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A Dual-Reservoir Remote Loading Water Target System for ^{18}F and ^{13}N Production with Direct In-Target Liquid Level Sensing

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GENERAL FEATURES

This report describes our universal water target loading system that serves both [^{18}F] and [^{13}N] production targets, and a radionuclide delivery system that is specific for [^{18}F]fluoride. The system was designed and fabricated around the operation of a single pneumatic syringe dispenser that accesses one of two reservoirs filled with [^{18}O]enriched water for [^{18}F]fluoride production from the $^{18}O(p,n)^{18}F$ reaction (Wieland and Wolf, 1983; Kilbourne, *et al.*, 1984, 1985; Huszar and Weinreich, 1985; Keinonen, *et al.*, 1986; Iwata, *et al.*, 1987.), and natural abundance water for [^{13}N]nitrate/nitrite production from the $^{16}O(p,\alpha)^{13}N$ reaction (Parks and Krohn, 1978; Tilbury and Dahl, 1979; MacDonald, *et al.*, 1979; Slegers, *et al.*, 1980), and loads one of two targets depending on the radionuclide desired.

The system offers several novel features for reliable radionuclide production. First, there exists an in-target probe for direct liquid level sensing using the conductivity response of water. In addition, transfer of [^{18}F]fluoride to the Hot Lab is completely decoupled from the irradiated water through the actions of a resin/recovery system which is located in the cyclotron vault, thus maintaining transfer line integrity. This feature also provides a mechanism for vault-containment of long-lived contaminants generated through target activation and leaching into the water.

SYSTEM DESCRIPTION

Target Loader: Figure 1 shows the valve layout of the dual-reservoir remote water target loading system configured for multiple radionuclide production. The system is designed around a pneumatic syringe dispenser (Hamilton Co.) that is operated with 40 psig of air. The dispenser is actuated electronically by pulsing a solenoid which momentarily vents pressure on the pneumatic arm. This forces the

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syringe to retract its plunger a preset distance thus withdrawing a charge of water from one of the reservoirs. A position sensitive pneumatic-operated three-way valve (part of the Hamilton syringe dispenser) then positions itself prior to the syringe completing its stroke thus allowing the withdrawn charge of water to be automatically dispensed through a short 1 m x 3.2 mm i.d. polyethylene transfer line enroute to the target. This sequence is again repeated whereupon the syringe withdraws and dispenses an equivalent volume of helium gas at 6 psig pressure. This action not only completes the transfer of water to the target volume, but also purges the syringe and extraneous plumbing of any residual water prior to the next target load. This minimizes cross-contamination of enriched and natural abundances waters which helps to insure proper maintanence of isotopic integrity.

The two water reservoirs are fabricated from standard pyrex glass tubes (10 cm x 2.5 cm i.d.). All flexible lines leading to and from the reservoirs via assorted valves are fabricated from 3.2 mm o.d. polyethylene tubing. All valves that are part of this plumbing were manufactured by General Value Co. (Fairfield, NJ). Tubing lengths are kept to a minimum in order to avoid possible organic contamination to the water. Access lines leading into the reservoirs themselves are fabricated from small i.d. glass tubes. Each reservoir requires a different i.d. access tube in order to restrict the volume of water withdrawn. This is necessary because the ^{18}F and ^{13}N targets require different volumes of water for filling. Connections are made between the glass reservoirs and flexible tubing using standard Instac fittings (Lee Valve Co. Westbrook, CT).

Target Components: The side-views of both production targets depicted in Figure 1 show the key features of the target design. The main target chambers are fabricated from silver. Key features to the target components include a 316 stainless coiled loop (2 mL volume) which serves as an expansion volume for the water during initial target loading. This is necessary because the target volume is somewhat smaller when the front window (1 miL thick titanium foil) is flat. Upon pressurization for irradiation (14 psig), the water is displaced back into the target volume as the front window deforms outward.

Another feature includes the miniature 3-way solenoid valves manufactured by Lee Valve Co. (12 VDC LFHX series, with Minstac fittings and capable of operating up to 100 psig) which direct loading water to the bottom inlet port of the target, seal the target for pressurization during irradiation, and then direct the irradiated water

out of the same port to the resin/recovery system for [^{18}F]fluoride production, or directly to the Hot Lab for [^{13}N]nitrate/nitrite production. One of the ports on the 3-way valve was plugged so that it operated as a 2-way shut-off valve.

Figure 2 shows a front-faced view of a water target without its window. The water sensor is inserted through one of the upper target ports. This sensor is fabricated from two platinum wires that are encased in polyethylene for electrical insulation. Each length is then fed through a 2 mm length of 6.25 mm o.d. teflon tube which is used to seal the sensor lines onto the back-end of the target chamber using a standard HPLC fitting with metal ferrules crimped onto the teflon tube. This made a gas-tight seal onto the target. Positioning of the sensor leads within the target does not appear to be crucial to the performance of the conductivity meter. The leads are fixed with approximately a 1 mm gap between the tips. The sensor is mounted up in the keyhole region of the target so as to avoid direct beam strike and radiation damage to the polyethylene insulators.

Liquid level sensing is accomplished by measuring the conductivity of the water. The meter used was manufactured by Crystalab Inc. Hartford, CT (120 volt AC, 60 cycle, 1/25 watt). The [^{18}O]-enriched water used (Isotec, Miamisburg OH) for [^{18}F] production has more than adequate levels of ions for good sensor response. However, response does fall-off with subsequent distillations of the water during reprocessing. In addition, response is low to the natural abundance water used (steam distilled) for [^{13}N] production. Satisfactory response levels are maintained by placing a strand of silver wire in each of the water reservoirs.

[^{18}F]Fluoride Resin/Recovery System: Figure 3 depicts a general schematic of the [^{18}F]fluoride resin/recovery system that connects to the [^{18}F] production target outlet line. The system for processing [^{18}F]fluoride consists of an anion exchange column (Bio Rad AG1X8 carbonate form; 200-400 mesh) similiar to previously reported (Schlyer, *et al.*, 1990), a [^{18}O]-enriched water recovery vial, two "single-shot" carbonate reservoirs, a timing relay with timedown selector switches, and several switching valves used to direct the flow of carbonate solution and the radionuclide. All valves used in this system are manufactured by Lee Valve Co as described earlier.

The anion resin is supported on a fine glass frit when packed into the glass column. Column dimensions of approximately 6 mm x 1 mm i.d. yield optimized fluoride extraction and recovery. Repacking of the anion resin column, as well as the refilling of the carbonate reservoirs is carried out on a per run basis.

A second charge of carbonate solution is available for resin rinsing sometimes to maximize radionuclide recovery, depending on the nature of the [^{18}F] synthesis, but most often it is done as a way to obtain nonproduction levels of [^{18}F]fluoride for research purposes later in the day.

In operation, the resin/recovery system energizes when the target is commanded to unload, but counts down 160 seconds before carrying out its designed tasks. During this timedown, irradiated water passes through the resin column, and is collected, while [^{18}F]fluoride is retained on the resin. Once the timedown is complete, a charge of carbonate solution is dispensed automatically through the resin to remove the bound fluoride, and deliver the radionuclide to the Hot Lab. Transfer times generally are less than 5 minutes (exclusive of the initial 160 second timedown) through 40 m x 0.5 mm i.d. polyethylene tubing.

System Control: The target control box, resin/recovery control box, and regulated 12 VDC power supply, depicted in Figure 4, control target loading and unloading, water recovery when [^{18}O]-enriched water is used, and [^{18}F]fluoride or [^{13}N]nitrate delivery to the Hot Lab. These control systems are all located in the Hot Lab for easy access by the operator.

The sequences for target loading, target pressurization for irradiation, and target unloading are controlled by a single selector switch on the main system box. The state of this switch is interlocked with the operation of all peripheral hardware in the vault through a series of hardwired relays. When in the target "Load" state, the dispenser is interlocked to a position sensitive microswitch which displays a ready-light, and energizes the start-load button only when correctly positioned. An electronic counter associated with the selected reservoir is also energized at this time, and registers the event so that operators can monitor the fill status of the reservoir. The conductivity meter is interlocked to the sequence selector switch so that it is only energized during the target "Load" state to avoid possible electrolytic breakdown of the water.

When in the "Irradiate" state, the target is pressurized with 14 psig of helium gas, and is ready to accept beam. To unload the target, the selector switch is turned back to the "Load/Unload" position. A latching relay couples the target "Unload" status to a sequence of events that allows it to be activated only after the selector switch passes through the "Load" and "Irradiate" states of the target. Irradiated water is then automatically processed through the resin/recovery system.

The resin/recovery control box designed for [^{18}F]fluoride delivery, allows the operator to divert the radionuclide delivery path to one of two hot cells located in the Hot Lab, or to a shielded vial residing in the vault. The operator also has control to release second charge of carbonate solution through the resin column, once the first charge has been recovered in the lab. This second charge can be delivered to either of the three locations.

SYSTEM PERFORMANCE

The universal water target loading system has been installed at BNL and operated for approximately 8 months, and is used almost daily for [^{18}F] production. Since its installation, we have never had a failure in the loading mechanism nor in the sensor performance. Target loading times have been greatly reduced through an operation that now requires less than a minute to carry out. Target unloading times have also been reduced significantly.

The methods implemented for [^{18}O]-enriched water storage in the sealed glass reservoirs, for target loading through the syringe dispenser, and for liquid level sensing using conductivity probes have had no deleterious effects on [^{18}F] target yields, nor on [^{18}F]radiotracer specific activity. Typically, the system will deliver approximately 360 mCi of [^{18}F]fluoride in the first rinse of carbonate solution from a 20 minute irradiation (using 17 MeV protons on target at 15 μA intensity) to yield final product (FDG) specific activities ranging between 1 and 3.5 Ci/μmol (at end-of-bombardment). An additional 10 to 30 mCi of [^{18}F]fluoride can usually be recovered in the second rinse of carbonate solution.

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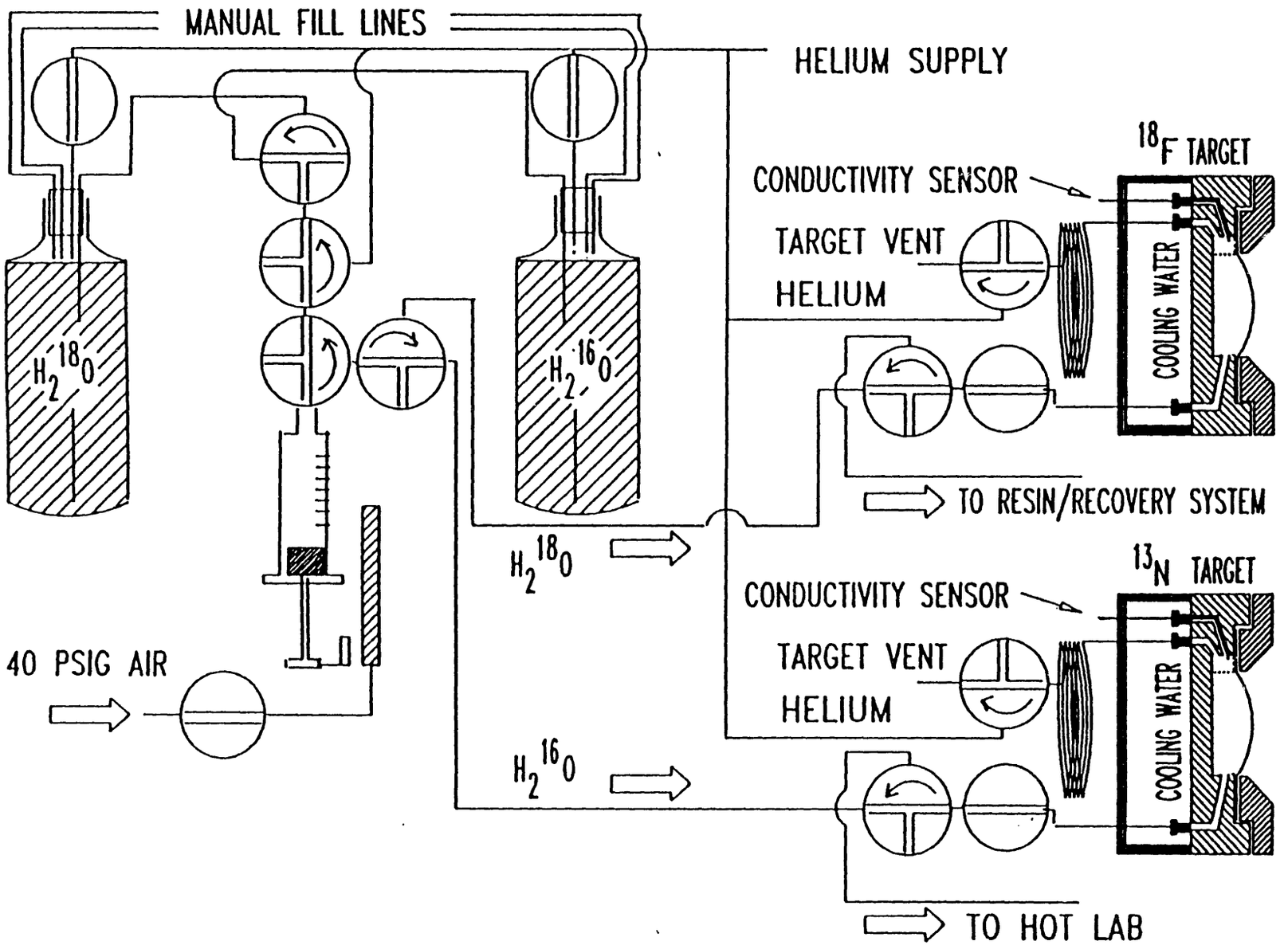

Figure 1 Schematic diagram of the water target loading system and production targets for [^{18}F]fluoride and [^{13}N]nitrate/nitrite.

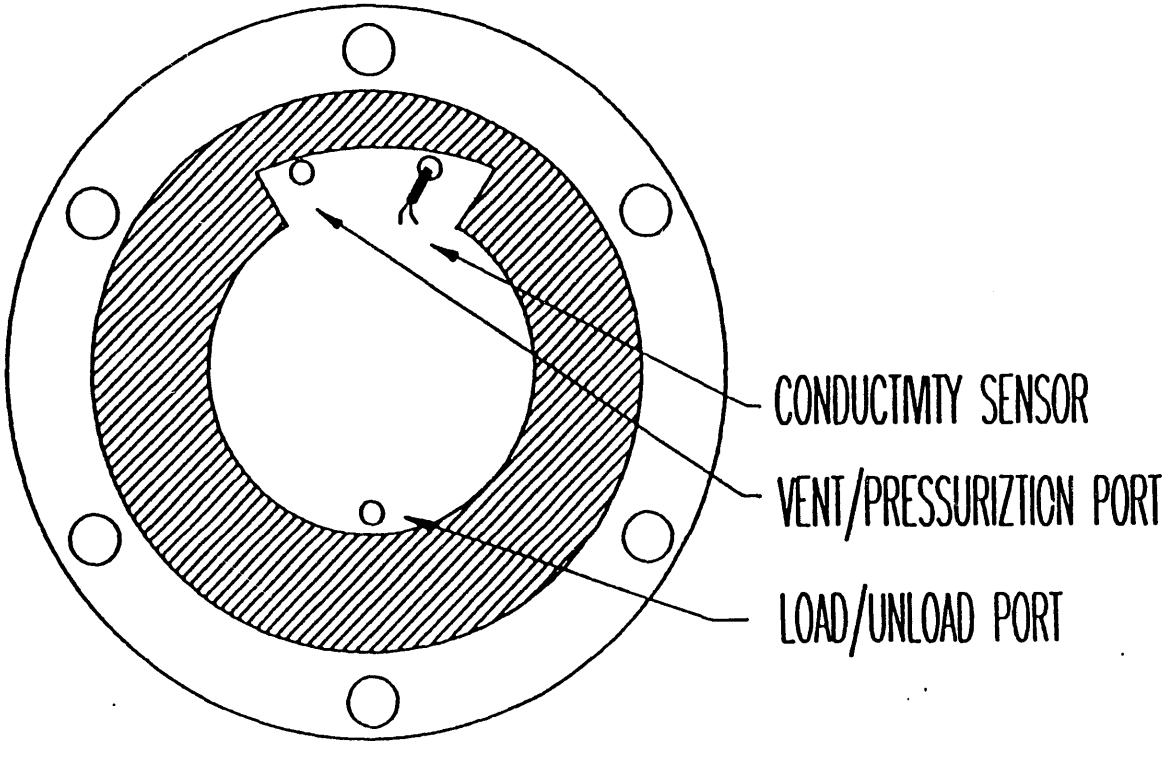

Figure 2 Front-faced view of a water target showing the conductivity sensor positioned in the keyhole region of the target body.

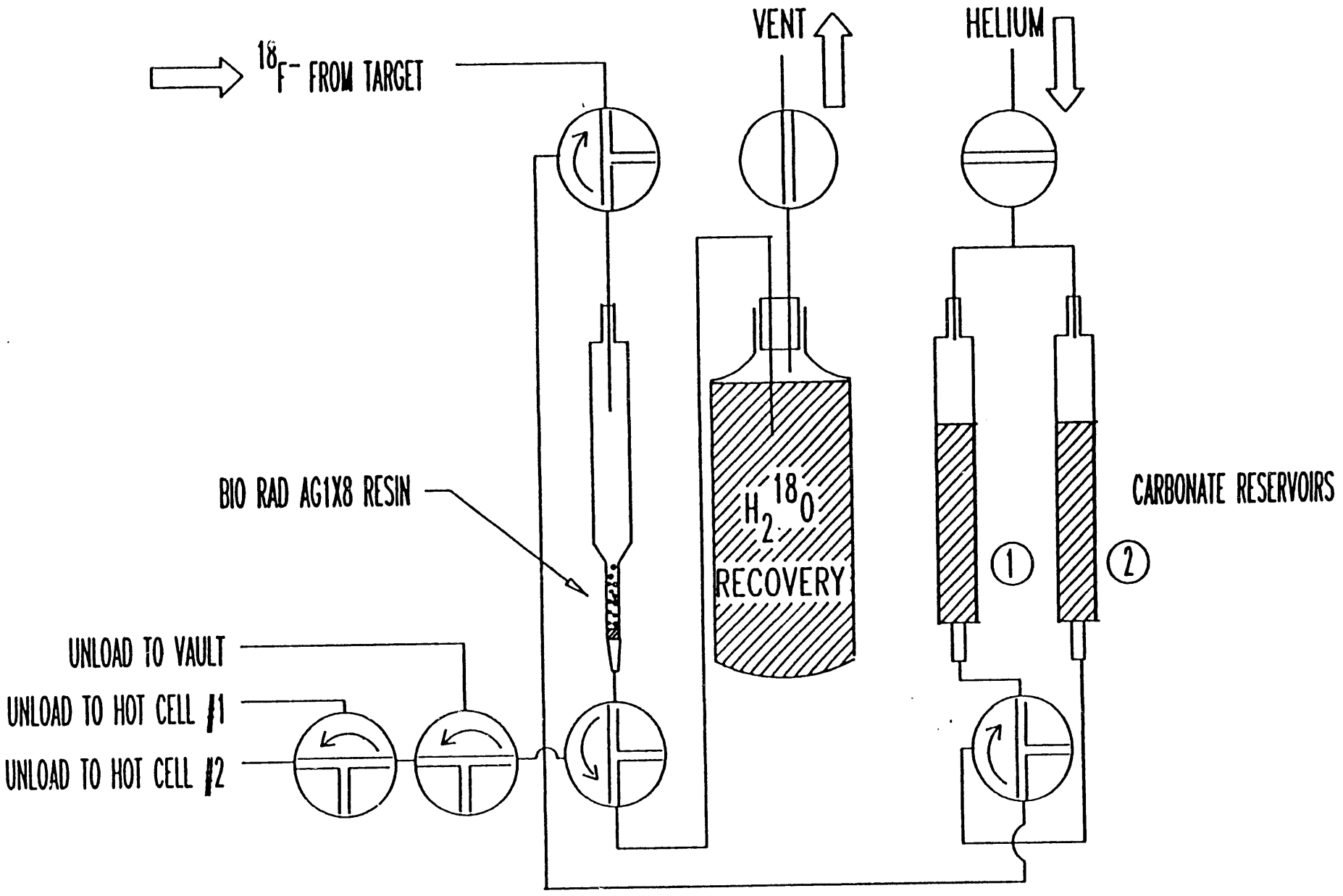

Figure 3. Schematic diagram of the resin/recovery system for processing [^{18}F]fluoride.

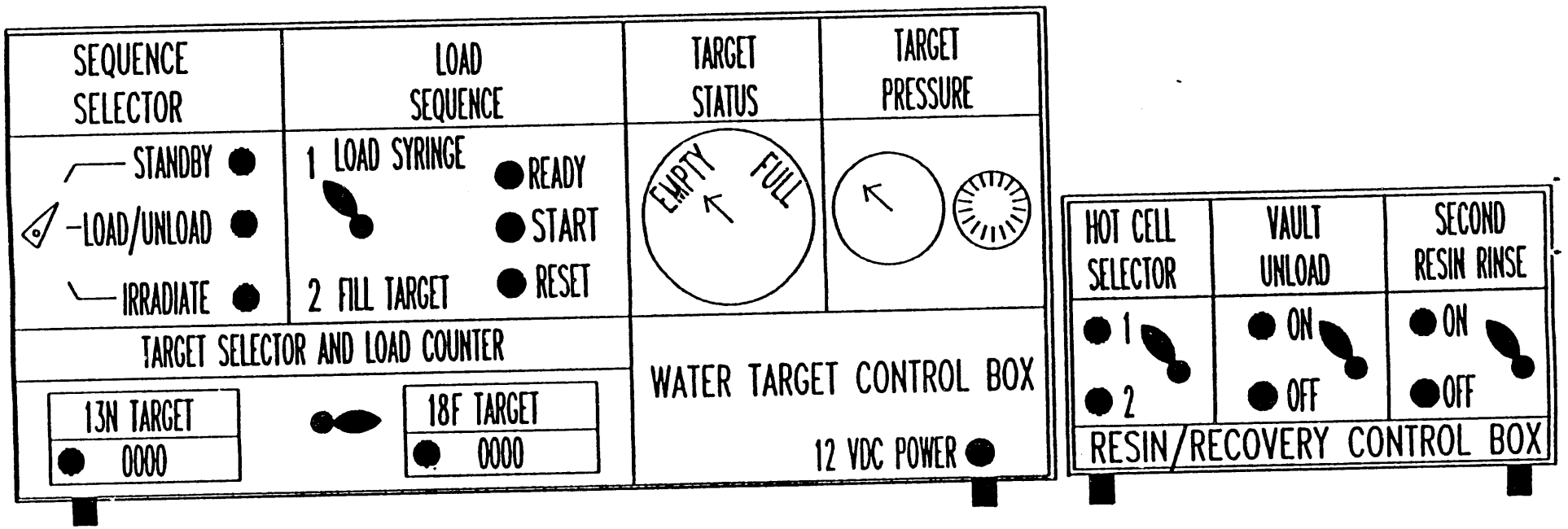

Figure 4. Front-faced view of the Water Target Control Box and the Resin/Recovery Control Box for [^{18}F]fluoride processing..

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