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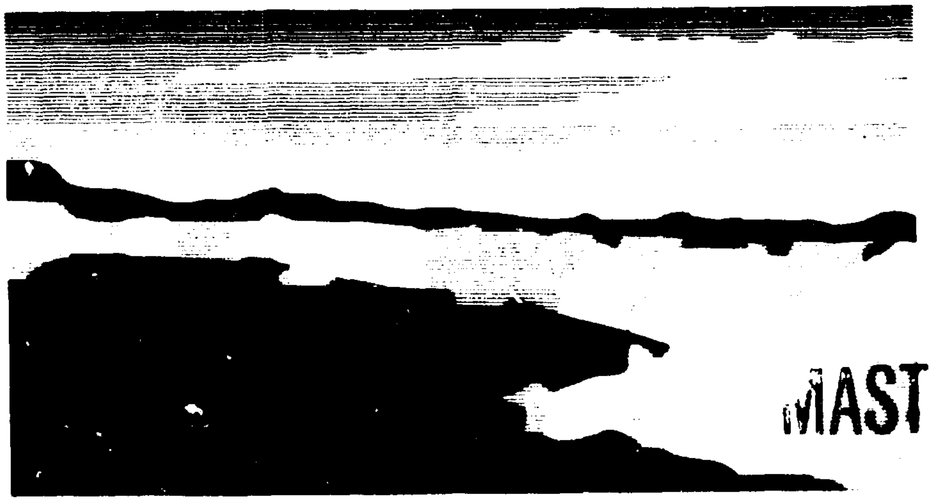

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A LARGE MULTIPLICITY COUNTER FOR THE MEASUREMENT OF BULK PLUTONIUM*

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ABSTRACT

We have considered the problem of designing a thermal neutron counter for the neutron multiplicity measurement of large, high-mass plutonium items. Three neutron multiplicity counters have been built at Los Alamos to date; two are used for in-plant applications. The third counter is an experimental prototype and is used for research. The sample cavities of these counters can accommodate only relatively small samples. The largest item that can be measured optimally by the largest of these counters is 20 cm wide by 36 cm high. Now that the multiplicity technique has proven to be accurate and timely for the measurement of plutonium-bearing items, several facilities in the DOE complex have identified the need for larger counters. Several sources have identified a counter that could measure items contained in 30-gal. drums as the most desirable. For a multiplicity measurement to be successful, the neutron counter must have a large detection efficiency independent of changes in the neutron energy of the emitted neutrons, a short die-away time, and a uniform response over the sample cavity. These requirements have been achieved in the smaller counters by using large numbers of ^{3}He tubes placed in concentric rings. This method can be extrapolated to a larger counter but at great expense. We have conducted a design study for a 30-gal. drum counter. The goal of this study is to reduce the number of ^{3}He tubes required for such a large counter while still maintaining good measurement performance. This paper will report on the results of this study.

INTRODUCTION

In the past several years, neutron multiplicity counting has been shown to be a timely and accurate method for measuring impure plutonium samples.[1-3] At Los Alamos we have designed and built three multiplicity counters: two for in-plant use and a third counter that is a research prototype. In addition, the Active/Passive Counter[4] at the Los Alamos Plutonium Processing Facility has been converted to a multiplicity counter by replacing its conventional coincidence counting electronics with a multiplicity electronics package.

The first in-plant use of one of the Los Alamos multiplicity counters was at the Lawrence Livermore National Laboratory (LLNL). The In-plant or Pyrochemical Multiplicity Counter was loaned to LLNL to aid them in an inventory reduction program. A scaled-down version of the In-plant counter was next built for the International Atomic Energy Agency. This counter, the Plutonium Scrap Multiplicity Counter,[5] is currently used with good results to measure mixed oxide scrap samples in Japan. Most recently, the Active/Passive Counter was refitted and is being used to investigate the measurement of weapons components by multiplicity counting.

The largest container that any of these counters can accommodate and measure optimally is 20 cm wide by 36 cm high. With the current national commitments to disarmament and the moratorium on plutonium production, DOE facilities are redirecting efforts to address the need for the long-term accounting and storage of bulk plutonium oxide and plutonium metal. Historically, bulk plutonium has been stored in drums that hold one or more smaller items surrounded by some kind of shielding material. Plans for future long-term storage of bulk plutonium involve a container about the size of a 30-gal. drum.

Existing neutron counters that have cavities large enough to measure materials in 30-gal. drums generally were not designed to meet the rigid criteria needed to be successful at multiplicity counting. These design criteria include high detection efficiency, low die-away time, uniform response throughout the sample region, and relative insensitivity to variations in the emitted neutron energy spectrum. These design criteria have been achieved in current multiplicity

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counters by using large numbers of ^{3}He tubes optimally placed in a polyethylene moderator. To scale up an existing counter design to accommodate a 30-gal. drum, however, would be prohibitively expensive. For example, if the In-plant Multiplicity Counter were scaled to accommodate a 30-gal. drum, two hundred and twenty 152-cm active length ^{3}He tubes would be needed. The cost of this many tubes approaches $250k.

To address the problem of assaying bulk items by multiplicity counting we undertook a design study of a counter that could accommodate a 30-gal. drum and that would meet the above design criteria in a cost-effective manner. This paper presents the results of that study.

PERFORMANCE PARAMETERS

We began our design study by defining target efficiency and die-away characteristics for a 30-gal. drum multiplicity counter. The three multiplicity counters built at Los Alamos have had detection efficiencies of 53% to 57% and die-away times under 50 μs. Such high efficiencies and low die-away times are required to measure low-mass items or items having moderate (α,n) rates, in a timely manner. Bulk plutonium materials, however, tend to be stored in 2 to 5 kg quantities and have relatively low levels of impurities so that the ratio of (α,n) neutrons to spontaneous fission neutrons emitted by them is relatively low. Thus, the detection efficiency for a counter to measure these materials need not be as high, and the die-away time can be somewhat longer. We decided that we wanted this counter to have a detection efficiency and die-away characteristic such that materials in this range could be measured to a precision of 1-3% in about 30 minutes assuming that the material was relatively pure. Using a figure-of-merit code developed by Ensslin,[6] we determined that for oxide materials in this mass range, a detection efficiency of about 35 to 40% and a die-away time of 50 to 70 μs would yield the desired assay precision.

Next we considered the mechanical layout for this counter. Present multiplicity counters are cylindrical in shape, have multiple rings of ^{3}He tubes, and are loaded from the top. The size and weight of a loaded 30-gal. drum or the proposed AT400 long-term storage container is such that top loading, although not impossible, would raise significant safety concerns. Thus, we decided that the counter would be loaded from the side. To ensure a good spatial response for the counter we decided we would specify 152-cm active length ^{3}He tubes, but that we would change from a cylindrical shape to a hexagonal shape for the counter so that only one third of the counter would have to be opened to load a sample.

CALCULATIONS

With these parameters defined, we began calculations to determine optimal tube number and placement. We used the Monte Carlo neutron transport code MCNP[7] version 4A to do these calculations. All Monte Carlo calculations were performed to a 1σ precision of 1-2%.

We began by scaling the tube spacing used in the In-plant Multiplicity Counter to the hexagonal shape. This model required 210 tubes in 4 rows, and the calculations predicted it would have about the same performance as the In-plant Counter. Next we began selectively removing tubes from the model to reduce the number of tubes and thus the detection efficiency. As we performed these calculations, we also studied the effect of tube removal on the energy and spatial characteristics of the model. We found that the tubes in the fourth row yielded the least efficiency for the most cost and that most of the fourth row could be removed and still achieve the target detection efficiency. However, the tubes in the fourth row of this model were very important to maintaining insensitivity to variations in the emitted neutron energy spectrum.

At this point, we decided to modify the four-row tube arrangement to a three-row model. We then focused on calculating the optimal placement of the three rows to achieve good spatial and energy characteristics and on the reduction in the total number of tubes to reduce cost. With these calculations we learned that the tubes in the corners of each sixth of the counter yielded low detection efficiency relative to their cost but were important to the overall energy characteristics of the counter. We found, however, that if the tubes and polyethylene in the corners were replaced with aluminum reflectors, neutrons would be reflected to the tubes near the corners and good energy characteristics could still be achieved. Also, by carefully choosing the size of the reflector, the resulting counter model was modular and could be adapted to several different door designs.

RESULTS

The counter model that resulted from the above process is shown schematically in Fig. 1.

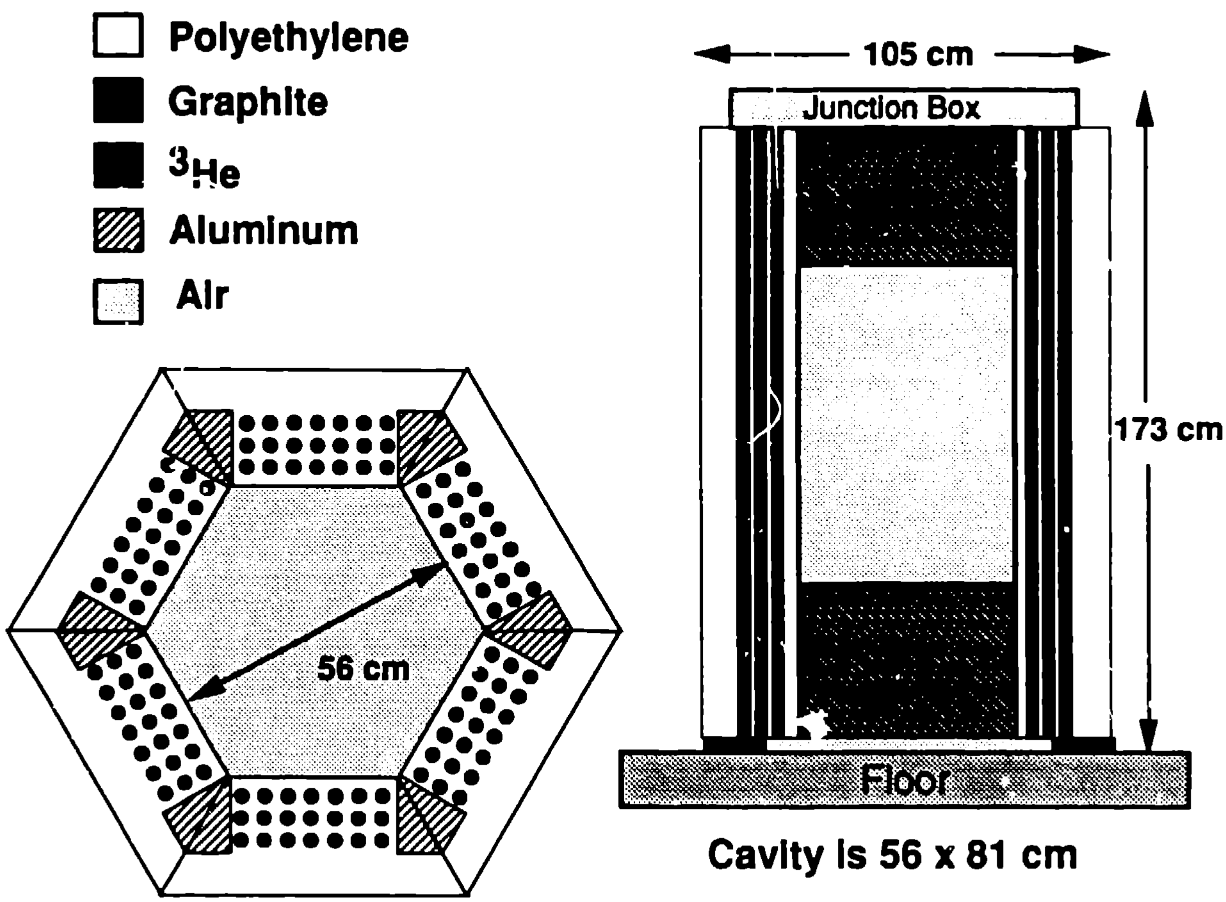

Fig. 1. Schematic diagram of the calculational model for the 30-gal. drum multiplicity counter.

Calculations predict that this counter, which has one hundred and twenty six 152-cm-active-length ^3He tubes, will have an efficiency of 40.6% and a die-away time of 53 µs. Figures 2 and 3 show the calculated spatial characteristics of this counter for a ^{252}Cf point source. These results are plotted relative to the response to that source in the center of the sample chamber. The axial response is comparable to that of the In-plant Multiplicity Counter. The horizontal response is an improvement, however. For cylindrical counters, the horizontal response behaves like that for the narrowest dimension for the 30-gal. drum counter model. Most bulk materials have circular symmetry in the horizontal dimension, however. Because the horizontal response varies in an opposite manner in the narrowest dimension relative to the widest dimension in the hexagonal counter, *on average* there will be smaller overall variations for this counter relative to a cylindrical one if the material is challenging the dimensions of the counter.

Figure 4 gives the calculated response of the counter as a function of the initial energy of detected neutrons. These results are for monoenergetic point sources centered in the cavity and are plotted relative to the response at 2 MeV. For comparison, the energy response of the In-plant Multiplicity Counter is also plotted. The knee in the response at 3-4 MeV for the 30-gal. drum counter is caused by resonant scattering by the large amount of carbon in this system. Figure 5 gives the detection efficiency as a function of row. Row 1 is the innermost row; row 3 is the outermost. When this counter is built, the individual rows will be wired together so that the differences in the energy responses of the different rows can be used to detect sample differences.

Figure 6 gives the predicted assay performance for the counter based on counting statistics. These

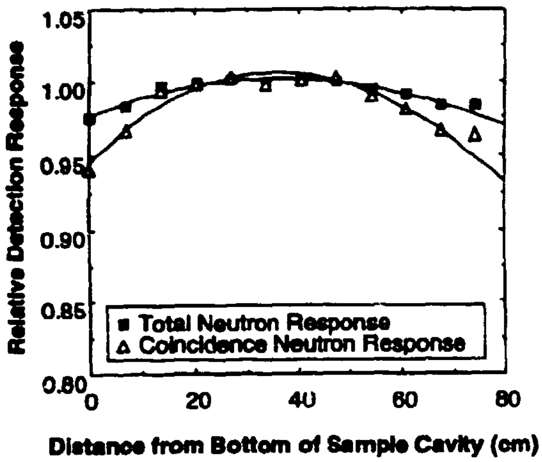

Fig. 2. Calculated axial response of the 30-gal. drum multiplicity counter.

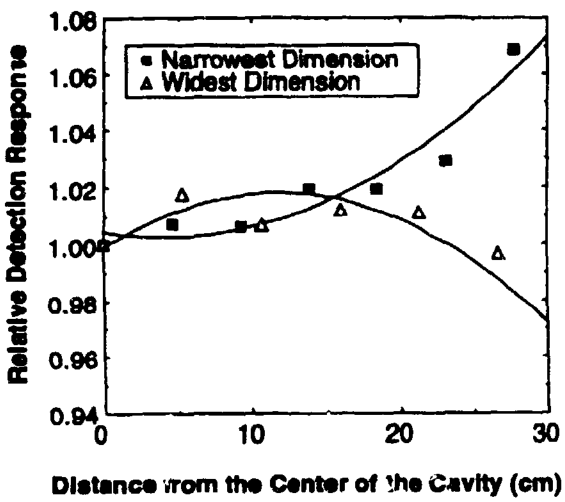

Fig. 3. Calculated horizontal response of the 30-gal. drum multiplicity counter.

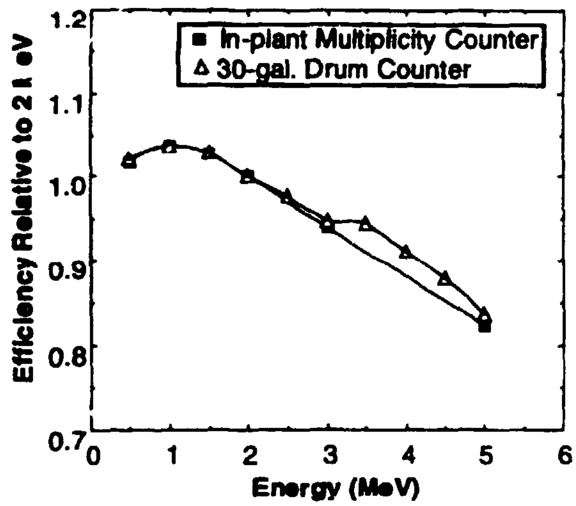

Fig. 4. Calculated response of the 30-gal. drum multiplicity counter as a function of neutron energy.

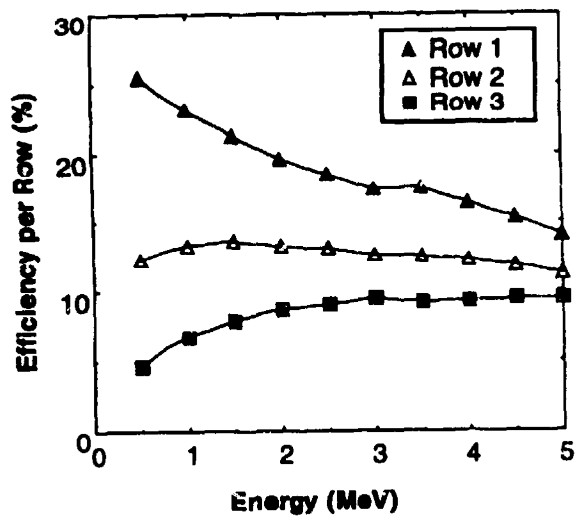

Fig. 5. Calculated detection efficiency of the 30-gal. drum multiplicity counter as a function of row. Row 1 is the innermost row.

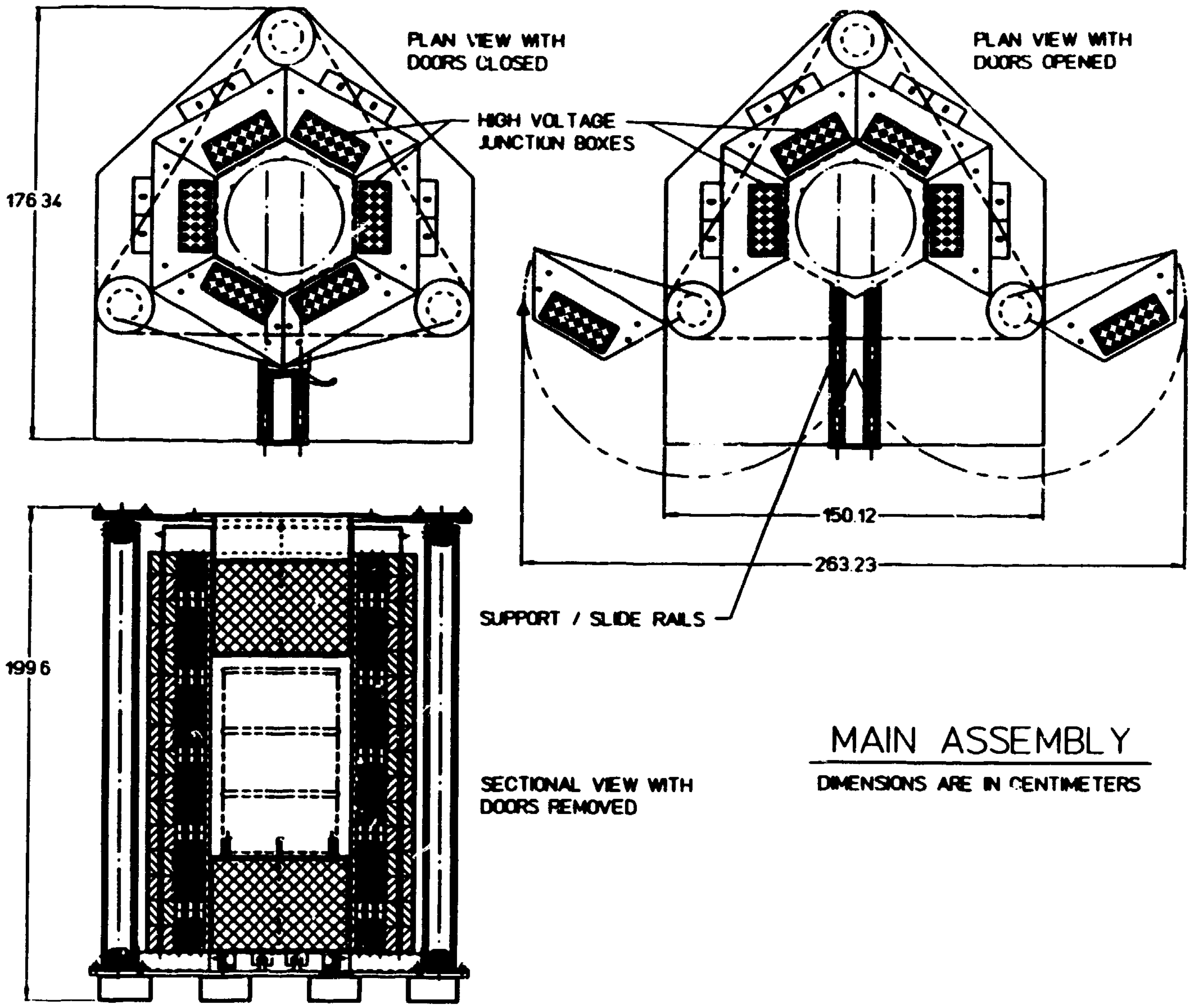

Fig. 7. Mechanical schematic for the first version of the 30-gal. drum multiplicity counter.

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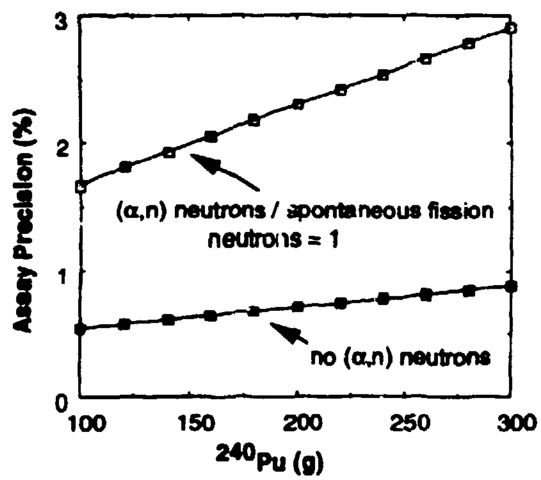

Fig. 6. Predicted assay precision for a neutron multiplicity counter with 40.6% detection efficiency, and a 53 μs die-away time, and a 30-minute count time.

latter calculations were performed using Ensslin's figure-of-merit code and assume that the material being measured is plutonium oxide. The measurement time is assumed to be 30 minutes. Performance for plutonium metals would be superior to this if the metals' geometry is such that the induced fission rate was higher than for a typical oxide.

COUNTER FABRICATION

This counter is scheduled to be fabricated beginning this fiscal year and completed in fiscal 1995. The first version of this counter will have a "French door" loading arrangement as shown in Fig. 7. Each sixth of the counter will be a separate module so that the mechanical design packages for the polyethylene assembly and the junction boxes will not have to be modified if the counter were to be adapted to a different loading scheme The counter modules will be mated to a large plate and hinge assembly that has fork lift tine receivers. The large plate and supports will make the counter very stable mechanically. For facilities having large seismic potential, this plate could also be bolted to a concrete foundation. The lower graphite end plug of this counter will be mounted on rails so that the end plug can be easily removed from the counter for sample loading. This end plug will be moved mechanically for this first version. However, the design will be easily adaptable to motorized movement. The French doors can be opened in several different configurations to accommodate loading either from the front or at an angle to either side. Thus the counter could be placed in a corner to minimize its "footprint," but loading could still be accomplished from the side.

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