Particuology 14 (2014) 117–123

Contents lists available at ScienceDirect

Particuology

journal homepage: www.elsevier.com/locate/partic

Leakage of nuclear material powder from pressure container through a small orifice

Longbo Liu*, Yanmin Wu, Yongyang Su, Hanbing Tang, Zhihong Zhang, Ping Huang, Lili Zhang, Lixing Zhang

Northwest Institute of Nuclear Technology, Xi'an 710024, China

ARTICLE INFO

Article history:
Received 19 November 2012
Received in revised form 11 April 2013
Accepted 15 June 2013

Keywords:
Nuclear materials
Powder
Leakage
Risk assessment

ABSTRACT

Because of the radioactivity and toxic nature of nuclear materials, their containment within oxide matrices, encased in sealed containers, has been proposed as a suitable means for storage and transportation. However, container failures because of cracks or small orifices present a major leakage risk for nuclear materials, consequently posing a significant hazard to the environment and human beings. In this study, terbium oxide powder was used as a nuclear material representative to examine the leakage of nuclear material powder through orifices located at the base of a pressure container. The dependence of the orifice diameter, the powder layer thickness, and the internal pressure of the container on the leakage mechanism and amount was examined. A simplified model correlating the dependence of the above-mentioned parameters to determine the utmost leakage amount was also developed based on the present results. The leakage of the nuclear material powder was assessed by measuring its concentration using an optical particle counter. The diameter of the orifice determined the powder leakage mechanism, which in turn influenced the amount of leakage produced. Comparison studies showed that unlike the changes in the differential pressure, the volume of the container has little effect on the leakage amount. Under sufficiently high internal pressures, the oxide powder can be released as a fine aerosol. The work is not only crucial from the nuclear safety aspect, but is also beneficial for the safe application of powder and nanoparticles.

1. Introduction

Because of the high radioactivity and poisonous nature, nuclear materials are generally entrapped within a matrix host that is contained in sealed containers during storage, transport, and manufacture (IAEA, 1998). Containment of nuclear materials as an oxide powder form is highly recommended for long-term storage because of its superior chemical stability as opposed to that of a metallic or liquid form. Under specific conditions, the internal pressure of the containers may rise above the ambient pressure, therefore significantly increasing the risk of nuclear materials leakage. The airborne release fractions and the respirable fractions of released aerosols from major accidents, such as spilling, fire, and explosion, are crucial for risk assessment and have been summarized (DOE, 1994). However, minor accidents that could potentially escalate, thereby causing unexpected risks, also deserve complete consideration. For instance, the presence of cracks or orifices developed in the containers during storage could lead to the release of nuclear particles in the atmosphere, which could consequently amount to a significant hazard.

Powder leakage through orifices under pressure difference is similar to the discharge of particles from silos, which has been extensively investigated in the field of powder technology (Ahn, Başaranoğlu, Yılmaz, Buğutekin, & Gül, 2008; Nedderman, 1992). Three release mechanisms have been proposed: (1) no flow (arching); (2) funnel flow; and (3) mass flow, as depicted in Fig. 1. The type of release mechanism is governed by the diameter of the orifice and powder characteristics. The hole developed in the bulk powder in the case of a funnel-type flow is called a rat-hole. This effect has been studied in powder mechanics (Johanson, 2004).

Although significant progress has been made in determining the release flow mechanisms of powders from silos, the following differences in our system are likely to influence the resulting release mechanism that would consequently affect the amount of leakage produced. First, pressure difference rather than gravity drives the flow of the powder. Second, powder feeding is unavailable; hence no mass flow is expected to occur. Finally, the powder under

* Corresponding author at: P.O. Box 69-14, Xi'an 710024, China. Tel.: +86 29 84767720; fax: +86 29 83366333.
E-mail address: lomboliu@live.cn (L. Liu).

1674-2001/$ – see front matter © 2013 Chinese Society of Particuology and Institute of Process Engineering, Chinese Academy of Sciences. Published by Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.partic.2013.06.004

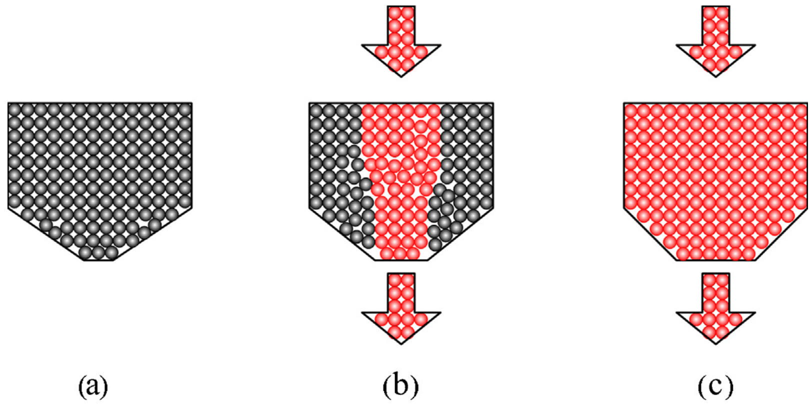

Fig. 1. Powder discharge mechanisms (from silos): (a) arching (no flow), (b) funnel flow, and (c) mass flow. The red balls represent mobile powder whereas the black balls represent stationary powder.

investigation is more cohesive and smaller orifices are being examined (See Section 3.1). Because of these differences, experimental studies, rather than theoretical extrapolations, are essential to yield better estimates of the amount of powder leakage.

Sutter et al. (1980) and Sutter, Johnston, Owzarski, Mishima, and Schwendiman (1981) simulated the leakage of plutonium dioxide powder from a damaged shipping container (~3.3 L). Depleted uranium oxide powder was used as a substitute to study the effect of the location and dimensions (diameter and length) of the orifices on the leakage amount. The authors also proposed a formula to estimate the leakage amount of the powder based on the container pressure and the orifice area. The extent of leakage was greater at orifices located below the powder layer than at orifices located above the oxide powder layer.

In the present study, the method as proposed by Sutter et al. (1980, 1981) was adapted to our system to analyze the dynamical characteristics of powder leakage and estimate the resulting leakage amount. Terbium oxide was selected as a nuclear material representative. This study is essential to understanding leakage mechanism.

2. Methods

The influence of the following parameters on the leakage amount is examined:

- initial pressure and volume of the container;
- orifice diameter;
- powder layer thickness.

For the purpose of this study, smaller-sized containers (that are smaller than the ones used in real-life practice) were employed in all experiments. However, it is know that a rapid decline in pressure will occur during leakage from a small container. In comparison, the rate of pressure drop in a much larger container is considerably reduced. To this effect, in our studies, gas was continuously fed into the small container during the leakage to avoid a fast decline in pressure, thereby mimicking the behavior in real-life large containers. In the first study (constant-volume), the volume of the container remained constant by releasing the gas and the change in pressure was monitored. In the second study (constant-pressure), the pressure was maintained by continuously feeding gas into the container during leakage to assess the effect of container volume on the leakage.

3. Experimental

3.1. Powder material

Terbium oxide (Tb_4O_7) powder (Shanghai Yuelong Rare Earth New Materials Co., Ltd., Shanghai, China) was used because of its comparable properties to transuranium elements (Kim, Joung, Lee, Oh, Koo & Heimgartner, 2008; Meyer, Newton, Cronenberg, & Loomis, 1994) and its relatively low ambient background. The density, bulk density, and the angle of repose of Tb_4O_7 are 4480 kg/m^3, 1930 kg/m^3, and 46.7°, respectively. The particle size of Tb_4O_7, as measured on a Mastersizer 2000 (Malvern Instruments Ltd., Malvern, Worcestershire, UK), is <10 μm, which is similar to the size of plutonium dioxide (Sutter et al., 1980). Based on its angle of repose (>40°) and particle size (<10 μm) characteristics, Tb_4O_7 powder is categorized as a cohesive material (Geldart, Abdullah, Hassanpour, Nwoke, & Wouters, 2006; Tykhoniuk, Tomas, Luding, Kappl, Heim, & Butt, 2005).

3.2. Constant-volume studies

The experimental set-up employed for this study, as illustrated in Fig. 2, consisted of a container with the following dimensions—diameter: 100 mm; height: 100 mm. The opening diameter at the base of the container was 8 mm. A stainless steel slab (thickness: 0.1–1.0 mm) with an orifice (diameter: 0.3–1.0 mm) was located at the opening. The discharged particles were collected on a polypropylene fibrous filter sampler that was connected at the end of the leakage tube.

The studies were conducted as such: the leakage valve was initially closed while nitrogen was fed into the container until a specified pressure. The gas inlet valve was then closed and the leakage valve was quickly opened. The change in pressure, as measured by a pressure transducer (AR3000, Zhejiang Supcon Instrument Co., Ltd., Hangzhou, China), was recorded every 1 s until the pressure dropped to the ambient pressure.

The procedure was repeated with the oxide powder loaded in the container. The discharged powder was collected by the filter and weighed. The filter was incinerated at 600 °C for 4 h. Any residue on the filter was then dissolved in hydrochloric acid solution and the mass of terbium in the solution was determined by inductively coupled plasma-optical emission spectroscopy (OPTIMA 3000, PerkinElmer Inc., USA).