

The Development of the ^{17}F Beam at the Holifield Radioactive Ion Beam Facility

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Abstract

This report details some of the key technological developments employed at the Holifield Radioactive Ion Beam Facility (HRIBF) to produce beams of ^{17}F using the $^{16}\text{O}(\text{d},\text{n})^{17}\text{F}$ reaction. The oxide fiber target material used at the HRIBF is described and a comparison is made between the ^{17}F yield achieved using light (Al) and heavy (Hf) metal oxide fibers. The development of the Kinetic Ejection Negative Ion Source (KENIS) employed in this work is also discussed along with the operational principles of the source. Finally, a detailed description of the HfO_2 target configuration used to produce 10^7 - 10^8 ^{17}F ions/s for over 850 hours of operation is provided. To date, seven nuclear physics experiments using accelerated beams of ^{17}F and ^{18}F produced using this apparatus have been performed over an energy range of 10-170 MeV.

Key words: 29.25.N Ion sources: positive and negative, 29.25.Rm Sources of radioactive nuclei, 07.20.K High temperature techniques and instrumentation, 26.30.+k Nucleosynthesis in novae, supernovae and other explosive environments, 29.27.-a Beams in particle accelerators, 25.45.Hi Transfer reactions with 2H

1 Introduction

The Holifield Radioactive Ion Beam Facility (HRIBF) at the Oak Ridge National Laboratory (ORNL) is devoted to the production of low-energy radioactive ion beams (RIBs) for nuclear structure and astrophysics research. Radioactive ions are produced by directing light ion beams accelerated by the $K = 100$ Oak Ridge Isochronous Cyclotron (ORIC) onto thick, hot, refractory targets. The radioactive atoms diffusing out of the target material are ionized and injected into the 25-MV tandem accelerator producing beams with energies of 0.1 - 10 MeV per nucleon for light nuclei and up to 5 MeV per nucleon for mass 80 (1; 2). Since the tandem accelerator requires injection of negative ions, the RIB can either be produced directly in a negative-ion source or first

positively ionized and then passed through a charge-exchange cell containing a metallic vapor to form a beam of negative ions.

There has been a long standing interest in performing cross section measurements with beams of ^{17}F because of the critical role this species plays in hydrogen burning reactions occurring in stellar explosions (3; 4). Several approaches have been taken to produce RIBs of this species. The ISOLDE group has produced ^{17}F by bombarding a 19 g/cm^2 SiC target, which was integrated into a high temperature target/ion source, with a few μA of 600 MeV protons. The presence of residual Al vapor contamination allowed the extraction of Al^{17}F^+ from the ion source at a rate as high as $\sim 10^7$ ions/s (5). Alternatively, an in-flight technique has recently been used to produce beams of ^{17}F at ANL (6). In this approach, accelerated beams of stable ^{16}O or ^{17}O are passed through a high pressure gas cell containing ^1H or ^2H and the reactions $p(^{17}\text{O},^{17}\text{F})n$ or $d(^{16}\text{O},^{17}\text{F})n$ occur. A superconducting solenoid is then used to collect and focus the emerging ^{17}F beam. Intensities as high as 2×10^6 ions/s on target have recently been produced at energies of 65-110 MeV. The HRIBF has recently produced $\sim 10^8$ Al^{17}F^+ ions/s from a positive-ion source using the $^{16}\text{O}(d,n)^{17}\text{F}$ reaction by bombarding a fibrous matrix of HfO_2 with $2\ \mu\text{A}$ of 45-MeV deuterons from the ORIC accelerator in the presence of a steady-state flow of elemental Al vapor (7). Unfortunately, the process of converting $\text{Al}^{17}\text{F}^+ \rightarrow ^{17}\text{F}^-$ by passage of the beam through the charge-exchange cell is quite inefficient and also results in considerable energy spread in the emerging $^{17}\text{F}^-$ beam. For this reason, we have developed a negative-ion source (KENIS) for producing F^- directly and eliminating the need for the charge-exchange cell. This article describes the target material, design of the target system, and ion source configuration that has allowed us to now achieve intensities of $^{17}\text{F}^-$ comparable to those of Al^{17}F^+ at the HRIBF.

2 HfO_2 target material

There are several mass transport processes radioactive nuclei must undergo between the time of their formation and the time of their ionization: diffusion through the target material, effusion through the void space in the target material, and effusion through the ion source enclosure to the ionizer. RIB yields will be compromised if any of these times are long compared with the half-life of the radioactive species. In solids, even at high temperatures, diffusion is a comparatively slow process and steps must be taken to reduce the distance traveled by particles diffusing through the material to lengths much less than the dimensions of the overall target. This is usually done by employing fine elemental powders, powders of compounds, powder mixtures with graphite or noble metals or for metallic species, very thin dimpled foils. Such materials are characterized by diffusion lengths of several micrometers. Unfor-

tunately, when heated to operating temperatures, powders tend to sinter quite readily (8). This is particularly true for oxides where extremely high oxygen self-diffusivities can rapidly densify the material (9).

Commercially available metal oxide fibers are an attractive alternative to powders and are available in the form of flexible sheets that are easily rolled or cut into desired shapes. The ISOLDE group was the first to employ such targets for RIB generation (5) and later investigations were conducted at the HRIBF using fiber targets of Al_2O_3 (10; 11) and HfO_2 (7) for ^{17}F production. The oxide fibers used at the HRIBF have been manufactured by Zircar Corporation using their precursor technique (12). In this process, a solution containing the metal chloride is prepared (e.g., $\text{ZrOCl}_2 \cdot 8\text{H}_2\text{O}$) and allowed to chemically impregnate an organic host textile (e.g. rayon). During impregnation, a process akin to fossilization occurs where the solution is actually drawn into the host fibers. Once this step is complete, the sample is dried and fired to drive off the organic material and a fossil replica of the original textile remains (13). Figure 1 shows a Scanning Electron Microscope (SEM) image of a single HfO_2 fiber taken at a magnification of $5000\times$. The resulting structure is mechanically resilient and consists of an interlocked network of fibers which is quite resistant to sintering at high temperatures. For example, we have heated HfO_2 fibers to temperatures ~ 2150 C in a vacuum furnace for a period of ~ 10 hours and observed little evidence of sintering. The HfO_2 targets have been operated on-line for times of ~ 1000 hours while maintaining nominal ^{17}F yield. Standard materials available in this form through Zircar Corporation include unstabilized/stabilized zirconia (ZrO_2), yttria (Y_2O_3), ceria (CeO_2), alumina (Al_2O_3), hafnia (HfO_2), and titania (TiO_2). These products are available as 4-6 μm fibers in the form of a flexible cloth several mm thick with an uncompressed density of 3-8% of the pure material. Also available through Zircar Corporation on a custom fabrication basis are fibers of La_2O_3 , Nd_2O_3 , Sm_2O_3 , Eu_2O_3 , Gd_2O_3 , Tb_2O_3 , Dy_2O_3 , Ho_2O_3 , Er_2O_3 , Tm_2O_3 , Yb_2O_3 , and Lu_2O_3 .

Given the large number of available metal oxides, we must now select the best material for ^{17}F production using the $^{16}\text{O}(\text{d},\text{n})^{17}\text{F}$ reaction. Targets constructed of oxides of low-Z metals provide higher production rates of ^{17}F nuclei due to the greater range of the production beam within the material. Higher-Z metal oxides, on the other hand, make thinner targets but have a more refractory character and can therefore operate at higher temperatures and withstand higher production beam intensities. To explore this tradeoff, let us consider both a high- and a low-Z metal oxide: Al_2O_3 and HfO_2 . Estimations of ^{17}F production rate can be determined from experimental cross sections (14) together with calculated energy loss functions (15) assuming the deuteron beam is stopped within the target. According to these calculations 40-MeV deuteron beams incident on Al_2O_3 and HfO_2 targets have production rates of 8.5×10^9 nuclei/ μC and 4.6×10^9 nuclei/ μC , respectively. Thus, from this

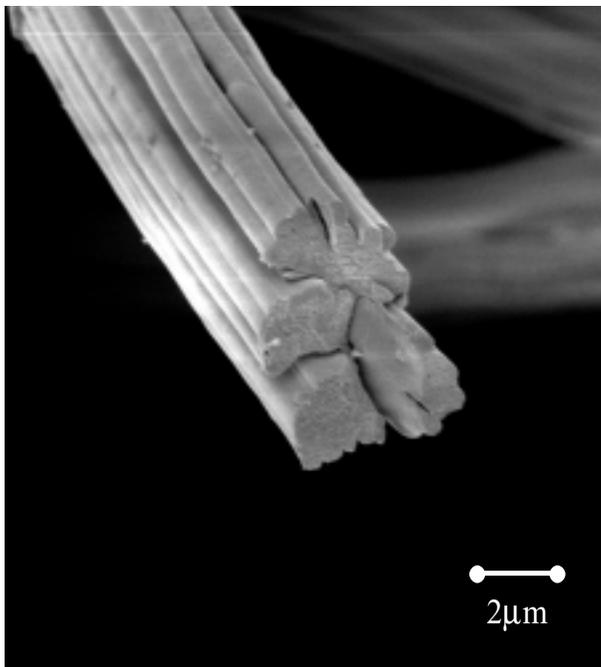


Fig. 1. SEM photograph of a single HfO_2 fiber taken with a magnification of $5000\times$.

point of view, Al_2O_3 targets will produce about a factor of two more ^{17}F than the HfO_2 target.

Thermodynamic equilibrium computer codes such as HSC (16) can be used to estimate the vapor pressures and maximum temperatures for thermal stability of various materials. According to the calculations, the temperature at which the total vapor pressure of Al_2O_3 and HfO_2 is $\sim 10^{-4}$ Torr is 1825 C and 2300 C, respectively. These codes can also calculate the temperature at which gaseous components dominate the equilibrium composition (maximum temperature for thermal stability) and this temperature was determined to be 1700 C and 2200 C for Al_2O_3 and HfO_2 , respectively. Thus, the calculations strongly suggest that the HfO_2 material should be able to operate at ~ 500 C higher temperatures than the Al_2O_3 material.

Two separate experiments were performed to compare the Al^{17}F^+ yield resulting from the use of Al_2O_3 and HfO_2 fibers as target material. Each material was loaded into identical Electron Beam Plasma Ion Sources (EBPIS) (10) and bombarded with a deuteron beam of variable intensity with an energy ~ 45 MeV from ORIC. Al vapor was present in both cases and ion source operating parameters were adjusted to optimize the yield of Al^{17}F^+ . Figure 2 shows the resulting Al^{17}F^+ yield as the production beam current was increased on target. The ionization efficiency of the source was monitored during these experiments using a calibrated leak of stable Xe gas and was found to be similar (3-5%) for each target material. The Al^{17}F^+ yield from the Al_2O_3 material is much less than the corresponding yield from the HfO_2 material for approximately

equal target temperatures. This suggested that the Al_2O_3 fiber material had been damaged by beam heating at even moderate beam intensities of $\sim 1 \mu\text{A}$ and subsequent inspection of the material confirmed this hypothesis.

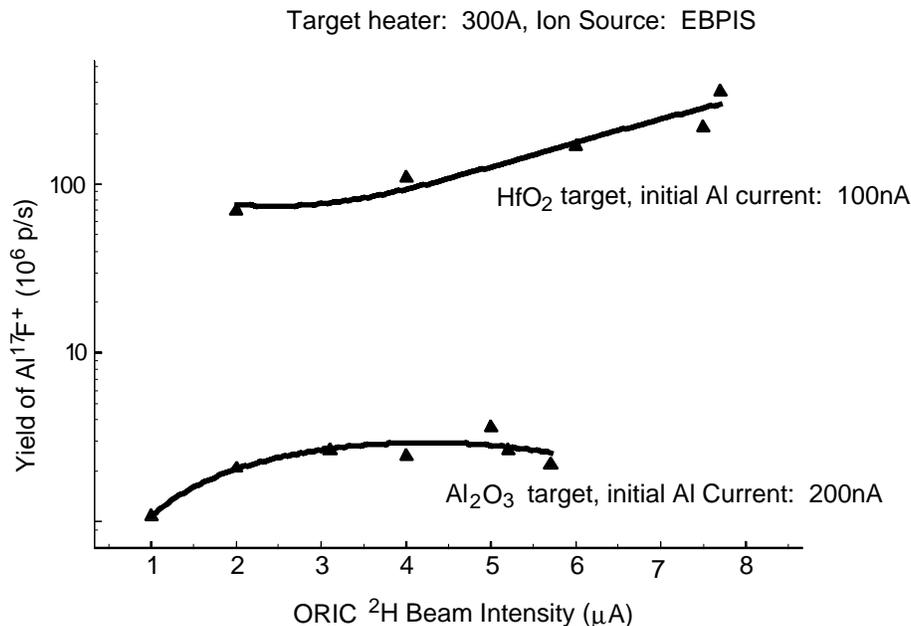


Fig. 2. Al^{17}F^+ yields obtained from an EBPIS with HfO_2 and Al_2O_3 target materials.

It is clear from this data that use of the HfO_2 target material produces beams of Al^{17}F^+ which are approximately two orders of magnitude greater in intensity than could be produced using the Al_2O_3 material. It appears that the Al_2O_3 material simply cannot be operated at temperatures high enough to insure efficient release and transport of ^{17}F from the target material. The use of the HfO_2 target was a significant milestone in the development of the ^{17}F beam since a suitable target material had now been found.

3 The negative-ion source

Over the last several years we have been developing a direct negative-ion source for $^{17}\text{F}^-$ because it was known that the efficiency for converting $\text{Al}^{17}\text{F}^+ \rightarrow ^{17}\text{F}^-$ by charge exchange is quite low. The development of the ion source was conducted in a dedicated, off-line test facility where many iterative changes to the ion source could be made with little impact on HRIBF facility operation. Before beginning to develop the ion source, we first needed an expedient method to measure the ionization efficiency and delay-times for reactive species such as F at very low flow-rates. We found that by injecting stable, gaseous molecules containing a high percentage of the species under investigation across a sharp

thermal gradient into the target reservoir, ionization efficiencies could be resolved. The molecule was chosen to thermally dissociate at the temperatures of the target/ion source allowing chemical reactions to occur as they would in the on-line process. Since these techniques use stable species, delay-times were explicitly measured using a high-temperature, fast valve technique (17).

Using these methods, we began investigating the ionization efficiency for F in a LaB₆ surface ionization source similar to the source described in reference (18) with the addition of an ionization surface composed of pure LaB₆. A LaB₆ ionization surface was chosen because it was successfully used in production of RIBs of Cl, Br, and I isotopes and recommended for targets not containing carbon (19). After a number of experiments it became clear that an efficiency $\sim 10^{-6}$ for F was the maximum we could achieve. This measurement was in contrast to the expectation of a very high ionization probability for F predicted from its large electron affinity (20).

The reason for this low ionization efficiency measurement became clear after inspection of experimental data taken from the positive-ion source during bombardment of a HfO₂ target in the presence of a small quantity of Al₂O₃. A mass scan revealed virtually all of the ¹⁷F activity was in the Al¹⁷F⁺ mass channel, with only a small fraction in the elemental ¹⁷F⁺ channel, which could likely be attributed to dissociative ionization of AlF under electron impact (10). Given the fact the AlF has a very low electron affinity (<0.5 eV) (21), the low values of the ionization efficiency measurements could be explained from the low surface ionization probability of AlF compared with F (20). Thus, in order to efficiently negatively ionize ¹⁷F, we needed to find a way to first dissociate the molecule and then ionize the highly electronegative F.

Since the late 1960's sputter-negative-ion sources were employed as heavy-ion injectors for tandem accelerators (22; 23). It was observed that halogen beams produced by these sources contained only weak molecular sidebands compared with the primary beam even though metal halide salts were typically employed as the sputter surface (24). Insight into the dissociation process occurring during sputtering can be gained from the field of Secondary Ion Mass Spectrometry (SIMS). In the case of negative SIMS, an energetic Cs⁺ ion beam is directed onto a surface and secondary negative ions are ejected from the surface through a sputtering/surface ionization process and then mass analyzed and detected. Such instruments are especially sensitive to trace quantities of halogens including F where they are detected as elemental beams (25). The basis for the high sensitivity to halogens is due to their high electron affinities as well as the efficient transfer of energy from the primary beam to the sputtered species promoting dissociation. During the sputtering process, particles are ejected in an energy distribution that has a maximum at energies comparable to the binding energy of the sputtered material (26). Thus, molecular species adsorbed on a Ta surface will, on average, be knocked off with an ap-

proximate energy of 8 eV or the sublimation energy of Ta, which is sufficient energy to dissociate simple metal halides.

In mid 1997, the first ion source based on this principle was constructed and tested at the HRIBF (27). The source consisted of a resistively heated vapor transfer tube ($\phi \sim 1$ cm) leading from the target reservoir to a tubular, coaxial, Negative Ionization Surface (NIS). The transfer tube was operated at nominal temperatures of ~ 1400 C and used to direct the flow of nuclear reaction products effusing from the target onto the much cooler NIS (~ 350 C) where ions were formed and extracted. Cs vapor was introduced into the ion source where it was efficiently ionized on the hot surface of the transfer tube while simultaneously condensing on the cooler, NIS. The NIS was biased negatively with respect to the transfer tube, causing Cs⁺ ions to bombard it's surface with energies of 300-500 eV. This bombardment caused the condensed radioactive species to be ejected from the surface and caught in the extraction field of the accelerating potential. This source was tested in the manner described above and yielded efficiencies for stable F of $\sim 1\%$. This was also a significant step forward in the development of the ¹⁷F beam, since a $\sim 10^4$ fold improvement in ion source efficiency was realized compared to the LaB₆ source.

After successfully demonstrating stable F could be ionized efficiently using the kinetic ejection process, we then designed an optimized source for use in on-line production of ¹⁷F. Figure 3 shows a view of the ion source and extraction electrode. By adding a grid that could be maintained at an independent voltage with respect to the transfer line and NIS, we were able to repel the ejected F⁻ out of the ion source and into the beam. Typically, the source was operated at a potential of 0 V on the transfer line, -300 V on the grid and a potential of -250 V on the NIS, effectively creating an electric field to repel F⁻. The NIS had also evolved into a conical shape to improve the extraction optics. A separate Cs vapor channel had also been added to allow the delivery of more intense Cs⁺ current to the NIS. Currently ionization efficiencies for stable F have reached $\sim 6\%$ (28).

4 On-line ¹⁷F production

Earlier on-line studies have shown that the yield of Al¹⁷F⁺ from the positive-ion source was greatly enhanced by feeding elemental Al vapor into the target reservoir containing a fibrous HfO₂ target (7). In contrast to this result, the yield of ¹⁷F⁻ from the KENIS showed no enhancement by flowing elemental Al vapor into the target reservoir also containing a HfO₂ target. In separate experiment, we have shown that the yield of ¹⁷F⁻ from the KENIS could be considerably enhanced by placing a small amount of Al₂O₃ fibers directly in the target reservoir containing a HfO₂ target.

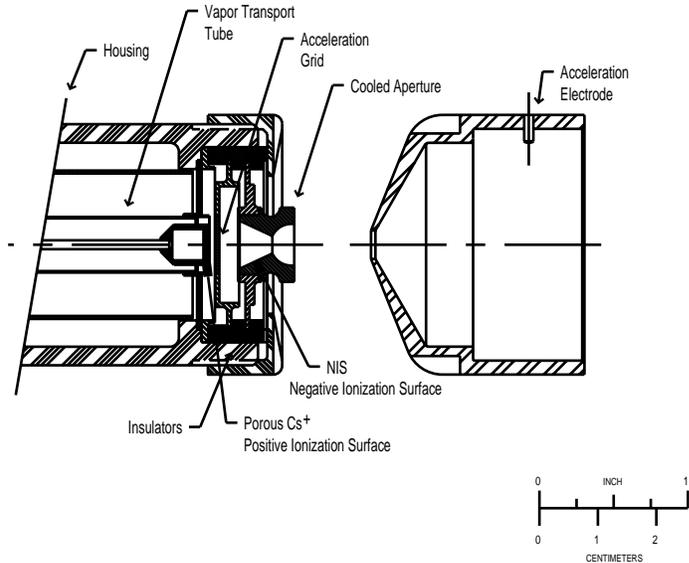


Fig. 3. View of the optimized KENIS and extraction electrode.

In December 1999, we installed a target/ion source on the RIB injector at the HRIBF. The target material was described in Section 2 and the negative-ion source was shown in Figure 3 of this paper. Figure 4 shows the target configuration employed. The cylindrical target reservoir ($\phi = 19$ mm, $l = 58$ mm) was made from Ta and lined with a blanket of Al_2O_3 fibers also from Zircar Corporation and the HfO_2 target material was rolled into disks and separated from each other by a few mm to allow more efficient radiative cooling. The total target density was 4.6 g/cm², effectively stopping the ^2H beam within the first two HfO_2 disks, allowing a reserve of material to be available if some of the target were consumed during the run. Care was taken to shield the radial Al_2O_3 liner from direct beam bombardment while the beam was allowed to pass through ~ 2 mm Al_2O_3 disk shown to the right in Figure 4.

The target was bombarded with 2-4 μA of 45-MeV deuterons and the target heater shown in the figure was operated with 500 A of heating current, which corresponds to a target reservoir wall temperature of ~ 1600 C. Initially, the $^{17}\text{F}^-$ yield was low ($\sim 10^5$ ions/s) and then increased an order of magnitude within ten days of operation. The yield steadily increased with time achieving a maximum of 6×10^7 ions/s after ~ 50 days of operation. In total, the $^{17}\text{F}^-$ yield remained in the 10^7 ions/s range for about 35 days. Very few changes were made to the source operating parameters during this run: Target heater: 500A; KENIS transfer line: 240 A; Cs oven: 180C; KENIS cone potential: 260V; KENIS grid potential: 300V. After this run, ^{18}F was also produced from this apparatus by applying a beam of 85-MeV alpha particles. Approximately 10^7 ions/s of ^{18}F were produced during bombardment with 0.5 p μA . Details of this experiment will be addressed in a subsequent publication. To date, seven

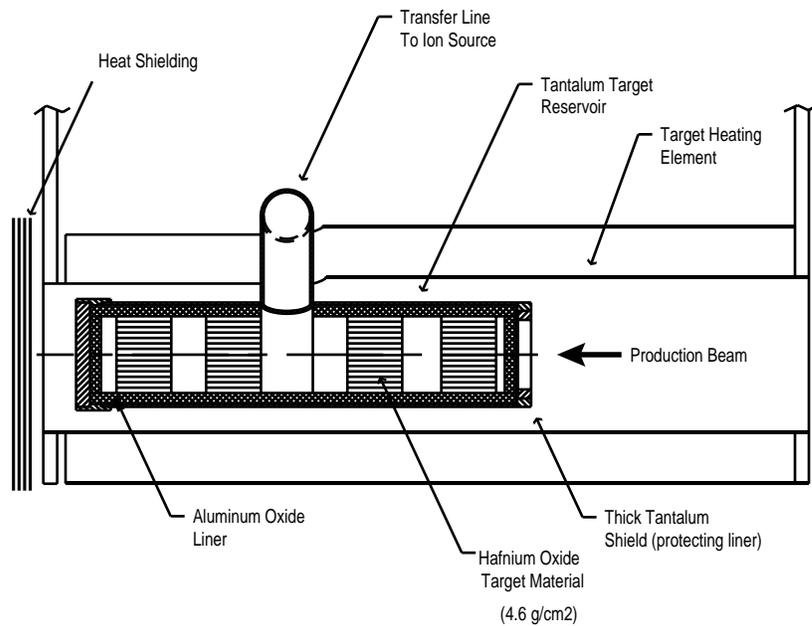


Fig. 4. Target configuration used for ^{17}F and ^{18}F production

nuclear physics experiments using beams of ^{17}F and ^{18}F produced using this apparatus have been performed over a beam energy range of 10-170 MeV, one of which has been published in reference 29.

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