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Xenon recovery from molybdenum-99 production

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Abstract

The U.S. Department of Energy Office of Nuclear Energy (DOE-NE) sponsors research and development on the recycle of used commercial nuclear fuel as an option for future nuclear fuel cycles that offers increased use of uranium and thorium resources and a possible reduction in the overall cost of nuclear waste management. The two alternatives, direct disposal of used fuel and fuel recycle, are broadly referred to as open and closed fuel cycles. One requirement of a closed fuel cycle is the safe management of radioactive off-gases, which includes ^{14}C, radioiodine and the noble gases, including radioxenon. The longest lived relevant radioxenon isotope is ^{127}Xe; with a half-life of just 36.35 days it is feasible to trap and hold the radioxenon to allow for decay to safe environmental levels. However, the very weak chemical bonds of noble gases, in this case xenon, make them difficult to trap, which led to an extensive DOE-NE study of noble gas adsorption on various molecular sieves as an alternative to costly cryogenics processes. Preliminary results indicate that xenon adsorption at near room temperature on molecular sieves, both synthetic and natural, may have both cost and efficiency advantages over cryogenic processes.

Xenon radioisotopes with short half-lives have value in the detection of underground nuclear explosions. However radioxenon released by medical isotope production facilities produces high backgrounds that complicate its use for nuclear explosion detection. Specifically, recovery of ^{99}Mo for medical applications from short-cooled irradiated ^{235}U targets results in the release of some radioxenon. In March 2015, by a unanimous vote of the five permanent members of the United Nations Security Council, commercial producers of ^{99}Mo were asked to decrease and, if possible, eliminate the emissions of radioactive xenon from their production processes. The use of materials currently being evaluated for Xe and Kr capture for use in UNF processing applications may provide a cost effective mechanism to achieve the United Nations Security Council goal.

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Keywords: xenon, medical isotopes, off-gas

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1. Background

The U.S. Department of Energy Office of Nuclear Energy (DOE-NE) sponsors research and development on the recycle of used commercial nuclear fuel as an option for future nuclear fuel cycles that offers increased use of uranium and thorium resources and a possible reduction in the overall cost of nuclear waste management. The two alternatives, direct disposal of used fuel and fuel recycle, are broadly referred to as open and closed fuel cycles. One requirement of a closed fuel cycle is the safe management of radioactive off-gases, which includes ^{14}C, radioiodine and the noble gases, including radioxenon.

The production and recovery of ^{99}Mo from short-cooled irradiated ^{235}U targets for medical applications also results in the release of volatile radionuclides. Owing to the very short time between irradiation and processing, much higher amounts of these volatile radionuclides with short half-lives are released into the medical isotope production (MIP) off-gas stream on a per unit mass of uranium processed basis. One of the most significant releases to the off-gas stream, in terms of activity, is radioxenon. The longest lived relevant radioxenon isotope is ^{127}Xe; with a half-life of just 36.35 days, it is feasible to trap and hold the radioxenon to allow for decay. Most, if not all, major ^{99}Mo processors have some level of control on the release of radioxenon. The trapping of radioxenon from MIP facilities may also help to support nuclear forensics efforts. Xenon radioisotopes with short half-lives have forensic value in the detection of underground nuclear explosions. However, radioxenon releases from medical isotope production facilities results in high backgrounds that complicate use of radioxenon for nuclear explosion detection.

In March 2015, by a unanimous vote of the five permanent members of the United Nations Security Council, commercial producers of ^{99}Mo were asked to decrease and, if possible, eliminate the emissions of radioactive xenon from their production processes. This was followed by the fifth Workshop on Signatures of Medical and Industrial Isotope Production (WOSMIP) held in Brussels, Belgium on May 12-14, 2015[1]. This workshop brought together representatives from the MIP and nuclear explosion monitoring communities with a focus on ways to mitigate the effects of radioxenon emissions from fission-based MIP on the verification efforts for the Comprehensive Nuclear-Test-Ban Treaty (CTBT). Discussions included new developments in International Monitoring System (IMS) noble gas network and sensors, updates from medical isotope producers on production processes and facilities, technologies used to measure radioxenon stack releases, research and development targeted at reducing radioxenon emissions, and methods for data sharing between the communities.The workshop had the largest representation of current and prospective isotope producers of any WOSMIP workshop to date. Fourteen current or prospective medical isotope producers attended the meeting and shared detailed information on current and future MIP. Several producers stated their intent to work toward achieving the voluntary radioxenon release goal of 5×10^9 Bq/day (0.135 Ci/day). One major producer stated that it is already meeting the goal in routine operations[1].

There are a number of similarities between the processing of used nuclear fuel (UNF) and MIP, and technology that is currently being developed to address the emission control needs of the UNF reprocessing may have direct application to the issues currently facing the MIP community. What follows is a brief analysis of the emission control challenges and requirements for both UNF processing and MIP in terms of source quantities and regulatory requirements.

2. Used Nuclear Fuel Processing

2.1. Used nuclear fuel processing, source terms and regulations governing emission

Atmospheric releases of radionuclides during the reprocessing of UNF must be controlled to levels that comply with air emissions regulations. Radionuclides that tend to form gaseous species that evolve into reprocessing facility off-gas systems are more challenging to efficiently control compared to radionuclides that tend to be contained in solid or liquid phases. Radionuclides that have been identified as “gaseous radionuclides” are noble gases (most notably isotopes of krypton and xenon): ^{3}H, ^{14}C, and ^{129}I. (Note that all radioactive xenon isotopes have very short half-lives relative to the time frames typically considered for UNF processing and will have decayed to the extent that they do not contribute to the dose.) For the purposes of this study, a relatively short cooling time (5 yr) for the UNF will be considered. Note that typically these values are reported in quantities per metric tonne initial heavy metal (tIHM) in the fuel, but for this analysis they are given per kg initial heavy metal (kgIHM) to allow direct