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Review

Capture of harmful radioactive contaminants from off-gas stream using porous solid sorbents for clean environment – A review

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HIGHLIGHTS

- Adsorption is an effective technique to remove contaminants from gas stream.
- Recent review on removal of harmful contaminants, I_2, Kr and Xe at reprocessing condition.
- Porous solid sorbents has several advantages for capture of volatile gases.
- I_2, Kr and Xe removal capacity of different sorbents are reported.
- MOFs are also effective sorbents for capture iodine and krypton.

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ABSTRACT

Nuclear energy production is growing rapidly worldwide to satisfy increasing energy demands. Reprocessing of used nuclear fuel (UNF) is expected to play an important role for sustainable development of nuclear energy by increasing the energy extracted from the fuel and reducing the generation of the high level waste (HLW). However, during the reprocessing of used nuclear fuel (UNF) gaseous radioactive nuclides including iodine, krypton, xenon, carbon, and tritium are released into the atmosphere through off-gas streams. The volatile iodine (^{129}I), and krypton (^{85}Kr) gases have long lived-isotopes; which have adverse effects on the environment as well as human health. Consequently, the capture of these two target radionuclides (species) is essential for the enhanced growth of nuclear energy. In this review we discuss several techniques for capture of volatile contaminants iodine, krypton, and xenon, focusing upon adsorption using solid sorbents, which has shown promising results for more than 70 years. Commonly used and recently developed sorbents are summarized in this article along with a short review of the results. Metal-organic-frameworks (MOFs), gaining favor in recent years as sorbents for the capture of off-gas contaminants are also discussed. Finally, some considerations of future trends and prospects for investigations of the capture of volatile radionuclides are presented.

Contents

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1. Introduction

The 21st century continues to see an unrelenting increase in energy demand worldwide due to rapid population and industrial growth, resulting in significant strain on the energy supply from the traditional fossil resources. Energy conservation and discovery of alternative primary energy resources are among the most pressing universal challenges to satisfy the current and future energy demands [1]. The U.S. Department of Energy (DOE) forecasts a nearly 28% increase in demand for electrical energy from 2012 to 2040 [2,3]. Currently, fossil fuels dominate the primary energy supply accounting for 67% (coal – 40%, natural gas – 22% and others) of electricity production [4]. However, the harmful effects of fossil fuels on the environment (and ultimately, human beings) have been well recognized [5,6], making it imperative to transition to a superior energy source that can satisfy these increasing demands in a way that does not exacerbate the environmental concerns.

Nuclear energy is one of these alternative sources for the production of electrical energy without excessive greenhouse gas emissions. It is safe, efficient, reliable and an inexpensive source of energy [7]. More than 400 nuclear reactors in 31 countries worldwide produce nearly 11.5% of global electricity [8]. It is attractive as a major source of electricity generation, as it does not suffer from the same technological and economic limitations as solar or wind energy. The nuclear electricity production is growing steadily to satisfy the worldwide energy demand and is projected to grow between 23 and 100% by 2030 [9]. However, nuclear energy is faced with the challenge of the management of radioactive waste produced at each stage of the nuclear fuel cycle, which can limit its growth. The radioactive wastes are classified as solid (high-level), liquid (low-level), and gases [10]. Gaseous radioactive waste presents an immediate threat to general population and the environment because of the ease of dispersal through the atmosphere.

Only a small fraction of radionuclide emission takes place during nuclear plant operation, primarily from minor leakage from the fuel rods [11]. The majority of radionuclide emissions take places during the aqueous reprocessing of used nuclear fuel (UNF) as it is chopped and dissolved in boiling nitric acid for subsequent extraction steps (PUREX process) [12]. Major gaseous radionuclides released into the atmosphere through off-gas include ^{129}I, ^{14}C, ^{3}H and noble gases (^{85}Kr and multiple Xe isotopes). ^{129}I and ^{85}Kr are a primary concern for the environmental pollution because of their long-lived isotopes that persist and accumulate in the environment [10]. ^{129}I is a highly mobile and volatile contaminant with a half-life of 1.52×10^7 year, and which bioaccumulates concentrating in the thyroid gland affecting biometabolism [13]. ^{131}I is another isotope of iodine, which has low half-life period 8.02 d. The long-lived isotope of Xe has a half-life of 36.4 d and will yield stable isotopes after typical UNF storage time of about 2 year. ^{85}Kr is a chemically inert radioisotope with a half-life of 10.7 y that poses more of a human health and environmental threat as it is continuously accumulated in the atmosphere [14].

Development of methods for the capture and long-term storage of radioactive gases is of crucial importance in order to manage their emissions, that are anticipated to increase significantly with the growth of nuclear energy. Several methods are available for removal of radioactive contaminants from off-gas streams, each with its own advantages and disadvantages. For more than 70 years, porous solid sorbents have been in the forefront of radioactive contaminant removal due to promising results and their advantages such as high removal efficiency, low maintenance cost, simple equipment design and operation over other techniques. Notably, in recent years, a new class of porous crystalline materials known as metal-organic frameworks (MOFs) has been shown to provide high capacities and wide customization options in the capture of iodine and noble gases.

In the present review article, we focus on several porous sorbents (traditional and newly developed), including an overview of promising MOFs for the capture of volatile radionuclides especially for iodine, krypton and xenon. Concerns and limitations of existing sorbents with operating conditions and their capacities for the capture of target species are also discussed.

2. Background

Fig. 1 shows the waste releases during various stages of the nuclear fuel cycle. Burger and Burns [15] reported that a small amount of elemental iodine is released from nuclear reactor due to fuel damage, but this is expected to have insignificant effect on overall iodine emissions from the fuel cycle during normal operations. Comparatively, large amounts of off-gas can be released during reactor accidents involving broad fuel ruptures. There are three main stages where off-gas released during reprocessing operations: (a) voloxidation off-gas (b) dissolver off-gas (DOG), and (c) vessel off-gas (VOG) [15,16].

The voloxidation process involves heating of the oxide fuel with oxygen at high temperature. The lattice structure of the uranium oxide fuel breaks down during the conversion of UO_2 to U_3O_8,

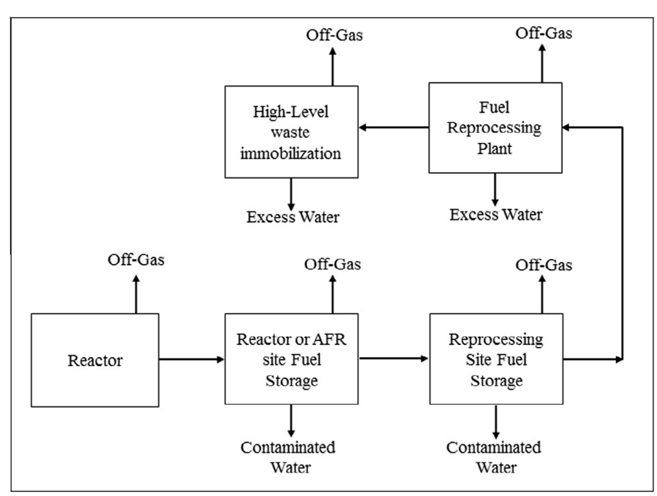

Fig. 1. Block diagram of off-gas generation stages at nuclear power station. Adopted from Ref. [15].