MASTER

42-38

RFP-2716

DESIGN AND OPERATION OF AIR-LIFT, CIRCULATED,
CASCADE DISSOLVER SYSTEMS

BY

CHARLES N. TESITOR

Rockwell International
Atomics International Division
P. O. BOX 464
Golden, CO.

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Department of Energy, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

THIS PAPER IS FOR PRESENTATION AT AN A.I.Ch.E. MEETING

THE SUBMITTED MANUSCRIPT HAS BEEN AUTHORED BY A CONTRACTOR OF THE U. S. GOVERNMENT UNDER CONTRACT EY-76-C-04-3533. ACCORDINGLY, THE CONTRACTOR, ROCKWELL INTERNATIONAL CORPORATION, AND THE U. S. GOVERNMENT RETAIN A NON-EXCLUSIVE, ROYALTY-FREE LICENSE TO PUBLISH OR REPRODUCE THE PUBLISHED FORM OF THIS CONTRIBUTION, OR ALLOW OTHERS TO DO SO, FOR U. S. GOVERNMENT PURPOSES.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

TABLE OF CONTENTS

Abstract.....	1
Summary.....	1
Introduction.....	2
Dissolver Development.....	4
Air-Lift Theory.....	6
Dissolver Description.....	8
Performance Data of Air-Lift Dissolvers.....	10
Feeder Design.....	12
Bibliography.....	16

ABSTRACT:

A continuous, cascade dissolver system, using an air-lift pump for circulation and suspension of solid particles has been developed. This dissolver system has been used to dissolve plutonium oxide, incinerator ash and other residues generated during the chemical processing of plutonium. An integral part of the dissolver system development involved the design and fabrication of a reliable continuous feed system for the dissolver.

SUMMARY:

A unique dissolver system based on the air-lift pump principle was designed and operated. Dissolution rates of 600 grams PuO_2 per hour were obtained. The dissolver system was also applied to the dissolution of incinerator ash. Plutonium recovery of up to 98% was realized utilizing the air-lift pump circulated dissolvers.

As part of the dissolver system, a continuous screw feeder was designed for use with the fine power being dissolved. These feeders have proven to be trouble-free and easy to operate.

INTRODUCTION

The handling and processing of plutonium results in the generation of a variety of residues. These residues may include such items as plutonium oxide, incinerator ash, slag and crucible if plutonium metal is produced, and sludges from a variety of filtration and other process steps.

The recovery and purification of the plutonium contained in these residues usually involves a dissolution or leaching process. The most common dissolution process consists of contacting the plutonium oxide or residue with a mixture of nitric acid and hydrofluoric acid at elevated temperatures.

The early equipment used in these processes, and still being used in many cases yet today, was patterned after laboratory studies and consisted of beakers of 2 to 4-liter capacity. Heating was supplied by electric immersion heaters in the 1000-watt range. The agitation was accomplished by using either the boiling action of the electric heater, an air sparge, mechanical agitation, or a variety of combinations of these methods.

When applied to a production scale operation, the batch-beaker method presented many problems. The fumes from the many dissolution beakers corroded the electric heater connections and presented problems for the glovebox air handling and filtration systems. The system was labor intensive and required much handwork in the glovebox. The efficiency of dissolution was not good; primarily as a result of poor suspension of the solids in the acid solution.

In an effort to improve the dissolution process, a system of continuous, cascade dissolvers was installed. This system consisted of air-sparged dissolver vessels, in series, overflowing by gravity from one to the other. The feed mechanism was a vibratory feeder of laboratory size.

While the continuous, cascade system was an improvement over the batch pots, serious problems still existed. The suspension of solids was difficult. A build-up of solids, termed "heel," would slowly fill the vessels rendering them inoperable. The large air sparge requirements necessary for even minimal particle suspension filled the glovebox hoods with fumes. Finally, the vibratory feeders were subject to transient equipment vibrations and fed erratically, if not continuously monitored. Clearly, an improved dissolution system was needed.

Mechanically agitated dissolver systems had also been tried. The most notable of these attempts was the multi-stage dissolver system. This system consisted of a set of propeller agitated dissolver vessels arranged in series. The propellers were driven by electric motors located externally to the glovebox. A mechanically sealed shaft passed into the glovebox through a guide bearing and into each of the dissolver vessels. At the end of each shaft a propeller was attached.

The initial startup of this system indicated excellent dissolution rates (unfortunately exact rates could not be found at the time of this writing); however, mechanical problems beset the system almost immediately. Glovebox shaft seals, propeller blade construction and guide bearing failures

occurred at a rate that was unacceptable to the production operation. After several attempts at repair and modification, the multi-stage dissolver system was abandoned.

Dissolver Development

A study of the existing air-sparged, cascade dissolver system was initiated. This study generated the major areas of deficiency where improvements in equipment design were needed.

By visual observation it was apparent that a lack of adequate particle suspension was preventing contact between the nitric acid solution and the plutonium oxide. Increasing the sparge rate increased the degree of particle suspension but had the adverse effects of cooling the dissolver and caused large volumes of acid vapor to escape into the glovebox.

It has been shown that complete suspension of particles with settling velocities up to 0.25 ft/sec. can be obtained by using 3 ft.³ of air per square foot of vessel cross section.⁽¹⁾ This data was generated using vessels of large cross sections. No data or correlation for vessels in the 4-inch diameter range could be found. It was decided to use the large vessel data to see if it matched the situation as experienced in operating the cascade dissolvers.

Using the equation

$$V_t = \frac{2G_c M_p (P_p - P_i)^{1/2}}{P_p P_i A_p C_d}$$

where

V_t = terminal velocity of a particle, ft/sec;

G_c = acceleration of gravity, ft/sec²;

M_p = the mass of the particle, lbs.;

P_p = density of the particle, lb/ft³ (641 lb/ft³)⁽²⁾;

P_l = density of the liquid, lb/ft³;

A_p = projected area of particle in axis of motion,
ft²;

C_d = drag coefficient, dimensionless

the terminal velocity of a particle of plutonium oxide can be estimated to be 1.35 ft/sec. Assuming the air sparge required would vary with square of the terminal velocity, a sparge rate of 8.2 ft³/min. was calculated as being required to suspend the plutonium oxide particles. Actual practice showed that sparge rates of 5 to 6 ft³/min. were sufficient to suspend most of the solids in the cascade dissolvers.

Sparge rates of this magnitude caused severe off-gas problems in the glovebox. The sparge air was saturated with acid vapors and the volume of gas was beyond the capacity of the off-gas collection system. The nitric acid vapor released escaped into the glovebox and condensed upon the glovebox windows and gloves, shortening their useful life. Therefore, since the calculated minimum sparge rate compared reasonably to the sparge rates required in operation, it was apparent that a significant reduction in sparge air rate would not be possible using the existing dissolver design. Since reduction of the air sparge rate was of major interest, attention was turned to the air-lift pump.

Air-Lift Theory

The air-lift pump principle has been used for many years primarily in the mining industry for the leaching of ores. As improved materials of construction have become available, the advantages of the air-lift pump have been reduced. However, for particularly rigorous applications, i.e., the dissolution PuO_2 in a Nitric-Hydrofluoric acid system, the air-lift retains its advantages.

The air-lift pump relies on the density difference between two-phase gas-liquid mixture and the liquid itself. Figure 1 shows a cut-away view of a simple air-lift pump. Air or other gas is introduced into the bottom of Draft Tube A.

FIGURE I
Cut Away View Of
Air-Lift Principle

$$\Delta p = h(\rho_0 - \rho_i)$$

As the gas rises, a two-phase system of liquid and gas is formed along the inside length of the draft tube. The density of the two-phase system inside the draft tube is, of course, less than the density of the corresponding single-phase liquid on the outside of the draft tube. This difference in density results in a pressure gradient between the bottom of the draft tube and the single-phase liquid surrounding the draft tube. Equation (1) expresses the pressure gradient in terms of the draft tube height.

$$\Delta P = h(P_o - P_i) \quad (1)$$

The pressure differential, ΔP , between the inside and outside of the draft tube causes the liquid to flow into the tube. The rate of liquid flow is determined by the pressure drop of the flowing, two-phase system inside the draft tube. At steady state conditions the pressure differential at the bottom of the draft tube equals the friction losses inside the draft tube. The theory of operation of air-lift pumps has been studied by others.^(3,4) Since the success of the air-lift pump dissolver is a function of mechanical design and not one of two-phase flow theory, this paper will not dwell on theory any further.

Experience with dissolver and other plutonium processing equipment has taught that two fundamental problems must be overcome for a process to be successful. The first of these involves proper selection of the materials of construction. The corrosive acid solutions and the abrasive nature of plutonium oxide, particularly if the oxide was formed at high temperatures, magnifies the impact of the material selections. The second problem area involves the maintenance of the process equipment in the glovebox environment.

Dissolver Description

An air-lift, circulated, cascade dissolver unit is shown in Figure 2.

Plutonium oxide and 8% by weight calcium fluoride are fed to the air-lift dissolver by means of a screw feeder. The design of the screw feeder was a critical factor in the success of the dissolver system and is discussed later in this paper. Nitric acid at a concentration of 12M is fed into a trough at the discharge of the screw feeder. The nitric acid washes the plutonium oxide-calcium fluoride mixture into the first air-lift dissolver unit. Air is supplied to the bottom of the draft tube as shown. The pumping action of the air-lift circulates the plutonium oxide-nitric acid slurry in the dissolver. The slurry overflows the first unit and flows by gravity to the next unit which in turn overflows into a third unit and so on; hence, the name cascade dissolver. The dissolver units themselves are made of heavy-wall, glass pipe, molded plastic and stainless steel. No welded, metal parts are in contact with the process solution. The average longevity of a dissolver unit is one year. The steam supply tubing is made from 304 stainless steel tubing and lasts from two to three months.

A Teflon[®] insert is placed in the bottom of the dissolver to streamline the draft tube inlet and prevent the accumulation of undissolved solids in the bottom corners of the dissolver. The steam supply tube is U-shaped and is inserted inside the draft tube. By having the steam supply inside the draft-tube, steam leaks in the steam supply tubing do not stop the air-lift pump as was the case with the steam supply on the outside of the draft tube. To

[®] Trademark of E. I. duPont de Nemours and Co., Wilmington, Delaware

FIGURE 2

AIR-LIFT, CIRCULATED,
CASCADE DISSOLVER UNIT

facilitate replacement of the steam tubing, the draft tube is extended through the lid of the dissolver. The glass section is assembled using standard, flared, heavy-wall glass pipe and flanges with Teflon[®] gaskets and compression rings. The top of the dissolver is made of molded Kynar.^{®*}

The dimensions of the dissolver units vary depending upon the application and glovebox availability. Dissolvers used for plutonium oxide dissolution are four inches in diameter by 30 inches high. Incinerator ash dissolvers are six inches in diameter by 40 inches high. In one application, the dissolvers are six inch diameter by eighteen inches high. The dimensions are limited only by the user's imagination and application.

Performance Data of Air-Lift Dissolvers

The first set of air-lift, circulated dissolvers were installed in 1971. This first set of dissolvers consisted of three units in series, each unit being four inches in diameter by 30 inches tall. Prior to installing the system in the glovebox, a full-size unit was operated using water and a silica-based oil-dry material as a stand-in for plutonium oxide.

The slurry of oil-dry and water was successfully circulated using an air flow rate of 0.2 SCFM. A total of 1.4 kilograms of oil-dry was added to the four liters of solution in the dissolver. This solid-to-liquid ratio was three times the solid-to-liquid ratio expected to be encountered in actual operation. The air-lift pump had no difficulty in keeping the solids in suspension in the liquid.

^{®*} Trademark of Pennwalt Corp., Philadelphia, PA.

The liquid circulation rate in the dissolver was determined by filming the dissolver in operation. The rate of fall of the oil-dry material in still water was determined. The rate of fall of the oil-dry material in the dissolver while operating was determined. By comparing the two rates of fall, the actual liquid velocity could be calculated. The average circulation rate in the dissolver, using an air rate of 0.2 SCFM, was 30 liters per minute.

A set of three cascade dissolvers was installed in an existing glovebox. The operating characteristics of the dissolver system is shown in Table I.

TABLE I

Operating Characteristics
Of Air-Lift, Circulated, Cascade
Dissolver System

PuO ₂ Feed Rate	700-1000 Grams/Hr.
Acid Feed Rate	8-10 Liters/Hr.
Air Rate	0.15-0.4 SCFM
Liquid Circulation Rate	30 Liters/Minute
PuO ₂ Dissolution Rate	500-600 Grams/Hr.

During normal operation between 60 and 75% of the PuO_2 was dissolved per pass through the dissolver system. The remaining solids were filtered, dried and recycled through the dissolvers. An overall dissolution efficiency of 95-97% based on the PuO_2 fed was achieved by recycling the undissolved solids.

Feeder Design

One of the most troublesome parts of the dissolver process was the solids feeding mechanism. During the operations reflected in Table I, a vibratory feeder was used as the feed mechanism. This unit was subject to erratic behavior due to extraneous equipment vibration and corrosion. The vibratory feeder was replaced by a simple screw feeder with a 1.5-liter capacity feed hopper. The screw feeder was subject to packing in the screw section and bridging of material in the hopper.

A new feeder was designed to overcome these problems. The basic design continued to be a screw-type feeder. Several modifications were made to permit uninterrupted operation without the plugging and bridging problems.

The auger was redesigned to incorporate a variable pitch design as shown in Figure 3.

FIGURE 3
Screw Feeder Auger

FIGURE 3. Screw Feeder

The variable pitch design prevents the auger from packing material into the auger barrel and plugging since the feed capacity of the auger increases along its length.

The solids hopper was redesigned to incorporate a chain-driven agitator inside the hopper and the walls of the hopper were inclined as shown in Figure 4.

FIGURE 4
Screw Feeder Hopper

The inclined walls prevent packing and bridging of fine powders in the hopper. The agitator serves to also prevent bridging of the feed material and moves the material down into the auger. The auger rotates at a speed of 0.25 rpm to 0.33 rpm. The screw feeder is driven by a 1/15 hp, dc electric motor with a 1000:1 gear reducer and a speed-controller for the motor.

With the success of the air-lift circulated dissolvers processing plutonium oxide, a new dissolver system for incinerator ash residues was designed.

The dissolver system for incinerator ash is similar to the air-lift dissolvers previously described for plutonium oxide dissolution. The major difference being that the ash dissolvers were six inches in diameter. Johnson⁽⁵⁾ has described the operation of air-lift circulated dissolvers in processing incinerator ash residues. Johnson shows that in a two-year period over 9,000 kilograms of incinerator ash and ash heel were leached in the air-lift circulated dissolvers. By using five successive leaches, up to 98% of the plutonium in the original incinerator ash was recovered in the leach solution.

BIBLIOGRAPHY

1. J. H. Perry, "Perry's Chemical Engineer's Handbook," Third Edition, 1950
2. O. J. Wick, "Plutonium Handbook," Volume I, 1967
3. Nicklin, D. J., "The Air-Lift Pump: Theory and Optimization," Trans. Instn. Chem. Engrs., Vol. 41, 1963
4. Andeen, G. B., "Bubble Pumps," Compressed Air Magazine, January, 1974.
5. Johnson, T. C., "Recovery of Plutonium From Incinerator Ash at Rocky Flats," RFP-2520, December 21, 1976