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K. R. Givens[a]
[a] Chemical Technology Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee

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REMEDIATION OF BOROSILICATE RASCHIG RINGS

K. R. Givens

Chemical Technology Division
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37831-6046

ABSTRACT

A remediation methodology for borosilicate Raschig rings contaminated with radioactivity at a Department of Energy (DOE) Nuclear Facility is demonstrated in which > 99% of the residual radioactive contamination was successfully removed with a combination of nitric acid and ultrasonic agitation. The results obtained from this methodology are directly applicable to other radioactively contaminated Raschig ring waste streams. However, the universal application of the developed methodology is limited to similar historical processing applications of the borosilicate Raschig rings.

INTRODUCTION

Throughout the United States, borosilicate Raschig rings were commonly used for nuclear criticality control at DOE Nuclear Facilities between the period of 1950 to 1970 (1). The boron impregnated in the Raschig rings is used for absorption of neutrons released from fissile materials in solution; while, the Raschig rings are used for the resultant high surface area (2). Guidelines still exist for the use of borosilicate Raschig rings for nuclear criticality control; however, safe design features have become the preferred mechanism for nuclear criticality control of fissile solution storage and processes (3). The criteria for Raschig rings to completely fill tanks and other voids that could have potentially posed a nuclear criticality has led to the generation of large volumes of low-level, high-level, and transuranic wastes. There is no accurate estimate of the total volume of Raschig rings that are radioactively contaminated and will require disposal; however, the current estimate approximates the volume to be more than 10,000 drums. These Raschig rings now compose a unique radioactive waste stream that pose a significant disposal dilemma.

Due to the large volumes these Raschig rings occupy, remediation of the rings could potentially be a viable and cost effective method for disposal of this radioactive waste stream. The disposal costs depend directly on the type of radioactive waste stream (i.e., low-level, high-level, transuranic waste). Currently, there is no established cost for disposal of transuranic waste; however, an estimate was made to show the potential economic potential for remediation of contaminated Raschig rings in Figure 1.

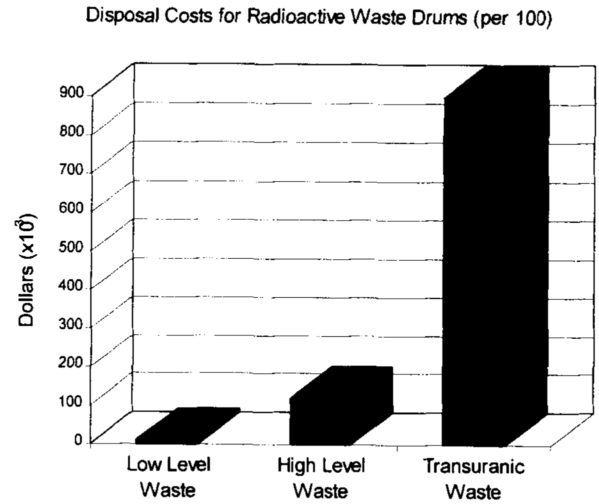

FIGURE 1. Approximate disposal costs of 55-gallon drums of low-level, high-level and transuranic waste.

A focused study was conducted on an uranium/thorium radioactive Raschig ring waste stream at the Radiochemical Development Facility at Oak Ridge National Laboratory. This waste stream was chosen to develop a methodology that could be applied to other similar radioactive contaminated Raschig ring waste streams and to recover a previously undesired isotope (i.e., thorium-229) for ongoing alpha radioimmunotherapy research (4-7).

MATERIALS AND METHODS

The borosilicate Raschig rings used for this study were from a legacy radioactive waste stream at Oak Ridge National Laboratory. The Raschig rings were used for criticality control during a uranium-233/thorium-229 separation in the mid 1970s. The original process involved a nitric acid dissolution of the material followed by an ion exchange separation of the thorium from the uranium (8). The thorium was then removed from the ion exchange media using an acetate complex. This material was kept in a storage tank and later processed for disposal. This left approximately 2 m^3 of radioactively contaminated borosilicate Raschig rings for disposal. The Raschig rings were stored in the empty tank until 1994 when they were removed for potential recovery of a residual isotope (i.e., thorium-229) for research and development applications in the nuclear medicine industry (9).

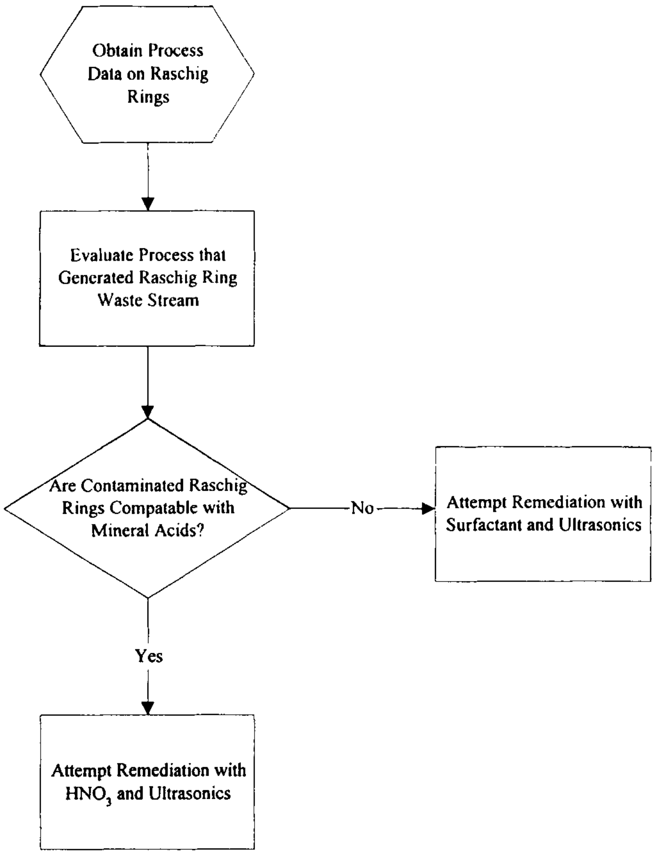

FIGURE 2. Basic schematic for determination of remediation of contaminated Raschig rings.

The residual radioactive contamination on the Raschig rings is a direct function of the process from which the Raschig rings were originally utilized. Typically, the radioactive contamination on the Raschig rings varies from a thin dry film to a sludge-like material that completely envelops the ring (10). However, by analyzing the process that led to the generation of the waste stream, a remediation scheme is proposed for effective removal of the radioactive contaminants from the Raschig rings (Fig. 2). The selected Raschig ring waste stream originated from a nitric acid dissolution process; therefore, a nitric acid/ultrasonic remediation method was chosen to demonstrate the remediation effectiveness on the Raschig ring waste stream. In support of this methodology, Raschig ring remediation with nitric acid and ultrasonic agitation was also recommended for the Raschig ring waste streams at the Rocky Flats Environmental Technology Site (10).

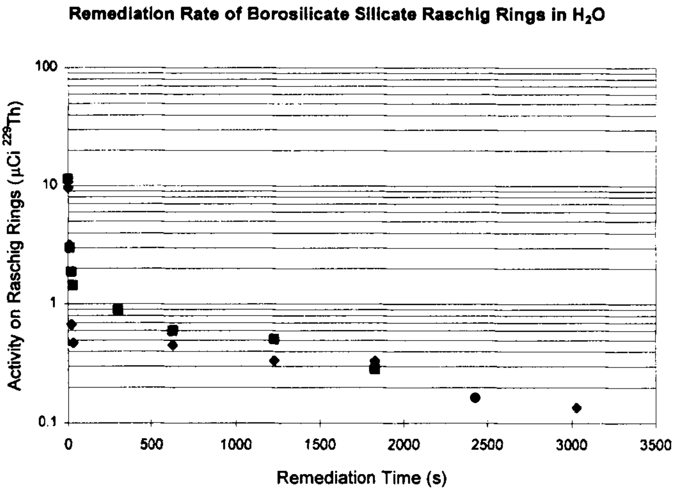

FIGURE 3. Remediation rate for two independent waste streams of radioactively contaminated borosilicate Raschig rings with deionized water and mechanical stirrer agitation.

RESULTS AND DISCUSSION

Initial remediation experiments were conducted with a mechanical stirrer and deionized water at room temperature (Fig. 3). This proved somewhat effective in remediation of the selected Raschig ring waste streams and provided a baseline to measure the effectiveness of the nitric acid/ultrasonic agitation remediation experiments.

The effectiveness of the remediation was measured through gross gamma radiation measurements and gamma spectroscopy. The gamma spectroscopy was used to determine the activity of the primary isotope that remained on the Raschig rings (i.e., thorium-229). The frequency of the measurements was developed from coarse remediation experiments to determine feasibility of the process. Remediation methodology focused on minimizing the amount of nitric acid necessary by utilizing only the amount necessary to submerge the Raschig rings. This was to support future large scale applications that would operate on a principal to minimize waste generation. In addition, to further minimize waste generation the nitric acid was recycled through an anion ion exchange column to remove the radioactive contaminants and therefore allow continued use of the nitric acid for remediation of additional Raschig ring waste streams. To effectively recover the radioactive contaminants (i.e., primarily thorium-229), a 7.5 N HNO_3 solution was used in conjunction with Reillex™ HPQ, a poly-4-vinylpyridine anion resin.

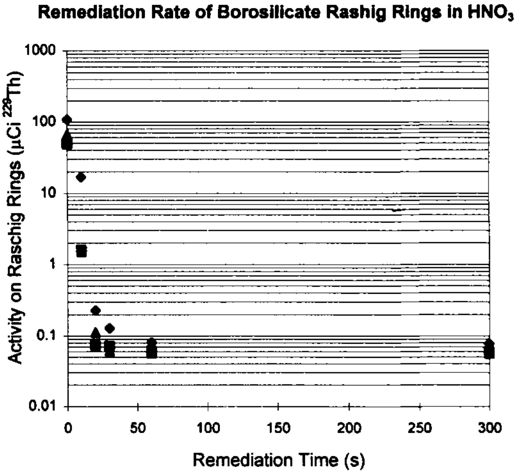

FIGURE 4. Remediation rate for three independent waste streams of radioactively contaminated borosilicate Raschig rings with 7.5 N HNO_3 and ultrasonic agitation.

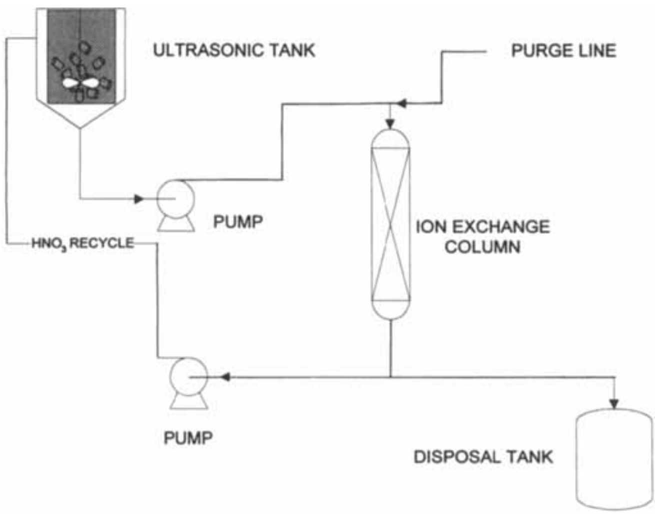

FIGURE 5. Benchtop remediation system for radioactively contaminated Raschig rings.

The 7.5 N HNO_3 in combination with ultrasonic cleaning system at room temperature effectively removed > 99% of the radioactive contaminants from the Raschig rings in less than one minute (Fig. 4). The Raschig rings were then removed from the acid solution and the solution was passed through the anion ion exchange column to remove the radioactive contaminants. After which, the nitric acid was reused to remediate additional Raschig ring waste streams. This combination proved the most effective for remediation of the Raschig rings and contaminant recovery.

The successful application of this remediation technology has lead to the design of a benchtop scale Raschig ring remediation system (Fig. 5). This system is currently under construction to test the potential applications of a large scale remediation process for Raschig ring waste streams that are contaminated with radioactivity and favor remediation with nitric acid. Additional development work is still necessary to develop an efficient process for remediation of Raschig ring waste streams with surfactants and ultrasonic agitation for waste streams not compatible with mineral acid.

SUMMARY AND CONCLUSIONS

Remediation of Raschig ring waste streams that are contaminated with radioactivity has been successfully demonstrated to be effective at removing > 99 % of the contaminants with nitric acid and ultrasonic agitation at room temperature. The economic incentives for application of a remediation process for radioactively contaminated Raschig rings are substantiated by the disposal costs of untreated high-level and transuranic waste. In addition, a basic remediation methodology has been suggested for application to all radioactively contaminated Raschig ring waste streams. However, variations in the application of the proposed remediation methodology are possible due to the various historical processes in which Raschig rings were involved. Additional development work will provide a broader empirical and theoretical base to support the remediation of other radioactively contaminated Raschig ring waste streams.

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