Large radio-frequency gas catchers and the production of radioactive nuclear beams

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Abstract. Gas catchers provide a means to transform radioactive recoils from various production mechanisms into low-energy beams of good ion optical properties. Recent developments with large radio-frequency gas catchers have pushed back purity and space-charge limitations in this technology to the point that it can now be used reliably for producing radioactive beams intense enough for various secondary experiments to be possible. The basic technology available and the current demonstrated capabilities are presented in the following. A number of examples of such systems currently under commissioning/construction/design at ANL to produce beams from fusion-evaporation, fission, deep-inelastic and fragmentation reaction products will also be presented together with the specific challenges to each approach and the chosen solutions.

1. Introduction

The ability to make numerous measurements of interest to nuclear physics and astrophysics depends on the availability of radioactive beams with the proper properties. The energy of a beam as well as the intensity, degree of contamination, timing and optical properties often determine if an experiment is possible or not. This has been a severe limitation for low-energy measurements such as the study of direct reactions at astrophysical energies or single- or two-nucleon transfer studies at Coulomb barrier energy, or even at the very lowest energies such as work on rare isotopes in traps. The isotopes of interests are typically obtained through nuclear reactions that either have them created as recoil at an energy determined by the production reaction or makes them available at rest in a large production target. Both approaches have limitations: in the case of direct recoil extraction [1], the energy of the recoils is often not that best suited for the particular experiment and the energy spread from the reaction is not matched to the experimental requirements, for the ISOL type [2] in target production, many isotope species cannot be extracted efficiently using this method. The net result is that although through various reaction mechanisms a significant fraction of the isotopes stable against particle emission can be produced in some quantity, only a very small fraction has been amenable to precision studies at low energy.

These limitations were overcome somewhat by the IGISOL technique pioneered at Jyvaskyla [3] where a small gas volume is used to thermalize the recoils and extract them via gas flow. This approach is fast and universal, and has been highly successful, but it is however limited by the small stopping volume that can be rapidly evacuated. To efficiently stop faster recoils, a new approach with larger stopping volume, obtained either through ion guides surrounding
larger structures [4, 5, 6] or wrapped stopping paths [7], was proposed to accept with high efficiency fast reaction recoils and transform them into a low-energy beam that can be easily manipulated or reaccelerated to the energy of interest for the particular experiment. The device used to collapse the phase-space of the beam is called a gas catcher, a large volume of high-purity helium gas surrounded by a guiding structure where the ions lose their energy and are extracted rapidly into a beam forming region. Gas catchers [8] were developed for the collection of fusion-evaporation [9, 10, 11], fragmentation [12, 13, 14] and fission products [15]. And although devices with both high efficiency and fast extraction times were built, limitations due to space charge buildup when too large an ion current was injected in the device limited the application of gas catchers for radioactive beam facilities where high-intensity operation is required. Recent developments have however allowed one to raise the operation limits of such devices by many orders of magnitude so that they can now be contemplated as main components of next generation radioactive beam facilities. The main components of such systems and the developments that have led to the high intensity operation breakthrough and the numerous applications being developed for these devices are presented in the following.

2. Basic physics behind gas catcher operation
In its simplest form, a gas catcher is a stopping chamber in which radioactive recoils lose their kinetic energy in a gas while remaining charged so that they can be manipulated by electromagnetic forces to be rapidly extracted from the gas and transformed in a cool ion beam. The gas catcher is an extension of the IGISOL system [3] where the extraction of the ions from the stopping chamber is performed by gas flow alone. Limitations in the gas flow that can effectively be extracted from a nozzle limit the maximum stopping volume from which ions can be rapidly extracted in this way to volumes of a few to a few tens of cm$^3$. The small stopping volume severely limits the fraction of recoils that can be stopped and hence extracted. The gas catcher overcomes this limitation by using electromagnetic fields (see figure 1) to act on the radioactive ions so that they can be extracted much faster than the evacuation time of the chamber. A well designed device can maintain tens of millisecond ion extraction time although the gas evacuation time is tens of minutes.

Figure 1. Schematic view of a gas catcher showing the large volume filled with helium gas used to stop the radioactive recoils, the DC electric field pushing the slow ionized recoils towards the extraction nozzle (right side of figure) and the RF wall and cone (also shown in more detail on top of the figure) used to focus the ions towards the nozzle. The left side of the gas catcher drawing shows a port to insert a fission source. In on-line experiments, that port and the plate on which it is located is replaced by a large area thin metal window to let the recoil ions in. The whole assembly is built to UHV standards to maintain the helium gas purity.
For this approach to work, the fast recoils must keep some charge when they stop otherwise they will not be affected by the electromagnetic fields. This is accomplished by ensuring that the first ionization potential of the gas is higher then that of the recoils so that it will become energetically forbidden for the ions to neutralize as they come to rest. The highest first ionization potentials are found in the noble gases with Helium having the highest of all at 24.6 eV. This, and the fact that Helium can be easily purified to limit the number of contaminants on which the ions could also neutralize, make it the ideal gas for this application.

The size of the stopping volume is determined by the energy, energy straggling and divergence of the recoils that need to be stopped. The product of the length and operating pressure of the stopping chamber must correspond to enough gas atoms to absorb the straggling range of the recoils. This can range from a few tens of mbar-cm for superheavy elements produced by cold fusion reactions to roughly $10^5$ mbar-cm for radioactive recoils produced by high-energy fragmentation. Note that in all cases the gas catcher only needs to be able to handle the range straggling of the radioactive ions, not the full range since the higher energy part can be absorbed in a solid degrader before the gas catcher. Extraction for the gas catcher is performed by pushing the radioactive ions quickly towards an extraction nozzle using electromagnetic fields and letting the gas flow take over once the ions are within a few mm from the nozzle where the gas velocity is highest. The extraction nozzle is typically a choked supersonic nozzle with the gas flowing at about 1000 m/s so that the extraction time is dominated by the drift time to the nozzle. The velocity of ions in gas in the presence of electric fields can be obtained from ion mobility data [16]. The velocity is directly proportional to the mobility of the ion and the electric field, and inversely proportional to the gas density and hence its pressure over its temperature:

$$v_d = K \cdot E \cdot \frac{P}{P_0} \cdot \frac{T_0}{T}$$

with $v_d$ the drift velocity in cm/sec, $K$ the ion mobility, $E$ the electric field, $P$ the pressure in mbar ($P_0 = 1000$ mbar) and $T$ the temperature in kelvins ($T_0 = 273$ K). Typical values for ion mobilities in Helium gas are around 18 cm$^2$/V·s so that typical median extraction times are of the order of 10 ns for a 50 cm long gas catcher operating at 100 mbar and a total voltage of 700 volts across.

Bringing ions stopped in a large volume to within a few mm of an extraction nozzle is however complicated by what DC electric fields can actually be created and by physical limitations on what field can be held in a gas. The damping of the ion motion in the gas implies that the ions follow the electric field lines and hence all field lines within the volume from which the ions need to be extracted must lead to a region of a few mm around the nozzle for the ions to get to that point. This implies a concentration of field lines at the nozzle and hence a stronger electric field gradient there. Electric fields in a gas are however limited by the possibility of electrical discharges that would ruin the operation of the gas catcher. The required voltage over a given distance for these discharges to occur has been studied in details and is given by the Paschen curve [17]. It limits the field in the extraction region which then limits the field in the rest of the gas catcher to much lower values which limits the extraction time. Diffusion and space charge are also issues since they spread the ions around the field lines and move them to field lines that do not end up close enough to the nozzle. To obtain the fastest extraction times and overcome diffusion and space charge, it is therefore necessary to have another force than the DC electric field to provide the focusing towards the extraction nozzle. The most successful approach has been the use of RF electric fields, on top of the DC field, whose effect can be shaped much more freely than that of DC fields. This has been successfully implemented using RF walls, also called RF cones (see figure 2) or RF carpets. These structures are composed of closely spaced electrodes on which alternating electric potential of opposing phases are applied to the even and odd plates (see highlight in figure 1). This creates a strong RF field close to the structure.
that falls of quickly with distance from the structure with a fall off rate related to the structure electrode spacing. In an inhomogeneous RF field, the average force exerted on charged particles by the RF gradient is given by

$$F_{\text{vac}} = -\frac{e^2}{2m\omega^2} \int |E| dE$$  \hspace{1cm} (2)

with $e$ the electronic charge, $m$ the mass of the ion, $\omega$ the angular frequency of the RF field and $E$ its amplitude. The force pushes along the RF gradient and charged particles are therefore repelled by strong RF field regions. Hence, the ions are repelled from the wall once they get closer than a distance of about a few electrode spacing from the structure. This approach has been implemented using either stacks of thin electrodes [8] or circuit boards with thin electrodes traced on them [12]. The former allows cones of UHV compatible material to be formed providing the optimum field configuration for focusing while the ions are pushed forward by a DC field, while the latter is technically simpler to realize and can practically reach smaller electrode pitch but is so far limited to flat geometries where they must fight off directly the DC field and introduce material with poorer vacuum properties in the gas catcher. High intensity operation has only been obtained with the former this far and we will concentrate on this approach in the following sections. The repelling force provided by the RF walls is weakened by the presence of the gas damping which reduces the amplitude of the ion’s oscillations in the inhomogeneous RF field so that the force is reduced according to

$$F_{\text{damp}} = -\frac{e^2}{2m\omega^2} \int |E| dE \cdot \frac{1}{1 + \left(\frac{eKm\omega}{\kappa}\right)^2} = F_{\text{vac}} \cdot \frac{1}{1 + \left(\frac{eKm\omega}{\kappa}\right)^2}.$$  \hspace{1cm} (3)

In the presence of gas, the repelling force therefore scales roughly as the RF amplitude squared over the pressure squared. Since the RF amplitude is also limited to voltages below the Paschen curve, this favors operation of RF walls at lower pressure. Typically, the RF walls become too weak at pressures above about 200 mbar to overcome the other forces present in the gas catcher. This is however not a serious limitation since, as will be seen below, for most applications gas catchers of practical dimensions can provide the required stopping power to absorb the range straggling of the radioactive recoils.

**Figure 2.** RF cone for a 25 cm inner diameter gas catcher. The cone is made of about 280 independent electrodes of 0.4 mm thickness separated by 0.5 mm. The even plates have the RF offset by 180 degrees with respect to the odd plates. In addition, a DC gradient is applied along the structure to drag the recoil ions forward.

3. High intensity operation
Gas catchers has been implemented successfully for physics measurements in a number of applications with radioactive recoils produced by fusion-evaporation [18, 19], fragmentation [20]
or fission reactions [21]. In all cases however, they were limited to fairly low intensity of extracted recoils, compatible with direct measurements but not with the formation of radioactive beams for acceleration. In this latter case, most applications will have to deal with important losses in the acceleration process (charge breeding, bunching and transport) and must provide enough particles after acceleration for nuclear reactions with cross-sections typically of mbarn size to be observed. This implies operation with recoil rates inside the gas catcher of $10^6$ to $10^9$ ions per second or higher, far above what was used in the applications mentioned above.

Figure 3. On the left is shown the result of a self-consistent calculation of the potential buildup by 60 millions ions entering a small gas catcher and depositing energy as they stop within the gas catcher (the outline of the gas catcher is shown as a dashed red line). The potential buildup is the result of the excess of positive charge ions left behind in the gas as the electrons are removed much faster by the electric fields. The potential build up is repelling the ions from the center of the gas catcher towards the side. These ions can be recovered and transported to the nozzle by adding an RF structure on the walls. Shown on the right is the RF body of a 10 cm inner diameter gas catcher with its UHV-compatible RF feeding circuit.

Recoils entering the gas catcher lose energy by ionizing the helium gas, creating electron/Helium-ion pairs. The electrons have an ion mobility inside the gas about 3 orders of magnitude larger than positive ions [16] and therefore leave the ionization region very quickly, leaving behind the positive charge. This positive charge excess creates a repelling potential for the stopped recoils that hampers their progress towards the nozzle and in the most intense cases totally overcoming the applied DC fields. The potential created by such a space-charge accumulation is illustrated in figure 3. The requirement for high intensity operation for facilities like CARIBU [15] and RIA/FRIB [22] generated an intense R&D effort to overcome these limitations. This led to an improved design for high-intensity gas catchers that was demonstrated in high-intensity tests at ANL and is now in routine operation at the CARIBU facility. Three basic features yield tolerance to high space charge. First, the DC electric field gradient has to be kept as high as possible. It determines how quickly the space charge is removed and the size of the electric field the space charge must build up to disrupt ion extraction. The intensity limit therefore scales as the electric field squared. The second feature is addition of RF focusing over the full body of the gas catcher. The RF cone or RF carpet at the extraction end of the gas catcher recovers ions that are pushed away from the nozzle region by space charge as it starts building up but when it gets too large many ions do not even get to the RF cone. Adding an RF structure over the full body of the gas catcher (see figure 3) allows one to recover even ions that are pushed against the side wall very far from the nozzle and transports them to the cone and then the nozzle (see left side of figure 4). This greatly increases the capacity of the gas catcher to operate at high intensity. The final feature that is essential at the highest intensities
is maintaining the highest gas purity so that as little as possible of the ionized helium charge exchanges to impurities and then run the RF structure in a way that it has no focusing effect at all on helium ions so that those neutralize on the walls as quickly as possible. Otherwise, the total number of ions directed towards the nozzle will overwhelm the extraction region and lead to important losses. This approach has been tested using an especially designed beamline at ANL [23] to carry, using a large superconducting solenoid, all reaction products in a fusion evaporation reaction to a large intensity gas catcher equipped with all features mentioned above. As shown on the right of figure 4, the device maintained high efficiency (~ 35%) at well above $10^8$ recoil ions per second entering the gas catcher. This level of performance now enables operation as a facility component for radioactive beams. It forms the basis for the CARIBU gas catcher which was designed for final operation with a 1 Ci $^{252}$Cf source yielding $10^9$ fission fragments per second and about $10^{10}$ alpha particles per second entering the gas catcher and currently operates successfully with a 100 mCi source as operational experience is being gained with the facility. This is the first operation of a gas catcher as an accelerator facility component and opens the way for a number of other applications, some of which will be described in the next section.

**Figure 4.** On the left is shown a schematic representation of the areas of the gas catcher from where radioactive recoils can be extracted when operating at high intensity. With only DC systems, only the region closest to the cone is accessible, the ions in the other regions being moved to trajectories not leading to the extraction nozzle by the space charge. The addition of an RF cone/carpet and an RF body allow ions from a larger and larger portion of the total volume to be recovered and channelled to the nozzle via the RF wall structures. Results from online tests performed at ANL with a gas catcher using RF over all surfaces showing the improved performance that is achieved when compared to systems with no or limited RF refocusing.

### 4. New applications for high-intensity radio-frequency gas catcher at accelerator facilities

The demonstration of reliable gas catcher operation at high intensity at CARIBU shows that this technology is now mature enough to be integrated into high-intensity radioactive ion beam facilities. Radioactive ions can be produced by a number of means, using either fusion-evaporation reactions, fragmentation reactions, fission and deep-inelastic reactions, and each has advantages in specific regions of the chart of nuclei. Each is also amenable to production of low-energy beams using the gas catcher approach. The gas catchers best for each approach are different in the exact geometry of the stopping region, but using the basic guidelines given
above, a device best matched to each application can be built. The main parameters to consider are the range straggling and transverse size of the radioactive recoil beams, the total particle rate and energy deposition expected in the particular application, and finally the mass range of the recoils of interest. Five applications of radio-frequency gas catchers being developed at ANL will be briefly presented here, all of which are either currently under construction or in operation. This sample does not cover all facilities presently in preparation but is meant to be representative of the possibilities that the technology opens up.

The first facility in operation using a high-intensity radio-frequency gas catcher for radioactive ion beam production is the CARIBU facility mentioned above. It is meant to convert fission products from a 1 Ci $^{252}$Cf source into a low-energy beam for experiments at low energy or at Coulomb barrier energy after post-acceleration. Fission is an effective means of producing neutron-rich isotopes but has a number of severe drawbacks. The recoils are emitted in $4\pi$ solid angle, are difficult to separate, and the source emits also a large number of neutrons which makes it a severe radiation issue. At CARIBU, these difficulties were overcome by using a large 50 cm inner diameter, 1 meter long, gas catcher so that most recoils emitted in $2\pi$ solid angle are stopped in the gas. The RF cone matching to the 50 cm diameter body is the largest that has been built so far and the full system contains over 1000 independent RF electrodes. The gas catcher is located in the center of 12000 lbs of shielding to protect users. The radioactive ions are extracted through an RFQ cooler which penetrates out of the shielding. The heat produced by the RF in the gas catcher must be removed from inside the shielding. This is done with a recirculating cooling circuit that is used both to cool the gas catcher in normal operation and heat the gas catcher after source installation. The extraction RFQ leads to a 50 kV acceleration section that forms a cocktail beam containing all radioactive species stopped in the gas. Very limited selection is available from the stopping range in the gas and the main recoil selection is performed by passing the cocktail beam through a compact isobar separator located on the same high-voltage platform. The device is in operation, with all elements remotely controlled, on a high-voltage platform located in a new building equipped with the necessary safety features to monitor and control operation and effluents coming out of the pumping system. It will provide the ATLAS user’s community with neutron-rich radioactive beams for research in astrophysics, nuclear structure and applications.

A large number of neutron-rich nuclei, many close to stability, are not produced in fission and cannot be produced by fusion-evaporation and are only weakly accessible via fragmentation. Deep-inelastic reactions offer access to these regions and are also considered as a possible mechanism to access more neutron-rich superheavy isotopes [24]. Deep-inelastic reactions involve collision close to the grazing angle and the reaction products are therefore characterized by a very wide reaction cone opening and a large energy spread. It is difficult to obtain isotope separation and a significant collection efficiency. The preferred approach is therefore to use a large solenoid to capture as many of the recoils as possible, collect them in a large gas catcher and then perform separation after the ions have been cooled into a good optical properties beam. This implies ion numbers of $10^8$ ions per second or higher entering the gas catcher. This approach is being implemented in a component of the radioactive beam upgrade at the Texas A&M cyclotron facility. High-intensity heavy-ion beams at 20-30 MeV/u will be used to produce radioactive recoils by deep-inelastic reactions that will be focused by the BIGSOL superconducting solenoid into a 25 cm inside diameter, 1.2 m long, gas catcher built using the high-intensity approach developed at ANL. The extracted beams will be available for charge breeding in an ECR ion source for reacceleration through the K500 superconducting cyclotron to reach energies up to 50 MeV/u. The cyclotron acts as an effective mass separator for the reaccelerated beams.

A strong incentive behind the development of the gas catcher concept comes from next generation radioactive beams facilities such as RIA/FRIB that will use the gas catcher to provide a wide range of isotopes not available by the ISOL approach at low and Coulomb barrier energy.
Fragmentation is a fairly universal production technique that, with the primary beam intensities that will be available with the new facilities, should provide the widest range of radioactive recoils. The energies of the reaction products are very high (50-1000 MeV/u) which is both an advantage and an inconvenient. The large range allows fast isotope separation by physical means so that the recoils of interest can be isolated from the bulk of the production and transported as a beam to the gas catcher. The range straggling is also large which increases the volume of gas required to stop the recoils although this can be reduced significantly by achromatization [25] of the recoils before injection into the gas catcher. For a 200 MeV/u facility like the proposed FRIB, a stopping volume of the order of 0.3 atmosphere-meter of helium gas is sufficient to cover the range straggling of all but the very lightest isotopes. A high-intensity gas catcher of 25 cm inner diameter and 1.5 meter length that will operate at pressure from 50 to 200 mbar is under construction at ANL for this application. It will be tested initially at the ReA12 facility that will be a precursor to FRIB at MSU. Combined selection from the fragment separator and the stopping range will yield a fairly pure recoil beam which will then be charge bred in an EBIS source before further acceleration for experiments.

The three approaches presented above will combine to yield low-energy beams of essentially all radioactive isotopes discovered so far and a large number of as yet undiscovered ones. The one major production mechanism not mentioned so far is fusion evaporation reactions which yields access not only to most neutron-deficient isotopes but also to the superheavy elements which are not accessible by fragmentation or most other mechanisms. Although the superheavy isotopes cannot be produced in large amount, the yield by fusion-evaporation is sufficient for their properties to be studied in detail if they were available at low energy. To take advantage of this possibility, gas catchers are proposed behind the gas-filled separators at Dubna and LBNL, and behind the high-intensity separator S3 at GANIL. The gas catchers will transform the rare recoils into a low-energy beam for direct mass identification of the new elements discovered by hot fusion and the general study of the atomic, chemical and related properties of the longest-lived of these isotopes. The issues for the gas catcher in this application are connected to the high efficiency and reliability required for this application and the extremely low energy of the recoils. The beam spots for the superheavy recoils at the back of gas-filled separators are large due to angular straggling in the gas and remaining momentum dispersion, typically 10 cm wide in the horizontal dispersive direction and a bit smaller in the vertical direction. A 14 cm inner diameter by 30 cm long gas catcher operating at 30 to 150 mbar is most suitable for this application. A thin UHV-compatible all-metal entrance window, supported by a high-transmission grid, separates the gas of the gas-filled separator from the ultra-pure helium of the gas catcher. The rare recoil ions are extracted from the gas catcher in a few ms through RFQs to form a low-energy beam that is mass separated and deposited at well defined location for further identification or study. All but the shortest-lived superheavy elements will be available in this form. A total efficiency of 30-40% is expected in this process but it will yield information not accessible with the current setups and the efficiency loss might be partly compensated by improved detection efficiency around the smaller collection points. The gas catcher behind the S3 separator is planned to be used also for the study of neutron-deficient isotopes such as $^{100}$Sn also produced by fusion-evaporation reactions and will therefore be a larger device similar to that proposed for deep-inelastic reactions to cover the range straggling in these reactions.

5. Conclusion
Gas catcher technology has made significant progress in the last few years. With the demonstration of the capability to operate at high intensity, it has opened up a wide range of possibilities for the production of low-energy and reaccelerated beams of rare isotopes at intensities sufficient for secondary reactions. These are capabilities that were so far available only for a very limited set of beams at these energies. Gas catchers can be matched to a wide
variety of reaction mechanisms, each with unique advantages in specific regions of the chart of nuclei. The first facility using a gas catcher to produce reaccelerated radioactive beams, the CARIBU facility, is now in operation. A number of new facilities aiming to take advantage of these capabilities are now under construction. This will greatly increase the radioactive beam community access to these rare beams in an energy regime critical for many studies in nuclear structure and astrophysics.

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