump.

Figure 2 shows an ERL’s main components. It begins with an injector, which provides the electron beam bunches, typically by a laser-driven photocathode. This beam is then accelerated by one or more SRF cavities. Typical accelerating gradients are 10–15 MV/m.

The cavities are submerged in a helium bath within a cryomodule. The beam is steered and focused with magnets and beamline components until it is delivered to the target apparatus. In our case, the e-beam would pass through a radiator, with around 99% of the beam passing straight through. Instead of sending this beam to a dump (and wasting its energy and causing activation), the spent beam is steered back through the cavities, 180° out of phase, so that it is decelerated and the energy returned to the RF structure, ready to accelerate the next bunch that passes through.

Available isotopes and radiator materials

The optimum electron beam energy for each particular isotope depends on its production rate, and in particular, on the reaction threshold energy. The most common photonuclear production channels are (γ,n) and (γ,p) with a threshold of about 10 MeV. However, some isotopes can only be produced via higher order channels, such as $(\gamma,2n)$, $(\gamma,3n)$, and (γ,np) . These reactions have higher energy threshold and the optimum electron beam energy for production of such isotopes can reach 80-100 MeV. Table 1 summarizes some of the isotopes which can be produced using a 40 MeV and a 100 MeV linac.

Table 1: Some of the isotopes which can be produced using an energy recovery linac.

Radioisotope	Reaction	Threshold, MeV
V-48	V-51($\gamma,3n$)V-48	32
	Cr-50(γ,np)V-48	21
Cr-48	Cr-50($\gamma,2n$)Cr-48	24
Zn-62	Zn-64($\gamma,2n$)Zn-62	21
Co-56	Ni-58(γ,np)Co-56	20
Cs-136	Ba-138(γ,np)Cs-136	16
Sm-145	Sm-145($\gamma,2n$)Sm-147	20
Ag-111	Cd-113(γ,np)Ag-111	16
Sc-47	Ti-48(γ,p)Sc-47	11
Y-88	Y-89(γ,n)Y-88	11
Cu-67	Zn-68(γ,p)Cu-67	10

The isotopes shown in Table 1 have numerous applications in research and industry. For example, vanadium-48 is a high energy gamma-emitter (984 keV and 1312 keV), which has both medical and non-medical uses. Its half-life of 16 days makes it suitable for investigation of biochemical and molecular actions of vanadium compounds which were shown to provide protection against all stages of carcinogenesis – initiation, promotion, and progression. Vanadium-48 also a positron emitter and has been suggested as an alternative for the Ge-68 to calibrate PET instruments (Hichwa et al, 1995). V-48 can be produced by $^{48}\text{Ti}(p,n)$ ^{48}V or $^{45}\text{Sc}(\alpha,n)$ ^{48}V . However, using ERL can be an alternative to the aforementioned production methods.

One of the common quality criteria of the produced isotopes is high specific activity. In many cases, especially for medical applications, the final product also has very stringent purity requirements. Thus it is often necessary to separate the produced isotope from the target material.

If the product and the target are chemically different species (for example ^{50}Cr and ^{48}V), this process requires a radiochemical facility. Niowave is currently setting up a radiochemistry lab in its R&D facility in Lansing, MI and will be willing to assist developing radioisotope separation and purification processes to start supplying certain radioisotopes to our customers.

Comparison to conventional linac

Choice of the radiator thickness is critical. On one hand, the radiator must be thick enough to provide a high photon flux in comparison with a conventional straight-through linac. On the other hand, it must be thin enough that the beam quality after the radiator is adequate for energy recovery. We first consider the energy efficiency aspect of using an ERL for isotope production. To fully realize ERL potential, it is better to use a relatively high-energy (~100 MeV) high-current (~10 mA) electron beam.

To compare the efficiencies of the ERL and one-pass linac, we considered a 100 MeV electron beam incident on a 0.25 mm thick tungsten converter. The ratio of yields per electron per unit converter thickness in an ERL (0.2 $\gamma/\text{e/mm}$) and in a one-pass linac (0.09 $\gamma/\text{e/mm}$) is

$$R = \frac{Y_{ERL}}{Y_{conv}} = 2.2 \quad (1)$$

The yield in a one-pass linac was calculated assuming an optimal 3 mm converter thickness. It gives maximum photon production per electron but already causes some attenuation of the photon beam. That is why the yield normalized to unit converter thickness is significantly lower than from a thin radiator in an ERL, which provides virtually no attenuation of the photon beam. Now, let's consider losses during energy recovery and their effect on the efficiency of the ERL for a photonuclear reaction with $E_{th} = 40$ MeV. Assuming that 3 MeV is not recovered from a 100 MeV beam:

$$\frac{V_{dump}}{V_{ERL}} = \frac{3MV}{100MV} = 0.03 \quad (2)$$

The ratio of thicknesses of tungsten converters for ERL (250 μm) and one-pass linac (3 mm) is given as:

$$\frac{t_{ERL}}{t_{conv}} = \frac{250}{3000} \quad (3)$$

The ratio of currents necessary to produce the same amount of activity using ERL and one-pass linac can be found from the ratio of thicknesses and ratio of yields:

$$\frac{I_{conv}}{I_{ERL}} = \frac{t_{ERL}}{t_{conv}} \cdot R = \frac{2.2 * 250}{3000} \approx 0.183 \quad (4)$$

The RF power one needs for the ERL (assuming 100% recovery and discounting the power that goes into x-rays) is:

$$P_{ERL} = I_{ERL} V_{dump} \quad (5)$$

And the RF power one needs for the one-pass linac at the same 100 MeV beam energy is:

$$P_{conv} = I_{conv} V_{conv} \quad (6)$$

If the power requirement are identical to produce the same amount of high energy photons, their ratio should be equal to unity, however, in our case it is:

$$\frac{P_{ERL}}{P_{conv}} = \frac{I_{ERL}V_{dump}}{I_{conv}V_{conv}} = \frac{0.03}{0.183} \approx 0.16 \quad (7)$$

Thus to produce an equivalent activity, ERL requires about one fifth of the power. Even if we take the recoverable particle fraction (93%) into account, the ERL is still going to be competitive with a one-pass linac. In this case:

$$P_{ERL} = \frac{I_{ERL}V_{dump}}{0.93} \quad \text{and} \quad \frac{P_{ERL}}{P_{conv}} = \frac{0.03}{0.93 \times 0.183} \approx 0.17 \quad (8) \text{ and } (9)$$

Interaction of beam with radiator

The interaction of the electron beam with the photon radiator was simulated using GEANT4/G4Beamline via MuSim to optimize the balance between energy recovery and isotope production. The linear optics upstream of the radiator was designed to obtain an electron beam waist at the radiator position. A 3D render of the simulation setup is shown in Fig. 3.

Different radiator materials were simulated to compare the photon fluxes from comparable radiator thicknesses. Tungsten and lead-bismuth eutectic (LBE) were simulated as representative materials for solid and liquid radiators, respectively. The relevant parameter for quantifying the radiator thickness is the ratio of the thickness to the material radiation length. For small thickness ratios (~few percent radiation length), the ratio corresponds to the average energy lost in the electron beam after passing through the radiator. This average energy loss is integrated over the entire electron energy distribution, however, and the peak of the distribution is more favorable for energy recovery than is implied by the total loss percentage. This characteristic of the electron energy distribution is shown in Fig. 4 (left) for 100 MeV/c electrons passing through a 3.5% (250 μm) LBE radiator. The distribution extends down to 0 MeV/c but is truncated here at 95 MeV/c. The average momentum of surviving electrons is 96.4 MeV/c, but the distribution peaks at 99.8 MeV/c with a full width at half maximum (FWHM) of ~0.1 MeV/c.

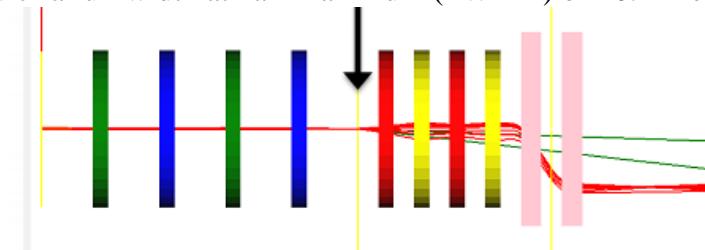


Figure 3: 3D render of simulation setup in MuSim. The arrow indicates the radiator position. Several electron (red) and photon (green) tracks are also plotted.

The rms angular spread due to multiple scattering is given by Eq. (10) [2]. For a 100 MeV/c electron beam, the induced angular spread depends only on the radiator thickness. For radiator thicknesses of ~3%, the angular spread due to multiple scattering is 20 mrad. The allowable angular spread in the spent electron beam depends on the size of the emittance that is tolerable in the downstream sections of the beamline as the spent beam is prepared for energy recovery.

$$\theta_{rms} = \frac{13.6}{E[\text{MeV}]} \sqrt{\frac{w}{X_0}} \left(1 + 0.038 \ln \frac{w}{X_0}\right), \quad (10)$$

For a 3.5% LBE radiator, the total number of photons generated per incident electron and photon fluxes for several energy ranges are listed in Table 2. The photon fluxes count all photons incident on a 1 cm diameter disk that is 1 m downstream of the radiator.

Table 2: Photon production by 100 MeV electron beam in a 0.25 mm thick LBE radiator.

	E_γ [MeV]	N_γ/N_e	Φ [$10^{15} \gamma/\text{cm}^2/\text{s}$]
3.5% (0.25 mm) LBE radiator	> 10	0.077	2.9
	> 20	0.049	1.9
	> 40	0.025	0.94

Let us now consider the possibility of beam energy recovery after the radiator. The average energy loss after a 250 μm LBE radiator is about 3.5%. By itself, this number is not challenging. For example, momentum acceptance of more than 10% has been demonstrated in JLab's FEL. Moreover, as shown in Fig. 4 (left), the peak of the distribution is much narrower. However, multiple Coulomb scattering in the radiator also introduces a large angular spread. It completely dominates over the initial angular spread of the beam. Managing this angular spread is the main challenge from the beam dynamics point of view. Assuming an angular acceptance of ± 100 mrad (plane-projected) and momentum acceptance of 15%, as illustrated in blue in Fig. 4 (right), the recoverable fraction of the beam is about 93%. We briefly demonstrate below that these numbers are reasonable.

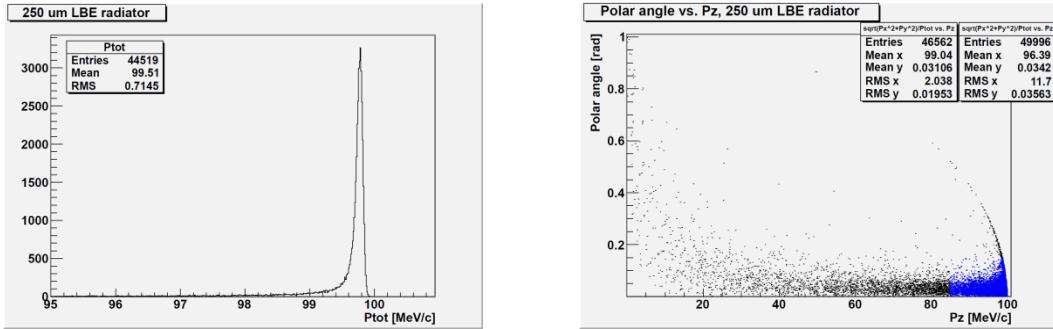


Figure 4: Beam momentum distribution after a 250 μm LBE radiator (left). Polar angle vs. longitudinal momentum (right) after the radiator (recoverable particles are indicated in blue).

Beam parameters at radiator

Our approach to handling the post-radiator electron beam is two-fold. We first optimize the beam parameters at the radiator. The angular beam distribution after passing through the radiator is determined primarily by multiple Coulomb scattering through small angles. As shown above, the angular spread after a 0.25 mm thick LBE radiator is about 20 mrad.

For any reasonable beam size at the radiator, the spread due to multiple scattering greatly dominates over the initial intrinsic beam angular spread. Therefore, the beam rms emittance $\varepsilon_{x,y}$ in each plane after the radiator can be written as

$$\varepsilon_{x,y} \approx \sigma_{x,y} \theta_{rms}, \quad (11)$$

where $\sigma_{x,y}$ is the horizontal/vertical beam size at the radiator. Obviously, minimizing the beam size at the radiator lowers the final emittance [3]. The downside of reducing the beam size is that it leads to a high instantaneous power density at the beam spot location on the radiator. We solve

this problem by considering liquid metal and spinning solid radiator designs as discussed in the Radiator design section below. Radiator design will be investigated in Phase II of the project.

The fundamental limit on the beam size at the radiator comes from the requirement that the transverse Twiss β functions at the radiator must be greater than or equal to the radiator thickness. This ensures that the beam size does not change significantly inside the radiator. This is an effect similar to the hour-glass effect in a collider, which results in increase of the effective beam size.

Table 3: Summary of the ERL beam parameters.

Injector beam energy	3 MeV	
Beam energy in the radiator section	100 MeV	
Beam energy at the dump	3 MeV	
Beam current	10 mA	
Bunch frequency	1497 MHz	
Bunch charge	6.7 pC	
Radiator material	LBE (or possibly W)	
Radiation length	7.1 mm	
Radiator thickness	0.25 mm (3.5% r.l.)	
Side of the radiator	Before	After
Twiss β function	4 mm	0.5 mm
Transverse geometric emittance (rms)	25 nm	\sim 200 nm
Longitudinal emittance (rms)	50 keV-ps	To be optimized
Bunch length (rms)	To be optimized	To be optimized
Momentum spread (rms)	To be optimized	\sim 0.1%
Transverse acceptance	Not an issue	\sim 5 μ m
Momentum acceptance	Not an issue	\sim 15%

Let us calculate Twiss β functions β_b and β_a before and after the radiator respectively. We assume a round beam, which is typical for electron linacs. In JLab's FEL, for example, a typical rms geometric emittance ε_b of a 100 MeV electron beam (without any interaction) is about 25 nm [4]. Let us also assume that the final rms geometric emittance ε_a after passing through the radiator at 100 MeV is 200 nm, which is consistent with acceptance of an ERL including adiabatic anti-damping during energy recovery. With 20 mrad rms angular spread θ_a just after the radiator, the rms beam size at the radiator is

$$\sigma \approx \varepsilon_a / \theta_{rms} = 10 \mu\text{m}. \quad (12)$$

10 μm beam size at the radiator corresponds to a Twiss β function before the radiator (Twiss $\alpha = 0$) of

$$\beta_b \approx \sigma^2 / \varepsilon_b = 4 \text{ mm} \quad (13)$$

and a Twiss β function after the radiator of

$$\beta_a \approx \sigma^2 / \varepsilon_a = \sigma / \theta_{rms} = 0.5 \text{ mm.} \quad (14)$$

There appears to be a discontinuity in the β function. This is because passing through the radiator is not a conservative process. The beam size, which is a real physical “observable”, of course, remains continuous. The obtained β functions are greater than the radiator thickness of 0.25 mm. Therefore, there is no significant change in the beam size inside the radiator. The resulting ERL beam parameters are summarized in Table 3.

The beam size and β function may seem small. However, they are within the state of the art of modern lepton colliders. In fact, below we apply a technique developed for colliders to manage the spent beam. This may be a nice application of what has traditionally been an “academic” technology, to a practical project. What also helps is that the initial beam emittance is smaller than in a typical collider.

Similar optimization is needed in the longitudinal dimension. It will be done in Phase II.

Non-linear compensation after radiator

The second component of our approach to beam handling is appropriate non-linear optics design. Despite nominally moderate emittance of 200 nm, spherical and chromatic aberrations in the downstream optics, especially in the nearest downstream quadrupoles, introduce significant beam smear and greatly degrade the beam quality. This problem is, in fact, very similar to compensation of non-linear impact of a final focusing triplet in a collider. Figure 5 (left) shows linear optics design of the post-radiator section. We compensate the chromatic kick of the large- β quadrupoles using a local Chromatic Compensation Block (CCB) developed earlier by a collaboration of Muons, Inc. and JLab [5]. Sextupoles in the CCB suppress chromatic β beat, as illustrated in Fig. 5 (right), without introducing significant geometric effects. For simplicity, at this stage, matching is done using linear matrices. Experience shows that non-linear impact of the matching section can initially be ignored with a good accuracy. A complete design will be developed in Phase II.

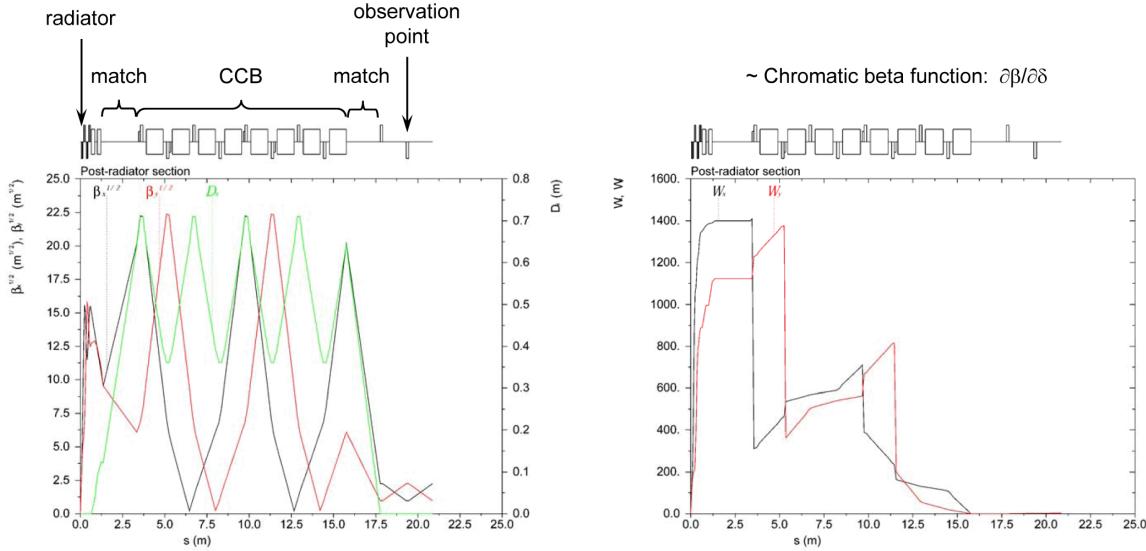


Figure 5: Linear optics design of the post-radiator section (left). Montague chromatic amplitude functions in the post-radiator section (right).

Three octupoles in the large- β region compensate spherical aberrations. The efficiency of the

octupole compensation is illustrated in Fig. 6 by tracking a beam with zero momentum spread using *Elegant* [6]. Setting the momentum spread to zero lets us distinguish spherical aberrations from chromatic ones. The spherical aberration effect is even more dramatic for more aggressive beam focusing designs. In addition, even though the tails of the final distributions may not look significant, filamentation would smear them all over the phase space ellipses (shown by the dashed lines in Fig. 6) that the final distributions are inscribed into. This would lead to beam loss and difficulties with collimation.

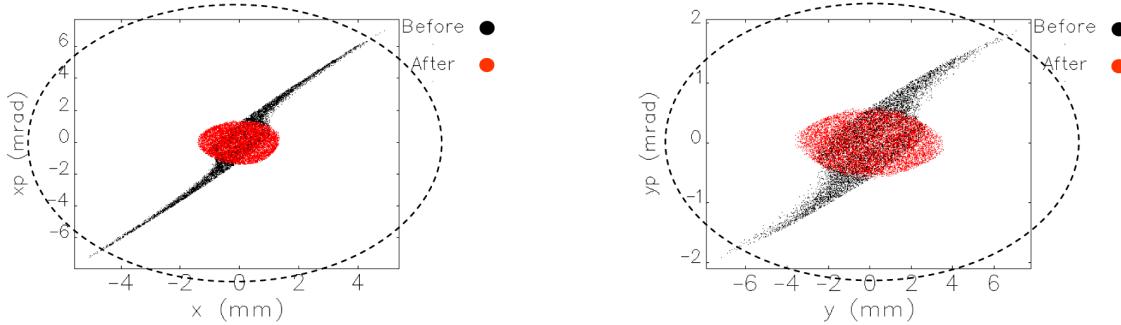


Figure 6: Horizontal (left) and vertical (right) beam phase space distributions at the observation point shown in Fig. 5 before (black) and after (red) octupole compensation. The initial beam size is $\pm 30 \mu\text{m}$, the initial angular spread is $\pm 60 \text{ mrad}$ and the momentum spread is 0. The dashed lines show matched phase-space ellipses that the uncompensated distributions are inscribed into.

Horizontal and vertical beam phase space distributions before and after compensation are shown in Fig. 7. They are obtained by tracking the beam from the radiator to the observation point using *Elegant*. Initial beam size is $\pm 30 \mu\text{m}$, initial angular spread is $\pm 60 \text{ mrad}$ and momentum spread is $\pm 3\%$. These quantities are not at their goal values yet. However, the CCB design has not been optimized in any way. We believe that, with a proper choice of a CCB scheme and appropriate optimization, we will significantly improve the non-linear performance. This will be done in Phase II.

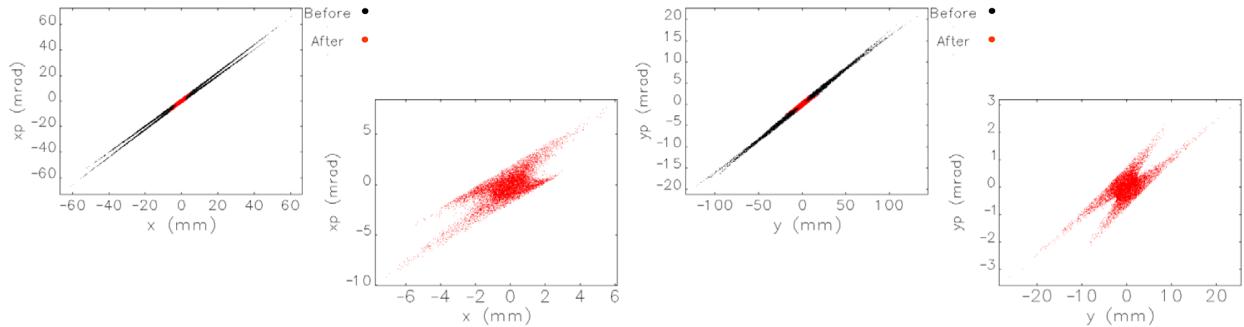


Figure 7: Horizontal (left) and vertical (right) beam phase space distributions in the post-radiator section before (black) and after (red) non-linear compensation. The initial beam size is $\pm 30 \mu\text{m}$, the initial angular spread is $\pm 60 \text{ mrad}$ and the momentum spread is $\pm 3\%$.

Beam collimation

Multiple scattering of electrons within the photon radiator results in a fraction of the spent beam that is not energy recoverable. This fraction requires both transverse and momentum collimation to provide controlled beam loss in the machine. Figure 8 shows the linear optics design for the post-radiator section. The beta functions shown in Fig. 8 indicate that the positions of largest transverse amplitude occur roughly at the centers of the second and third downstream quadrupoles. A first attempt at collimation utilizes the quadrupoles themselves as collimators by restricting the apertures of each of the downstream quadrupoles to 60 mm in diameter. In this case, the quadrupoles effectively act as 10 cm long beryllium collimators. The resultant phase space plots from this first collimation scheme are shown in Fig. 9.

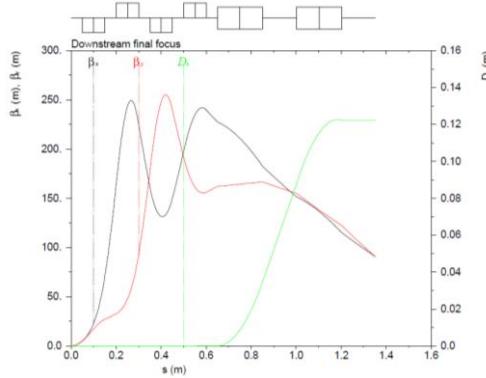


Figure 8: Linear optics design for the post-radiator region.

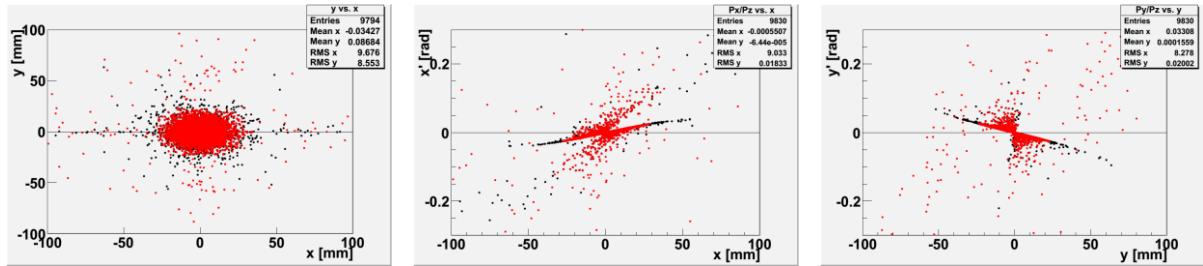


Figure 9: phase space plots for initial (black) and collimated (red) distributions at a plane near the midpoint of the final downstream quadrupole. The first collimation attempt aggravates the beam halo.

The phase space plots show that the large amplitude beam halo is not completely controlled with this collimation scheme, and a more sophisticated approach will be necessary. Here, limiting the quadrupole aperture scrapes off approximately 1% of the spent electron beam. Recent collimation work done at the JLab FEL [7] suggests that this fraction may be too high for effective collimation without further inducing beam halo; a fraction of a percent is most effectively scraped. This recent work also suggests using multiple collimation stages, and a strategy for the optics design to best accommodate this staged collimation. This strategy includes dual collimation stages for each transverse plane, with phase advance of 90 degrees between stages, and optimal collimator positions at positions of Twiss $\alpha=0$ [7]. In Phase II, we will utilize

this strategy to design appropriate optics for effective amplitude and momentum collimation of the spent electron beam.

Radiator design

Solid radiator

Because use of an ERL allows for higher beam currents due to the reduced beam power that must be dumped, care must be taken in the design of the photon radiator to handle the high power deposition from the beam-radiator interaction. In the case of a solid photon radiator, beam parameters must be chosen to maximize the photon production, minimize the beam size incident on the radiator, and avoid melting the radiator material. A high-Z material with a high melting point and good thermal conductivity is ideal for such an application. Assuming only radiative cooling of the solid radiator and a Gaussian electron beam, the relation between the radiator thickness, temperature, and beam parameters is given by:

$$\frac{w}{X_0} = 4\pi\sigma_{SB}T^4 \frac{\sigma_{beam}^2}{E \cdot I} \varepsilon, \quad (15)$$

where σ_{SB} is the Stefan-Boltzmann constant, T is the temperature, σ_{beam} is the rms beam size, E and I are the beam energy and current, respectively, and ε is the emissivity of the radiator material. For a 100 MeV, 10 mA electron beam incident on a 0.1 mm thick tungsten radiator, assuming a maximum temperature of 3500 K gives a minimum incident rms beam size of 28 mm. A large beam size and large angular spread induced by the radiator interaction results in a large transverse emittance after the radiator that will be difficult to transport and control. Larger incident beam sizes also reduce the maximum photon flux obtainable. Thus it is desirable to make smaller incident beam sizes feasible while maintaining integrity of the radiator. A possible solution is to employ active cooling with a rotating radiator. The size of this rotating radiator can be estimated by equating the surface area of the larger beam incident on the stationary radiator with that of the smaller beam incident on a rotating radiator disk. The beam extent on the rotating radiator will trace a ring on the rotating disk. For an initial beam size of 28 mm on a stationary 3% tungsten radiator, reduction of the rms beam size to 20 μm for manageable emittance gives a rotating tungsten radiator size:

$$\begin{aligned} \pi(28 \text{ mm})^2 &= 2400 \text{ mm}^2 = 2\pi R_{rotate} \cdot 2(0.02 \text{ mm}) \\ R_{rotate} &= 9500 \text{ mm} \end{aligned} \quad (16)$$

This estimate gives an impractically large rotating radiator. If the rotating radiator can shift in the transverse direction such that the beam traces a spiral covering the entire radiator surface, the radius of the rotating radiator can be reduced to $R_{rotate} = 5.5$ cm. Since active cooling of the radiator is necessary for obtaining small incident beam sizes, use of a flowing liquid metal may be another practical radiator solution.

Liquid radiator

High-power commercial superconducting accelerators for high-power x-ray production require converter targets capable of handling the power density of the full electron beam. Niowave is operating several types of high-power-density liquid-metal x-ray converter targets. The liquid metal loop which feeds high-Z liquid metal to the target region is shown as a CAD model accompanied by a photo of a system being prepared for test with heating tapes in Fig. 10. The target itself is a ribbon of lead-bismuth eutectic flowing between two thin windows, where the

thickness of the two thin windows and the liquid metal itself are determined for the particular operating parameters for the x-ray source. The beam window protects the vacuum and accelerator from the liquid metal and the output window confines the liquid and separates it from the atmospheric moisture content and other potential contaminants.

For production of radioisotopes with an ERL, a thinner target is required. Based on experience, the current designs can be modified to sub-mm thickness as required for this SBIR project. The issues that will determine the relevant thickness of windows and liquid metal, and therefore will comprise the physics design of the liquid metal loop x-ray radiator that can handle the full power of the electron beam, are:

- calculation of the electron beam penetration and scattering through the beam window to the liquid metal
- power deposition calculations considering heat from electrons and x-rays
- calculations of the temperature gradient across the liquid metal and across the beam window
- stress analysis on the beam window due to heating

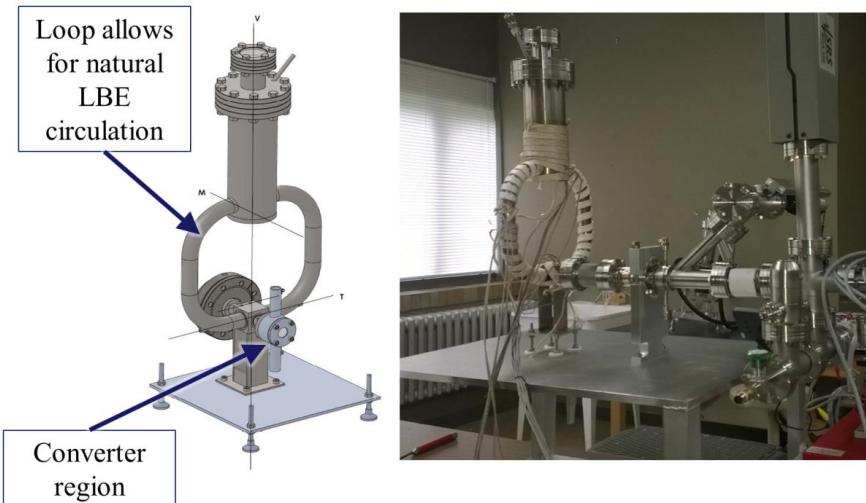


Figure 10: CAD model of a Niowave liquid-metal x-ray converter (using natural circulation to provide flow) and photograph of the system set up for testing using heating tapes.

Electron beam penetration and power deposition calculations will be performed using the Monte Carlo N-Particle eXtended (MCNPX) code, which is a general Monte Carlo code that handles particle and radiation interactions with matter. Temperature gradients, pressure gradients, and flow will be estimated using simple numerical models based on the coupled equations of mass and heat flow (radiative, conductive, and convective) in the system. Input into these models includes the density, specific heat, and dynamic viscosity of the lead-bismuth eutectic (all of which vary with temperature and for which data are available in the relevant temperature range of 400-700 K), the electron beam spot size, energy and current, and the cross sectional area of the liquid metal at the x-ray converter. For long term development and improved performance, very thin targets can be made by going to a window-less design, in which differential pumping protects the accelerator from the liquid metal, instead of a window.

Niowave is currently developing a positron source based on a liquid-metal target. This project requires handling power from a high-power electron beam after passing through the liquid metal, providing experimental benchmarks on performance of the target at 10 kW and more, and data on

the high-energy electron beam exiting the target. The experimental setup includes an achromatic dipole section to separate the electron and positron beams, which can also be operated to capture and perform diagnostics on the electrons. Experiments with thinner targets in this system will also progress toward an important commercial development of the ideas in this STTR project, which is the operation of multiple targets where the first target is thin and optimized for the use of high-energy x-rays, and the second target intercepts the rest of the beam and makes x-rays for low-energy-threshold isotope production or other x-ray applications.

Jefferson Lab SRF Energy Recovery Linac Expertise

JLab leads the world in the design, fabrication and operation of superconducting RF (SRF) linacs and SRF Energy Recovery Linacs (ERLs). The first practical demonstration of high-power energy recovery in an SRF linac was in JLab's IR Demo Free-Electron Laser (FEL) facility in 1999 (Fig. 2 [8]). Since then, that FEL has undergone upgrades and expansions and is regularly run with energy recovery at 135 MeV and 10 mA.

Niowave, Inc. Commercialization Expertise

Niowave, Inc. is developing a radioisotope production facility based on a superconducting electron linac with multiple liquid metal targets. The initial superconducting linac will be a 20 MeV, 50 kW machine, with a straightforward upgrade to a 40 MeV, 100 kW machine. These electron linac based production facilities are expected to cost between \$10-15 million with minimal licensing hurdles. This would allow the siting of many units around the country that are close to large metropolitan areas to supply a broad range of isotopes reliably and economically.

Niowave is currently licensed to operate accelerators up to 40 MeV and 100 kW through the state of Michigan. Niowave is also licensed through the Nuclear Regulatory Commission (NRC) to possess and machine source materials. The NRC is currently reviewing Niowave's license for isotope production from stable targets. This SBIR/STTR project offers a path to an even more efficient accelerator for making isotopes using an ERL, and a different operation regime in terms of the thermal effects and activation in the thin x-ray production target. Niowave brings expertise in commercial, robust systems for superconducting linacs – ensuring that the recirculation technology pioneered at the national laboratories can be made economically viable.

Named the 2010 SBIR/STTR Small Business of the Year for the Department of Energy, Niowave is the only company worldwide capable of building and testing a superconducting linear accelerator in its own facility, as well as delivering and commissioning complete accelerator systems for its customers. Its state of the art facilities produce:

Complete accelerator systems & cryomodules
Electron guns / injectors
Niobium and niobium alloys.

Helium refrigerator systems
Accelerating cavities & components

Industrial Applications of radioisotopes are numerous and may offer opportunities that we will explore. Many types of thickness gauges exploit the fact that gamma rays are attenuated when they pass through material. By measuring the number of gamma rays, the thickness can be determined. The isotope ^{241}Am is used in many smoke detectors for homes and businesses (as mentioned previously), in thickness gauges designed to measure and control metal foil thickness

during manufacturing processes, to measure levels of toxic lead in dried paint samples, and to help determine where oil wells should be drilled. The isotope ^{252}Cf (a neutron emitter) is used for neutron activation analysis, to inspect airline luggage for hidden explosives, to gauge the moisture content of soil and other materials, in bore hole logging in geology, and in human cervix-cancer therapy.

MuPlus/Muons, Inc. Simulation Expertise and MuSim Commercialization

MuPlus is a wholly-owned subsidiary of Muons, Inc., which is responsible for the development of G4beamline and MuSim programs, time-saving human interfaces to GEANT4 and MCNP6, respectively. Dr. Thomas Roberts, the creator and developer of these interfaces will be working on this project and is named as a key individual. G4beamline has become a workhorse in the global accelerator community, with over 200 projects using it at a level of \$17 M of manpower devoted to its exploitation outside of SBIR-STTR grants. In this project, MuSim, our newest simulation software interface to MCNP6 will be exercised by students, postdocs, and workers at Jefferson Lab, Niowave, and Muons/MuPlus and thereby strengthened for industrial use. We believe that there is a need and will be a demand for this software by many workers at Nuclear Physics, NNSA, DHS, and other facilities that would benefit from the ease and efficiency of MuSim and would pay license fees for maintenance and development. MuSim has an even better human interface and we believe a much larger potential user community. It will greatly accelerate studies in this project of radioisotope production as a function of beam and target parameters.

Anticipated Public Benefits

ERLs are increasingly the technology of choice for highly demanding applications. In energy recovery, more than 90% of the beam power is recycled and not deposited in a beam dump. Therefore, the energy of the waste beam is lower than the threshold for neutron production and the activation of shield components, thereby reducing both complexity and cost. Our first application will be for nuclear medicine, which has humanitarian and commercial benefits. According to LBNL studies [9], of the 30 million people who are hospitalized each year in the United States, 1/3 are treated with nuclear medicine. More than 10 million nuclear-medicine procedures are performed on patients and more than 100 million nuclear-medicine tests are performed each year in the United States alone. A comparable number of such procedures are performed in the rest of the world. There are nearly one hundred radioisotopes whose beta and/or gamma radiation is used in diagnosis, therapy, or investigations in nuclear medicine. Our aspirations include garnering some of the market share for the commonly used isotopes as well as developing techniques for isotopes for new medical, scientific, and industrial applications.

The LBNL study continues: By 1970, 90 percent of the 8 million administrations per year of radioisotopes in the United States utilized either ^{131}I , ^{60}Co , or $^{99\text{m}}\text{Tc}$. Today, $^{99\text{m}}\text{Tc}$, with a half-life of 6 hours, is the workhorse of nuclear medicine. It accounts for more than 10 million diagnostic procedures a year in the United States. It is used for brain, bone, liver, spleen, kidney, lung and thyroid imaging as well as for blood-flow studies. ^{131}I , with a half-life of 8 days, is used to diagnose and treat thyroid disorders. A very effective role for radioisotopes in nuclear medicine

is the use of short-lived positron emitters such as ^{11}C , ^{13}N , ^{15}O , or ^{18}F in Positron Emission Tomography (PET). For cancer therapy, the radioisotope ^{60}Co emits gamma rays that are used to destroy cancer cells.

The global nuclear medicine market is fragmented, with more than 60 companies selling radio-pharmaceuticals on a regular basis; however, three companies take more than half of the world market share whereas more than 50 companies share 14% of this same world market. While nuclear medicine is not a recent technology, the emergence of new radionuclides associated with the appearance of almost 30 new dedicated R&D companies opens a new era for this industry, which will consolidate as soon as it matures. The global nuclear medicine market is now in its third S-Curve, with a number of products that are still under development. But at the same time some major companies are already starting to acquire smaller players with novel and potentially blockbuster products.

This era opens new opportunities in the nuclear medicine landscape. These opportunities have been identified not only in the radiodiagnostic area but notably in therapeutic radiopharmaceuticals, with the first products scheduled to reach the market before the end of 2020. Opportunities exist for larger groups or investors to finance such development, to merge with some partners and/or to acquire companies.

The global market is expected to reach US\$ 24 billion (EUR 18 billion) in 2030, showing an annual average growth of 11%, mostly driven by the therapeutic radiopharmaceutical market which is expected to increase annually by 30% between 2013 and 2030. The diagnostic radiopharmaceutical market is expected to grow by 5% a year, mainly driven by volume increases.

In 2013 about 60% of the world market (in value) was based on Technetium-99m; ^{18}F -FDG accounts for about 20% of the market, while therapeutic radiopharmaceuticals correspond presently only to a small 4%. This latter is expected to grow strongly in the coming years.

Presentation

A.V. Sy, G.A. Krafft, C.H. Boulware, R.P. Johnson. "Energy Recovery Linacs for Commercial Radioisotope Production". Proceedings of 2015 International Particle Accelerator Conference, Richmond, VA, USA. http://accelconf.web.cern.ch/AccelConf/IPAC2015/talks/mobb3_talk.pdf

Participants

MuPlus, Inc.; R.P. Johnson, T.J. Roberts

Thomas Jefferson National Accelerator Facility; D. R. Douglas, G. Krafft, V. S. Morozov, A. Sy Niowave, Inc.: C.H. Boulware, Terry Grimm, V. Stoyanova

Patent

Non-provisional patent application 15/143,383 filed 04/29/2016 with priority date 05/02/2015, ENERGY RECOVERY LINAC FOR RADIOISOTOPE PRODUCTION WITH SPATIALLY-SEPARATED BREMSSTRAHLUNG RADIATOR AND ISOTOPE PRODUCTION TARGET, inventors: David R. Douglas, Rolland P. Johnson, Andrew Kimber, Geoffrey Krafft, Vasiliy S. Morozov, Amy Sy.

Conclusions

Using Muplus/Muons programs MuSim for MCNP6 and G4beamline for GEANT4 and other codes as described in the technical approach sections above, we demonstrated:

- 1) The ERL advantage for producing radioisotopes is at high electron beam energies (~100 MeV) where the fraction of recovered beam energy is highest and the ERL beam power is over 5 times more effective at making radioisotopes than a single-pass linac. High beam energy matches well with the desire to make radioisotopes with higher photoproduction threshold energies;
- 2) The range of acceptable radiator thickness is narrow (too thin and there is no advantage relative to other methods and too thick means energy recovery is too difficult). This result confirms that it is unlikely that an ERL with a combined radiator and radioisotope production target in the electron beam is practical. Realizing this, we submitted a non-provisional patent application based on figure 2 above, where only the thin radiator is in the electron beam;
- 3) Using optics techniques developed under an earlier STTR for collider low beta designs greatly improves the fraction of beam energy that can be recovered.
- 4) Unwanted residual radioactivity in the target area is reduced by using an ERL, instead of a single pass bremsstrahlung beam, to decelerate the beam below the neutron activation threshold.
- 5) Based on these encouraging results, we believe that many radioisotopes can be made that have never before been available in significant enough quantities to justify their study.

A Phase II project was proposed to start with a Conceptual Design Report (CDR) based on the results of Phase I and conclude with a Technical Design Report (TDR) that is a complete design of an ERL-based isotope production facility, with optimized performance based on end-to-end simulations.

References

- [1] L. Merminga, D.R. Douglas, and G.A. Krafft, *High-Current Energy-Recovering Electron Linacs*, Annu. Rev. Nucl. Part. Sci. 53, 387-429, (2003).
- [2] *Handbook of accelerator physics and engineering*, 3rd edition, edited by A.W. Chao and M. Tigner (World Scientific, 2006), p. 243.
- [3] D. Douglas, JLAB-TN-12-046 (2012).
- [4] D. Douglas, JLAB-TN-12-017-REDACTED (2011).
- [5] V.S. Morozov et al., Phys. Rev. ST-Accel. Beams 16, 011004 (2013).
- [6] M. Borland, Advanced Photon Source LS-287 (2000).
- [7] D. Douglas and S. Benson, private communication.
- [8] G. Neil *et al.*, *Sustained Kilowatt Lasing in a Free-Electron Laser with Same-Cell Energy Recovery*, Phys. Rev. Lett. 84, 662 (2000).
- [9] <http://www2.lbl.gov/abc/wallchart/chapters/13/2.html>