Accelerator-Driven Transmutation Technology for Energy Production and Nuclear Waste Treatment*

R. A. Jameson, G.P. Lawrence, S.O. Schriber
Los Alamos National Laboratory
Los Alamos, NM 87545, USA

Abstract

New concepts recently developed at Los Alamos show that the use of intense particle accelerators affords unique opportunities for electrical power generation, from plentiful fuel such as thorium, with little long term waste legacy. The concept can also effectively transmute existing actinide and fission product wastes. The physical processes to be used are different and more advanced than earlier ideas; the new concept uses the accelerator beam to generate intense flux levels (10^{15} - 10^{16} n/cm^2·sec) of thermal energy neutrons that efficiently transmute fuels or actinide wastes via a two-step capture/fission process, and also efficiently transmute fission products to stable or short-lived end-products. Effective cross sections for the actinide transmutation are enhanced at higher flux levels, and thermal-energy fission-product cross-sections are also higher than at fast neutron energies. The high neutron flux values and large cross-sections allow large transmutation rates with very small resident material inventories, a factor of 100 or more smaller than earlier methods. This feature, realized in a dilute, continuously flowing system, results in significant safety and engineering advantages.

Proton cw accelerators in the 800-1600 MeV, 50-250 mA class are required, depending on the desired plant configuration. The technology base for such accelerators has been thoroughly reviewed and is feasible. Beam dynamics and optimization issues related to insuring low beam loss along the linac are outlined.

1 INTRODUCTION TO THE ATW CONCEPTS

The need for clean energy supplies is a world problem. Nuclear power is presently under the onus of a legacy of existing long-lived radioactive waste that must be disposed of, and more waste must not be allowed to result from future systems. Indeed, until the waste problem is solved, public opposition to nuclear power will undoubtedly continue. The required solution must reduce both the volume of existing waste and its radioactivity, so the waste becomes benign by the end of a period over which a society might be able to retain control, say a few hundred years.

Studies[1-3] at Los Alamos indicate that an accelerator-driven thermal neutron source could address both the transmutation of wastes and clean generation of electrical power in a more complete, safe, and environmentally responsive way than earlier approaches. The thermal neutron source approach has the following advantages:

- Completeness — The system is unique in that it can process both long-lived actinides and fission product wastes, while producing net electrical power. In an energy production configuration, fertile material (^{232}Th or ^{238}U) is converted to fissile fuel (^{233}U or ^{239}Pu) and burned to produce power, including enough excess neutrons to convert its own high-level wastes to stable or short-lived products.
- Speed of Processing — Use of a sufficiently high flux of thermal neutrons, such as can be provided only by an accelerator-driven system, allows advantage to be taken of a high cross-section, two-step burning process for fissioning long-lived actinide wastes to stable or short-lived fission products, with excess neutrons left over. The high neutron cross section and high flux allows the amount of material required to achieve a given burnup rate to be two orders of magnitude or more smaller than in a reactor system with the same burnup rate. Further, fission product capture cross sections are higher for thermal neutrons, and there are excess neutrons available for burning them.
- Safety — The neutron producing target/blanket assembly is thus a small radionuclide source term and is strongly subcritical, removing any possibility of a criticality accident. The accelerator driver can be instantly shut off, by a variety of means.
- Environmental — The low material inventory, coupled with optimized chemical processing, results in minimization of waste streams and small end-of-life residues, allowing on-site management of remaining low-level waste.

The general features of the Los Alamos Accelerator Transmutation of Waste (ATW) concept are shown in Figure 1. The plant configuration has a wide range of options depending

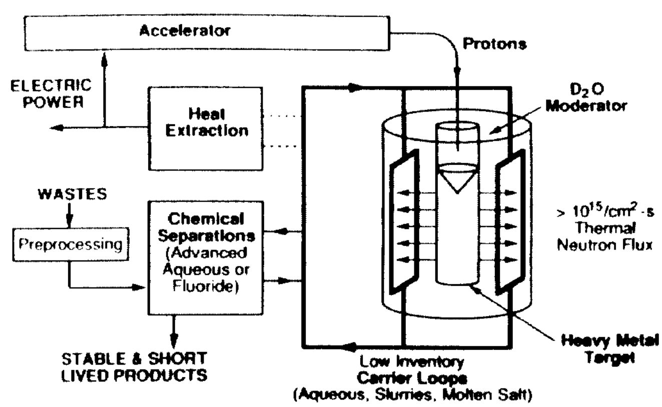

Figure 1. General features of the ATW concept.

on the emphasis among a number of applications, including transmutation of existing waste, generation of electric power, or production of special nuclear materials. Typical plants of 2000-6000 MWt capacity would require a linear accelerator with beam power of 200-400 MW, translating to proton accelerator energies in the range 800-1600 MeV at continuous currents of 150-250 mA. The proton beam is delivered to a heavy metal (Pb, W or U) target, producing neutrons. A heavy water moderator (several meters in diameter) surrounds the spallation target to slow the neutrons down to thermal energies. Fluxes greater than 2-3x10^{15} n/cm^2·sec are

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produced over a large active volume. The materials to be transmuted are carried in a dilute (by volume), low inventory system using continuous material feed in a carrier material such as aqueous media, oxide slurries, or molten salt. Because the inventory is small, chemical partitioning processes leading to the highest decontamination factors can be used. If a molten-salt carrier system can be successfully developed, the salt's high thermal-to-electric conversion compatibility can be used to substantially increase the efficiency of electricity production, and also would enable advanced fluoride chemistry or physical methods to be used for removal of fission products from the actinide fuel.

Base-case ATW systems have been laid out using current, credible technology[3], and options for substantial improvement through development have been identified. The base case is driven by a 250 mA, 1600 MeV accelerator requiring 900 MWe. The proton beam is split to four target blanket modules, each producing ~1.5 GWt from actinide fission for a total system power of 6 GWt. The system will burn the actinide waste from ~7.5 LWR's, i.e. about 2450 kg/yr. Each blanket also provides excess neutrons to transmute the ^{99}Tc and ^{129}I from 7.5 LWRs (~250 kg/yr) to stable products. The thermal-to-electric conversion efficiency is 30%, yielding ~1000 MW of net (after driving the accelerator) electrical power to the grid.

A crucial aspect of these systems is called the "mass balance"; an exhaustive analysis of all material entering and leaving the system. This is a revealing method for comparison among transmutation approaches, and the recently convened US National Academy of Science review on radwaste has requested that all approaches furnish a comprehensive mass balance. The preliminary results for the baseline LWR waste burner outlined above are, for the yearly transmutation rates cited: per kilogram of waste burned per year, 30 g of radionuclides with halflives ≥ 30 years are produced in the spallation target; per kilogram of fission products burned per year, one kilogram of Class C or better transmuted by-product is discharged; per kilogram of actinide burned per year, < 250 liters of non-TRU liquid discharge (mainly water); N_2 and CO_2 are produced; 156 g of inerts (phosphates, sodium, ash) are produced; and 65 g of radionuclides with halflife > 30 years (^{135}Cs, ^{107}Pd, ^{93}Zr) are produced.

2 ACCELERATOR REQUIREMENTS

In recent papers[4-6], we have outlined the basic accelerator requirements, presented point designs, discussed the design approach for near-term and longer-range improved options, and the technology issues and technology base. Very briefly summarized, the required room-temperature technology is in hand. An integrated "front-end" funneled system up to at least 40 MeV should be constructed as a testbed for final engineering development and reliability/availability development. Two major reviews, by ERAB[7] and JASON[8], have emphasized these points. We believe that superconducting rf technology may be appropriate in the longer range for energy-production systems where efficiency is paramount, and we are initiating studies in this area[5].

A key design factor for these long, high-intensity linacs is to insure that beam losses along the linac are kept low enough to insure "hands-on" maintenance, without remote manipulators, over the lifetime of the facility. The reference design concepts used to date[9] address low beam loss by:

- Use of the radiofrequency quadrupole (RFQ) to bunch and preaccelerate the beam with very good emittance preservation, with operation well below the current limits of the device. Current limitations on a single injection channel are one reason for the funneled approach; a second is to fill every bucket of the doubled-frequency downstream linac for minimum emittance at a given current.
- Attention to keeping the beam matched well across machine structure transitions (e.g. from RFQ to DTL to CCL, or where magnet or tank groups change), minimizing the number of such changes, and trying to make such changes at the lowest energy, where beam loss is less important.
- Attention to achieving high "rms aperture ratios", the ratio of transverse aperture to rms beam size (at flutter factor maximum), and longitudinal bucket width to rms beam length. The strong economic tradeoff between linac length and gradient argues for constant accelerating gradient along most of the linac (~1 MV/m at present rf costs); thus the longitudinal focusing weakens rapidly at higher energy, and some longitudinal emittance growth is typically allowed. In the transverse direction, strong focusing is provided, the aperture is kept large (at some penalty in rf losses), and rms aperture ratios of at least 10 are sought.
- The design is simulated in a fully nonlinear, $2^1/_2$ - 3-D particle-in-cell simulation code, using a large number of particles, and introducing various errors within engineering tolerances. If these runs show a well-matched beam, adequate aperture factors, and no particles lost (beyond those not accepted by the RFQ), the design is judged satisfactory.

The criteria are thus seen to be a combination of beam dynamics, numerical consideration, and cost and engineering factors.

The beam dynamics in the codes has been thoroughly validated in terms of rms behavior in different experiments at different laboratories. The behavior of the total beam is another matter - few accelerator laboratories, beyond LAMPF, have had to worry about very small beam losses in linacs (typically, fractional losses must be kept below 10^{-5} to 10^{-8}/m, more stringent at higher energy). The beam dynamics codes contain approximations that basically limit their validity. At LAMPF, detailed beam halo measurements were made to the 10^{-4} - 10^{-5} level that qualitatively agreed with simulations, but it was observed that the halo patterns were different from day-to-day. The best validation of loss prediction is a comparison[10] between the residual activity pattern along the LAMPF linac, accumulated over its 20-year operating history, compared to the beam spill prediction using an as-built datafile in the simulation code with a large number of particles. Losses patterns at spill areas where machine transitions occur (more abrupt than we would now allow) are accurately predicted. The simulations predicted all measured quantities to 10-15%, except for total beam loss, which is sensitive to the tail population. "Design" and "production" input distributions to the simulation overestimated the total loss by x3 and x10 respectively. With careful use, it is believed (RAJ) that the codes allow quali-tative assessment, and rough quantitative assessment, to fractional losses of ~10^{-3} - 10^{-4}.

The numerical aspects have also received detailed attention at Los Alamos. With supercomputers, brute force simulation of very large numbers of particles could be done (the required number of particles per bunch in the ATW machines is

~2x10^9). Careful attention[11,12] has been given to appropriate fitting of measured particle distributions; modified Weibull distributions give good fits, whereas combinations of Gaussians are very poor. The statistics of the outliers can be independently assessed, with predictive power that is undoubtedly better than the code physics deserves.

An engineering safety factor based on the rms aperture factors is then used, a practice based on experience such as LAMPF's and many circular accelerators and storage rings around the world. The transverse rms aperture factor near the end of the LAMPF CCL is about 6.3; our preliminary ATW designs strive for at least 10.

These criteria are rigorous, defensible in detail, and adequate to proceed confidently with construction plans for linacs in this class. For all that, every accelerator builder knows that there is always room for a better job. For these high-intensity, low-beam-loss linacs where very high efficiency is also crucial, it is necessary to know that the designs are optimum with respect to a complex combination of beam-loss, efficiency, cost and other criteria and constraints. Optimization, for example, to the best rf frequency, aperture size, focusing strategy, injection conditions, and so on. And it is the case, presently, that while we can demonstrate adequacy, we cannot clearly describe the optimum for the high-intensity linac where low-beam-loss and high-efficiency are the primary objectives. The following paragraphs outline initial attempts by one of us (RAJ) to define and address aperture factor optimization, within the constraints of other objectives.

3 LOW BEAM LOSS DESIGN OPTIMIZATION

The (transverse or longitudinal) (rms or total) aperture factors are the ratio of the accelerator bore or longitudinal acceptance to the corresponding (rms or total) beam size. Unfortunately, we do not have analytic relationships in general; there are (at least) two difficulties:

- Even the simplest rms formulas we have (see below) are nonlinear and coupled.
- Total beam size is of course the important quantity. "Halos" or "tails" can form around the beam from a variety of effects, but their extent cannot be predicted theoretically, nor even be related to the rms beam properties. Therefore optimization efforts must always keep the total beam size under surveillance using the full simulation codes.

3.1 Design Philosophy -> Matched System

A beam focusing system has beam kinetic and internal field energy and potential energy from the external fields. The definition of a matched system is that the beam and its surroundings are in perfect equilibrium, including any nonlinear or time-varying effects. Our design philosophy is to try to achieve matching; our optimization philosophy should start with the matched condition, but is subject to practical constraints.

An imbalance (mismatch) anywhere constitutes a source of free energy. There are many possible sources – independent or dependent on beam intensity, linear or nonlinear, static or time-varying. Via interaction with a nonlinearity, a mechanism, or dynamical path, exists for the free energy to convert to (coarse-grained) emittance growth, leading toward a new equilibrium (in the absence of further driving terms). The evolution can occur through single-particle or collective motion, and be a stable or unstable process., with a rate that varies accordingly. For example, if there is a strong enough anisotropy of divergence between the degrees of freedom, an equipartitioning energy transfer between planes will occur through incoherent instabilities[13-15]. Intra-plane effects will result from mismatch even without instability, through the couplings between planes.

Matching of the beam's rms envelope equations of motion forms a robust basis for the design philosophy, describing an equilibrium condition for a uniform beam density distribution in a periodic focusing system:

$$\varepsilon_{tn} = \frac{a^2\sigma^t\gamma}{n\lambda} \quad ; \quad \varepsilon_{ln} = \frac{b^2\sigma^l\gamma^3}{n\lambda} \qquad (1)$$

where ε denotes emittance (here total emittance), sub-or-super-t-or-l the transverse or longitudinal plane, sub-n the normalization to canonically preserved emittance during transport or adiabatic acceleration, a and b the transverse and longitudinal radii, of an ellipsoidal beam bunch with uniform particle distribution, n is an integer describing the number of $\beta\lambda$'s in the transverse focusing period, and σ is the phase advance over the distance $n\beta\lambda$.

The phase advances σ^t and σ^l are each comprised of a beam-independent (zero-current) part, $\sigma_0{}^t$ and $\sigma_0{}^l$, representing the external focusing fields of the linac, and a beam-dependent part that is the counteracting space-charge effect:

$$\sigma^{t2} = \sigma_o^{t2} - \frac{I\lambda^3kn^2(1-ff)}{a^2b\gamma^3}$$
$$\sigma^{l2} = \sigma_o^{l2} - \frac{2I\lambda^3kn^2ff}{a^2b\gamma^3} \qquad (2)$$

where we are working with a smooth, small-angle, approximation to the focusing forces, k = $(3Z_oq)/(8\pi\, m_oc^2)$, and ff = ellipsoid form factor ~ a/(3γb) for γb/a between approximately 0.8 and 5.

The relationships among the beam emittances, sizes and phase advances (or "tunes") are the most fundamental way to look at the beam physics situation along the machine. The balance between external focusing forces and the space-charge self-defocusing of the nonrelativistic beam is seen in σ^t and σ^l, equaling zero at exact cancellation, with laminar (parallel) particle trajectories and zero divergence. The ratios $\sigma^t/\sigma_0{}^t$ and $\sigma^l/\sigma_0{}^l$ are called the tune depressions. Beam current, energy, and the operating frequency, usually used as primary quantities, are seen to be subsumed into these more basic relationships. The equations locally agree very well with simulation results along a typical linac. They are relatively insensitive to injected non-matched density distributions; the mechanism and asymptotic behavior of the (rapid and usually small) emittance growth from this effect are known[14-15], and can usually be neglected for design and optimization. The consequences of ellipse and off-axis beam mismatch in these equations are also known[16]. Changes known apriori can be introduced into these equations, but they basically describe a linearized system in which there can be no emittance growth.

A third important rms equation is known[17], describing the condition known as "equipartitioned", when there is energy balance of the potential and kinetic energies between the transverse and longitudinal degrees of freedom:

$$\frac{\varepsilon_{ln}}{\varepsilon_{tn}} = \frac{\sigma^t}{\sigma^l} = \frac{\gamma b}{a} \qquad (3)$$

Rewriting this as $e_{ln}/\gamma b$ = b' = e_{tn}/a = a' shows that the divergences are equal when the beam is equipartitioned. The products $\varepsilon\sigma$, and a'^2, b'^2, are energies.

Under reasonable design conditions (non-linearities and rate-of-change of parameters not too large, certain resonances avoided), it is known that if Eqns (1) and (3) are locally satisfied, emittance growth is strongly avoided. These are the only general equations we have at present for the matched condition, so their simultaneous solution is now being explored as a design philosophy and basis for optimization.

There is a practical caveat, however. Matching using Eqn (1) alone is relatively easy and produces "smooth" beams. Achieving equipartitioning has hardly been explored yet, but in some cases, it appears costly. For example, it may be hard to achieve the proper ratio of emittances. Because coherent instability mode thresholds are involved in this mechanism, there is also leeway around balance (a rule of thumb for e_{ln}/e_{tn} » 1 is to keep $\sigma^l/\sigma^t \leq 1.5$). So the requirement for equipartitioning could be relaxed or abandoned, and some emittance growth might be allowed from this or other mechanisms. It is not at all clear whether a linac with no emittance growth will have a better aperture factor than a different linac, set up with a different prescription that allows some emittance growth. But it seems that a thorough understanding of a design approach that intrinsically avoids emittance growth is a very good starting point, from which the consequences of compromise might be assessed.

With three equations, three variables can be left free while the others are fixed. The coupled, nonlinear equations are generally impossible to solve for analytic scaling relationships (for example, solutions for a and b). Guided by numerical examples, some progress has been made that will be reported elsewhere. One of the most sought results is how to optimize the choice of frequency to maximize the aperture factors. It has become quite clear is that this is by no means obvious!

In the transverse plane, the linac aperture may remain constant with energy, or vary as some function, say of $\beta\lambda$, and this will strongly affect the optimization. The transverse and longitudinal aperture factors usually will not optimize at the same place, so a criterion to balance them must be chosen.

In some cases (choices of free vs. fixed variables), it has been found numerically that the aperture factor will maximize at the same tune depression in that plane, over a wide variation of any of the variables used to arrive at that tune depression. Such a result is surprising and important to understand, so a strong effort is being made to find an analytic solution that will elucidate the effect.

In the longitudinal plane, there is a further complication. Within the framework of the smooth approximation and no acceleration, it has been shown[18] that the longitudinal acceptance width would shrink as $(1 - (\sigma^l/\sigma_o{}^l)^2)$. With acceleration, numerical simulation shows that the bucket width does not shrink this fast. There is at present no theory that gives the effective acceptance under accelerating and space-charge conditions. It might be that a useful relationship could be derived using some of the recent understanding of how free energy affects the beam dynamics. For the ATW designs, we are exploring the acceptance shrinkage numerically, because it is crucial to the aperture factor design to know the acceptance behavior. A particle bunch is propagated through the linac, generating the appropriate space-charge forces, which are then applied to zero-current "test particles". The test particles are injected at some cell on a grid overlapping the acceptance; relating test particles that have survived downstream to their initial coordinates on the injected grid defines an acceptance boundary. In our initial studies, we assume that the downstream channel is defined by a linac with a constant characteristic - for example, constant $\sigma_o{}^t$ and constant accelerating gradient. Depending on whether there is emittance growth, this may mean that the tune characteristic of the downstream channel is not constant. Thus is is not yet clear how the effective acceptance should be defined.

We have been discussing matching using equations for rms conditions in periodic systems – a useful construct for rms injection matching or for local conditions along the linac. Introduction of acceleration and smoothly but fairly rapidly changing parameters is needed. In I. Hofmann's words[14], "An rms matched beam is (intrinsically) mismatched if the nonlinear field energy term [or externally supplied free energy term - my addition] changes rapidly within a coherent oscillation period...". Simulations indicate this is a factor in typical rf linacs. Acceleration has been incorporated in the envelope equations for 2-D beams[19]; a high priority is to extend this to 3-D bunched beams, as well as parameter variations. Again, from a practical viewpoint, non-adiabatic parameter changes (even though smooth) are unavoidable – it is of interest to determine if effective compensation (to prevent emittance growth) is possible.

3.2 Perspective on ATW CCL

What does an ATW CCL linac look like from the perspective of tunes, tune shifts, equipartitioning, and other matching aspects? Assume a coupled-cavity linac from 20-1600 MeV, with a constant $\sigma_o{}^t$ = 80°, constant real-estate accelerating gradient of 1 MV/m, frequency = 700 MHz, ϕ_s = -30°, aperture = 2.5 cm radius, current = 140 mA. An rms-matched beam is injected with transverse, normalized, rms emittance of 0.02 cm.mrad (about the smallest that might be achieved at this current from the ion source/RFQ/DTL), and longitudinal normalized rms emittance of 0.04 cm.mrad. Solution of Eqns (1) shows:

- $\sigma^t/\sigma_o{}^t$ rises as {0.4,0.62,0.91} at {20,200,1600} MeV.
- $\sigma^l/\sigma_o{}^l$ falls as {0.5,0.24,0.08 at {20 ,200,1600} MeV.
- the transverse rms aperture factor rises as {13,17,32}.
- the longitudinal rms aperture factor assuming that the bucket width remains $3\phi_s$ rises from 3.5 to 9; assuming the bucket shrinks as $(1 - (\sigma^l/\sigma_o{}^l)^2)$, falls from 1.2 to below 1.

Simulation shows the beam to be very smooth in the rms-matched sense of Eqn (1), without betatron or synchrotron oscillations. There is transverse rms emittance growth of x1.3, but emittance growth of the total beam peaks at x5 around 300 MeV, and levels off to ~x4. The transverse rms beam radius shrinks continually under this strong focusing and the transverse rms aperture factor looks good (~37); however, the aperture factor based on total beam radius is ~5.7.

The longitudinal rms emittance grows steadily to 1600 MeV, reaching x2.1, with total emittance growth of x7-8. Factoring these rms growths into local solutions of Eqn.(1) produces excellent agreement.

Inspection of the the coherent instability thresholds[13] shows that the operating tune depressions for the first few MeV are below the thresholds; emittance transfer from longitudinal to transverse is indeed seen in the simulation, stopping as soon as the tunes rise above threshold. Above 200 MeV, the ratio b'/a' is level at ~0.8, indicating no further transfer. σ^l/σ^t is <1 and decreasing (Hofmann indicates that modes in this regime are negligible) but $\sigma^l/\sigma_0{}^t$ is >0.8 anyway, well above the thresholds.

Additional runs were made with the transverse rms input emittance doubled to 0.04 cm.mrad, and doubled again. The tune progressions are similar. Transverse total emittance growth was ~x3 and ~x1.5 at 0.04 and 0.08 input, respectively. The total beam radius at 1600 MeV for the 0.02 input is only ~20% smaller that that for the 0.08 input. Longitudinal growths remained nearly the same. The initial instability is avoided at higher input emittance. Above 200 MeV, the b'/a' ratio is lower with higher input emittance, but no transfer occurs for the same reasons.

Thus there appears to be no emittance growth from rms mismatch or coherent instability effects; yet there is clearly a large and continuous source of free energy, converting primarily to longitudinal rms emittance growth and halo formation in both planes. It is believed this is due to rapidly changing parameters and acceleration, and so we plan to determine the amount of free energy available from these effects and how it gets converted into emittance growth. In this linac with constant real-estate accelerating gradient, $\sigma_0{}^l$ decreases as $\gamma^3(1 - (1/\gamma^2))^{1/2}$, the space charge terms decrease as γ^3, and the strong focusing, constant $\sigma_0{}^t$ recipe results in the rapidly changing transverse tune during the first few hundred MeV.

Summarizing, the intended goal of these studies is to describe an optimum design for ATW-class linear accelerators, and to understand the consequences of the inevitable compromises entailed.

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