Development of a Cryogenic Gas Cell Purification System

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Abstract

Multi-nucleon transfer reactions in heavy actinide target-projectile combinations may represent an important breakthrough for producing neutron-rich superheavy nuclei. Therefore, designing and constructing a proof-of-principle spectrometer for neutron-rich superheavy nuclei research applicable to multi-nucleon transfer reactions can not only lay the technical, methodological, and instrumental foundation for exploring the island of stability of superheavy nuclei, but also holds significant importance for studying the mechanisms of multi-nucleon transfer reactions. The gas cell in the proof-of-principle spectrometer for neutron-rich superheavy nuclei research requires a continuous supply of high-purity helium gas to stop the multi-nucleon transfer reaction products and extract the energy-degraded ions to subsequent experimental apparatus. This paper primarily introduces the newly developed cryogenic purification system for the gas cell of the proof-of-principle spectrometer for neutron-rich superheavy nuclei research, which mainly provides recyclable high-purity helium gas for the spectrometer. Experimental measurements demonstrate that the cryogenic purification system can purify 99% helium gas to over 99.999%, and when combined with a chemical purification unit, the purified helium meets the requirements of the gas cell for high-purity helium.

Full Text

Development of a Cryogenic Purification System for Gas Cells

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Abstract

Multinucleon transfer reactions involving heavy actinide projectile-target combinations may represent a crucial breakthrough for producing neutron-rich superheavy nuclei. Therefore, designing and constructing a prototype spectrometer for neutron-rich superheavy nucleus research suitable for multinucleon transfer reactions can not only establish the technical, methodological, and instrumental foundation for exploring the island of stability of superheavy nuclei, but also holds significant importance for studying the mechanism of multinucleon transfer reactions. The gas cell in the prototype spectrometer for neutron-rich superheavy nucleus research requires a continuous supply of high-purity helium gas to stop the multinucleon transfer reaction products and extract the energy-degraded ions to subsequent experimental apparatus. This paper primarily introduces the newly developed cryogenic purification system for the gas cell of the prototype spectrometer for neutron-rich superheavy nucleus research, which mainly provides recyclable high-purity helium for the spectrometer. Experimental measurements demonstrate that the cryogenic purification system can purify 99% helium gas to over 99.999%; when combined with a chemical purification unit, the purified helium meets the gas cell' s requirements for high-purity helium.

Keywords: multinucleon transfer reaction; gas cell; cryogenic purification system; high-purity helium

1. Introduction

Since the 1960s, the exploration of the superheavy nuclear stability island and the synthesis of new elements have become one of the most popular research topics in nuclear physics, and human investigation of the superheavy nuclear stability island has never ceased. Theoretical studies indicate that the island of stability for superheavy nuclei lies in the nuclear region centered around proton number 114 and neutron number 184, where atomic nuclei have relatively long lifetimes and greater stability [1-2]. Reaching the island of stability for superheavy nuclei can not only reveal the properties and structure of atomic nuclei under extremely strong Coulomb fields, but may also lead to the discovery of the largest nuclear magic numbers. Furthermore, synthesizing long-lived isotopes on the island of stability would facilitate studies of the physical and chemical properties of superheavy elements and unlock their enormous potential applications. Over the past decades, scientists have synthesized dozens of new superheavy nuclides using various nuclear reactions such as fusion-evaporation [3-4]. However, constrained by reaction mechanisms, all artificially synthesized superheavy nuclides to date are located on the neutron-deficient side of the nuclide chart. Experimentally synthesizing nuclides on the neutron-rich side of the island of stability remains a tremendous challenge.

With the development of high-intensity heavy-ion accelerator technology, multinucleon transfer reactions involving heavy actinide projectile-target combinations may represent a crucial breakthrough for producing neutron-rich superheavy nuclei [5]. Additionally, multinucleon transfer reactions can be used to synthesize neutron-deficient nuclides and neutron-rich nuclides with neutron number 126 that are relevant to the r-process [6-7]. However, multinucleon transfer reactions produce a wide variety of products with strongly anisotropic angular distributions and broad energy and charge-state distributions [8], making it difficult to separate and identify them using existing experimental methods and devices. Therefore, designing and constructing a spectrometer suitable for multinucleon transfer reactions for neutron-rich superheavy nucleus research is essential.

In nuclear physics, gas cells are widely used to stop nuclear reaction products. Early research primarily combined gas cells with online isotope separation devices, where nuclear reactions occurred inside the gas cell and reaction products were extracted from it. This technology has been applied in Jyväskylä [9], Leuven [10], and Mainz [11]. New-generation gas cells are mainly combined with In-Flight isotope separation devices, where nuclear reactions occur outside the gas cell, which then stops the resulting ion beams and extracts them. Research teams at ANL [12], NSCL-MSU [13], and RIKEN [14] have all developed and tested new-generation gas cells. China's major scientific infrastructure project, the High-Intensity Heavy-Ion Accelerator Facility (HIAF), will officially commence operation around 2025 [15], providing unprecedented experimental conditions for exploring the island of stability for superheavy nuclei using multinucleon transfer reactions and representing a major opportunity. Consequently,

developing a stable and reliable prototype spectrometer for neutron-rich superheavy nucleus research is imperative. Researchers at the Institute of Modern Physics are currently designing and constructing such a spectrometer suitable for multinucleon transfer reactions. The gas cell is the most critical component of the spectrometer, functioning to stop multinucleon transfer reaction products and extract the energy-degraded ions to subsequent experimental apparatus. After passing through the entrance window, ions continuously collide with gas molecules or atoms inside the gas cell, losing energy and exchanging charge. Therefore, the type and purity of the stopping medium in the gas cell directly affect the survival rate of ions in the gas cell [16-17].

Helium is a colorless, odorless inert gas that is the most difficult gas to liquefy in nature, with extremely stable chemical properties that generally prevent it from combining with any element under normal conditions, making it an excellent stopping medium. Furthermore, to accelerate ion migration speed in the gas cell, an electric field is typically established, requiring the stopped reaction products to maintain a certain charge state. Helium has the highest first ionization energy of all elements [18], ensuring that stopped ions maintain their charge state. Based on these excellent characteristics, the newly designed gas cell selects helium as the stopping medium. Helium purity is a critical factor affecting ion survival rates; if impurities are present, they can neutralize ions into atoms, reducing ion survival rates and affecting the separation and identification of subsequent reaction products. During experiments, the gas cell requires a continuous supply of high-purity helium at 99.99999% purity, resulting in enormous helium consumption. However, helium is extremely rare on Earth and unevenly distributed, and China is a helium-scarce country where high-purity helium is very expensive [19-20]. Therefore, to achieve continuous supply of high-purity helium while reducing consumption, ensuring experimental reliability while lowering costs, we have designed and constructed a helium cryogenic purification and circulation system for the prototype spectrometer for neutron-rich superheavy nucleus research.

2. Basic Principles

Helium can be purified through physical or chemical methods, with different purification approaches adopted according to different purity requirements. Current mainstream helium purification methods include low-temperature condensation, membrane separation, and chemical purification [21-23]. Membrane separation demands high-quality membranes and provides relatively poor purification precision. Chemical purification offers higher precision but typically requires special reagents and filters with longer purification times. Cryogenic condensation purification utilizes condensation, solidification, and low-temperature adsorption to separate impurities. Table 1 shows the basic physical properties of impurity gases in helium. In practical applications, reducing helium temperature below the boiling and freezing points of impurity gases can liquefy or solidify water, oxygen, hydrogen, nitrogen, and other impurities for separation

and removal. Cryogenic purification is widely used due to its high efficiency and relatively simple technical approach [16].

To maximize ion survival rates, the gas cell demands extremely high helium purity. Direct chemical purification would require long purification times and low efficiency, unable to continuously provide sufficient high-purity helium for extended periods, thus failing to meet experimental requirements. While cryogenic purification offers higher efficiency and simpler implementation, the resulting helium purity is slightly lower. To meet experimental demands for high-purity helium, this purification system employs a combination of cryogenic and chemical purification. The cryogenic purification system purifies 99% helium to 99.999% and above, after which a chemical purifier further purifies the 99.999% helium to 99.99999%. This combined approach not only improves overall purification efficiency but also achieves stable supply and recycling of high-purity helium, meeting experimental requirements while significantly reducing costs. This paper focuses primarily on the cryogenic purification system.

3. System Design

The primary design objective of the purification system is to provide continuous high-purity helium for the gas cell while recycling used helium. Therefore, purification efficiency and helium purity are the key performance indicators of the entire system. The high-purity helium for the prototype spectrometer for neutron-rich superheavy nucleus research becomes contaminated by molecular pumps, the gas cell, and transfer pipelines, with its purity expected to drop to approximately 99%. The cryogenic purification system controls temperature to liquefy or solidify different gas impurities, thereby separating them from helium and improving purity. The complete cryogenic purification system consists of helium buffer tanks, a refrigeration unit, booster pumps, various valves, and multiple pipelines. Valve control primarily employs pneumatic methods, supplemented by needle valves, manual ball valves, pressure reducing valves, check valves, and safety valves. Additionally, flow meters are installed in the pipelines to regulate helium flow rates in different lines. Currently, the entire cryogenic purification system has been manufactured and assembled by Anhui Vacree Cryogenic Technology Co., Ltd. [24], with the overall system design shown in Figure 1 [Figure 1: see original paper].

The refrigeration unit plays a critical role in the entire cryogenic purification system. Figure 2 [Figure 2: see original paper] shows the internal structure of the refrigeration unit in the system. The refrigeration unit' s housing chamber is made of 304 stainless steel and contains multiple temperature sensors and three heaters. Temperature sensors record temperatures at various critical locations to facilitate separation of different gas impurities, while the three heaters are installed near the cold heads and recuperative heat exchangers to regulate temperature and ensure smooth regeneration. The refrigeration unit employs Vacree' s GM415 cryocooler [24] and Sumitomo' s RDK408S cryocooler [25] as cold sources to provide sufficient cooling capacity, with each cryocooler having

both first-stage and second-stage cold heads. Table 2 lists the cryocooler parameters. During actual assembly, the first-stage cold heads of both cryocoolers are coupled in parallel to the first-stage heat exchanger. The GM415' s second-stage cold head provides cooling for the second-stage heat exchanger, while the RDK408S' s second-stage cold head serves as a backup cold source for use during system failure or insufficient cooling capacity.

Cryocooler selection primarily considered the cooling capacity relative to actual requirements. The first-stage heat exchanger mainly reduces contaminated helium to a temperature where impurity gases fully liquefy. Since nitrogen solidifies at 63.1 K, the outlet temperature is set to 65 K to prevent freezing and blockage in the first-stage heat exchanger. This temperature setting not only optimizes helium purity but also ensures normal heat exchanger operation. The primary heat load on the first-stage cold head comes from cooling 99% helium and 1% impurity gases to 65 K, plus heat leakage from the cold box. Based on experience, cold box heat leakage is set at 5 W, with a rough estimate indicating the first-stage cold head requires 55 W of cooling capacity at 65 K. After most impurities are removed from helium at the first-stage heat exchanger, the second-stage heat exchanger provides extremely low temperatures to solidify remaining impurity gases.

To efficiently utilize the cryocoolers' cooling capacity, the refrigeration unit also incorporates a DEVOR [26] BX4TH first-stage plate recuperative heat exchanger and a second-stage negative pressure recuperative heat exchanger independently designed by Vacree [24]. The second-stage negative pressure recuperative heat exchanger is made of copper spiral tubing with fins vacuum-brazed sequentially along the spiral direction, with perforated and non-perforated fins arranged alternately to significantly enhance heat transfer capability on both shell and tube sides. The first-stage plate recuperative heat exchanger warms the purified cold helium, using contaminated helium entering the refrigeration unit as the heat source, thus also serving as a pre-cooler for contaminated helium. The second-stage negative pressure recuperative heat exchanger is installed near the second-stage cold head, with cold helium from the second-stage heat exchanger simultaneously pre-cooling helium entering the second-stage heat exchanger. Finally, the cryocoolers and heat exchangers are wrapped with aluminum-coated film, maintaining the refrigeration unit chamber vacuum at 10^{-2} Pa during operation to reduce radiation and convection heat leakage, providing thermal insulation and ensuring cryocooler and heat exchanger efficiency. The physical diagram in Figure 2 [Figure 2: see original paper] shows the cryocoolers and heat exchangers wrapped in aluminum-coated film. During actual testing, the second-stage heat exchanger temperature can reach approximately 10 K, fully meeting practical requirements.

The entire helium purification process includes six stages: stop, regeneration, purge, dry-cool, pre-cool, and purification. System pipelines include helium purification lines, helium purity detection lines, and safety vent lines. Figure 3 [Figure 3: see original paper] shows the complete helium purification process

flow for the cryogenic purification system, with each stage described below:

1. **Stop Stage**: The cryogenic purification system is initially in a stopped state, or pauses when the purifier reaches saturation. During this stage, all valves entering the refrigeration unit are closed, the cryocoolers are not operating, and only purge vent valves are open.
2. **Regeneration Stage**: Before purification begins, this stage clears blockages in pipelines caused by impurity solidification and low-temperature adsorption, allowing the purification system to operate again. This stage primarily uses heaters to warm refrigeration pipelines, converting condensed impurities on pipe walls back to gaseous state.
3. **Purge Stage**: Impurity gases are expelled. All valves entering the refrigeration unit are opened, using high-purity helium to blow impurity gases out through vent pipelines. After purging, the purge vent pipelines are closed.
4. **Dry-Cool Stage**: The system temperature is lowered to remove residual moisture from the purging process.
5. **Pre-Cool Stage**: Cryocoolers reduce the temperature of refrigeration pipelines and helium in the refrigeration unit below target operating temperatures, preparing for subsequent purification.
6. **Purification Stage**: The most critical stage of cryogenic purification. After pre-cooling, low-purity helium is introduced into the system for purification. Purified helium passes through purity detectors and flow controllers regulate its flow rate.

From the stop stage through the purification stage constitutes the complete workflow of the cryogenic purification system. During actual experiments, stop, regeneration, purge, dry-cool, and pre-cool are preparatory processes. When experiments begin, the cryogenic purification system continuously operates in the purification stage. Additionally, to avoid interrupting experiments due to cryogenic purification system downtime, high-purity helium at 99.99999% purity can be directly supplied to the gas cell when the purification system is paused.

4. Test Procedures and Results

Helium purity is a critical indicator for evaluating cryogenic purification system performance. Purity is typically determined by measuring impurity content including water (H_2O), nitrogen (N_2), and hydrocarbons (C H). Multi-component dew point meters employ different measurement techniques and sensors to simultaneously measure dew or frost points of water vapor, nitrogen, and hydrocarbons in helium. These devices combine infrared absorption, electrolytic sensors, or thermal conductivity to detect impurity content. Dew point meters determine water content through humidity sensors that detect moisture changes and calculate dew point temperature; lower dew point temperatures

indicate lower water content. For hydrocarbon detection, dew point meters use infrared absorption and thermal conductivity detection. By measuring different impurity components, helium purity can be accurately determined. This test used a multi-component dew point meter (MDM300) [27] to measure water, nitrogen, and hydrocarbon content to determine helium purity.

Regarding purification effectiveness, stability, and recoverability, we conducted the following tests and obtained preliminary results:

1. **Purification Effectiveness Test**: After completing preparatory work, 99% purity helium was introduced into the cryogenic purification system while measuring the purified helium purity. Table 3 records measured helium purity and impurity content at different times, while Figure 4 [Figure 4: see original paper] shows the purity variation trend over testing time. Results demonstrate that after single-stage cryogenic purification, 99% helium reached 99.99725% purity at the outlet. The relatively low initial purity was due to residual impurities in system pipelines. After three hours of operation, pipeline impurities were essentially cleared, achieving 99.99972% helium purity with excellent purification effectiveness that met expected targets.

2. **Stability Test**: To ensure long-term operation in practical applications, system stability testing is essential. This test conducted long-term stability testing by continuously feeding 99% helium into the system while measuring output purity. During the two-week test period, the purification system operated without blockage, demonstrating good purification effectiveness and stability. Additionally, the cryogenic purification system was reused multiple times over three months without observed performance degradation, indicating it can operate stably for extended periods in experiments, providing a solid foundation for subsequent low-cross-section nuclear physics experiments.

3. **Recoverability Test**: Since the cryogenic purification system freezes helium impurities in transfer pipelines to achieve purification, long-term operation may cause pipeline blockages. To understand actual recovery time when blockages occur, we conducted recoverability testing by intentionally feeding helium doped with air for extended periods to cause pipeline blockages and testing regeneration and purging performance. Test results show that when pipeline blockage occurs, regeneration and purging can be completed within 24 hours, effectively solving blockage problems with good system recoverability. Additionally, as mentioned above, the purification system includes a bypass line to the gas cell, allowing direct supply of 99.99999% high-purity helium to the gas cell during refrigeration unit recovery without affecting normal nuclear physics experiments.

This test used 99% helium; additional purification experiments and system performance tests will be conducted subsequently.

5. Conclusion

Exploring the island of stability for superheavy nuclei and synthesizing neutron-rich new nuclides using multinucleon transfer reactions represents an important future direction for nuclear physics development. The prototype spectrometer for neutron-rich superheavy nucleus research, as the instrumental foundation for such experiments, relies on the support of helium purification systems. Testing demonstrates that the cryogenic purification system developed by our group achieves the expected helium purification goals, with final purified helium purity reaching over 99.999%. When combined with the chemical purification unit, the purified helium meets the gas cell' s requirements for high-purity helium. Moreover, the system exhibits excellent stability and recoverability, capable of long-term stable operation and purification of used helium, significantly reducing experimental costs. Therefore, this cryogenic purification system establishes a reliable instrumental foundation for exploring the island of stability for superheavy nuclei using multinucleon transfer reactions.

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