



Project Summary

Separation of Hazardous Organics by Low Pressure Membranes: Treatment of Soil-Wash Rinse-Water Leachates

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Soil washing is a promising technology for treating contaminated soils. In the present work, low-pressure, thin-film composite membranes were evaluated to treat the soil-wash leachates so that the treated water could be recycled back to the soil washing step. Experiments were done with SARM (Synthetic Analytical Reference Matrix) soils. Membrane performance was evaluated with leachates obtained from different wash solutions. The effect of fine suspensions in the leachates was also studied. A solution-diffusion model was modified to include an adsorption resistance term in water flux, and this term was correlated with bulk concentration using the Freundlich isotherm. This correlation was then used to predict water flux drop at different bulk concentrations or to predict water flux at different recoveries. Thin-film composite membranes were found to effectively treat the leachate from rinse water used to wash contaminated soil. In addition, feed preozonation significantly improved water flux.

This Project Summary was developed by EPA's Risk Reduction Engineering Laboratory, Cincinnati, OH, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

Soil contamination is one of the major environmental problems of today. Recently

enacted legislations and the high costs and high energy requirements associated with conventional excavation and incineration, with subsequent disposal in a landfill, have created a need for innovative, cost-effective technologies for cleanup. Soil washing is a promising technology for treating contaminated sites, and it is one of the most successful full-scale technologies developed in Europe for site remediation. Cost-effective remediation by soil washing would, however, require simultaneous development of effective rinsate treatment techniques for separating and concentrating dissolved hazardous pollutants in the wash water and recycling back the treated water for soil washing. These needs were the motivation for the present work. Previous research on removal of contaminants from leachates by low pressure membranes and pre-ozonation was also performed.^{1,2}

Membrane processes provide a very promising treatment technology for this particular purpose—treating soil-wash rinse-water leachates. Membrane separation processes consume less energy than conventional processes, and membrane systems are compact and modular. Microfiltration, ultrafiltration, reverse osmosis (RO), and electrodialysis are fully developed membrane technologies, and pervaporation is a developing membrane technology. In recent years, RO has emerged as a fully developed, mature technology, and the estimated worldwide sales of RO membranes in 1988 were \$118 million with a projection of \$335 million for 1998. Considerable information is now available for full-scale application of RO technology in terms of membrane materi-



als, module design, and cost estimation. High-pressure RO membranes are used for sea water desalination (osmotic pressure - 2.34 MPa), and low-pressure RO membranes are used traditionally for brackish water desalination (osmotic pressure - 0.1 to 0.28 MPa). Low-pressure RO membranes have lower capital and operating cost, and because of low pressure requirements, they can be used in spiral element design, thus minimizing membrane fouling. For 3,785 L of purified water, the energy requirements of a high-pressure RO process are about 3 to 4 times that of a low-pressure RO process (1.38 to 2.76 MPa), and distillation is about 8 to 18 times that of a low-pressure RO process. Thin-film composite membranes provide high water flux and higher rejection. The osmotic pressure of most hazardous waste streams is in the range of that of brackish water. In view of these attractive properties, it was decided to use low-pressure, thin-film composite membranes to treat the leachates from rinse water used in soil washing. The ultra-low-pressure RO process (or nanofiltration) can be used in combination with low-pressure RO because a nanofiltration membrane permeates monovalent ions but rejects divalent and multivalent ions, as well as organic compounds having molecular weights greater than 200.

This work deals with the use of thin-film, low-pressure composite membranes for concentrating and separating hazardous pollutants in the soil-wash rinse-water leachates from SARM soils. Soil washing experiments were done with different wash solutions (pure water, nonionic surfactant solutions, and EDTA solutions). The separation characteristics of the membranes were evaluated in terms of membrane feed total organic carbon (TOC), heavy metal concentration, dissolved solids, suspended solids, pH, presence of specific compounds in the feed, feed preozonation, extent of recovery, and water flux. In addition, the flux drop results were correlated by using a solution-diffusion transport model containing an adsorption term.

Experimental

Figure 1 gives an overview of the different experiments and studies done. Most of the experiments were done with SARM IV. For soil washing experiments, a 10:1 wash solution-to-soil ratio was used for all experiments. Mixing time was fixed for 1 hr, and for most of the experiments, a single rinse was done. For most of the experiments, suspended solids from the leachate were removed by vacuum filtration with the use of a .0.22- μ m pore-size filter. Membrane

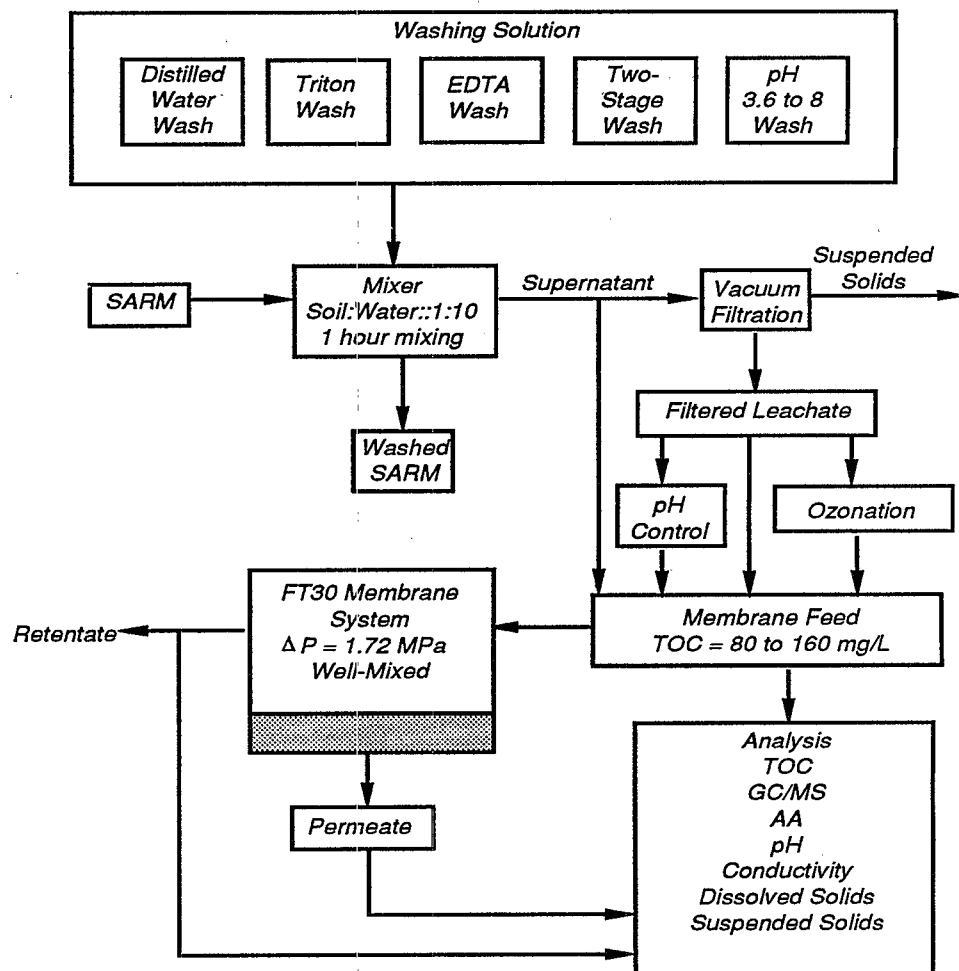


Figure 1. Schematic of the overall experimental plan.

studies were mostly done in a batch system at a system pressure of 1.75 MPa. High mixing conditions were maintained. The continuous run (in the presence of suspended solids) was also done at the same system pressure, and a pump provided a flow of solution through the continuous cell. The membrane was cleaned with a 10% to 30% water-methanol solution. Feed was preozonated in a 500-mL stirred reactor with a flow of 0.2 standard L/min O_2 containing 2% ozone.

Filtered leachates, membrane feeds, retentates, and permeates were analyzed by TOC analyzer, atomic absorption spectrophotometer (AA), and GC/MS. Some analyses were also done by a U.S. Envi-

ronmental Protection Agency support laboratory.

Results and Discussion

The consistency of distilled water fluxes and sodium chloride rejections over an extended operating period (> 200 days) demonstrated that the FT30* membranes used were stable. Figure 2 summarizes (1) the membrane performance with leachates obtained by washing SARM IV with different wash solutions and also (2) the performance with ozonated leachates. The distilled water fluxes for all the experi-

* Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

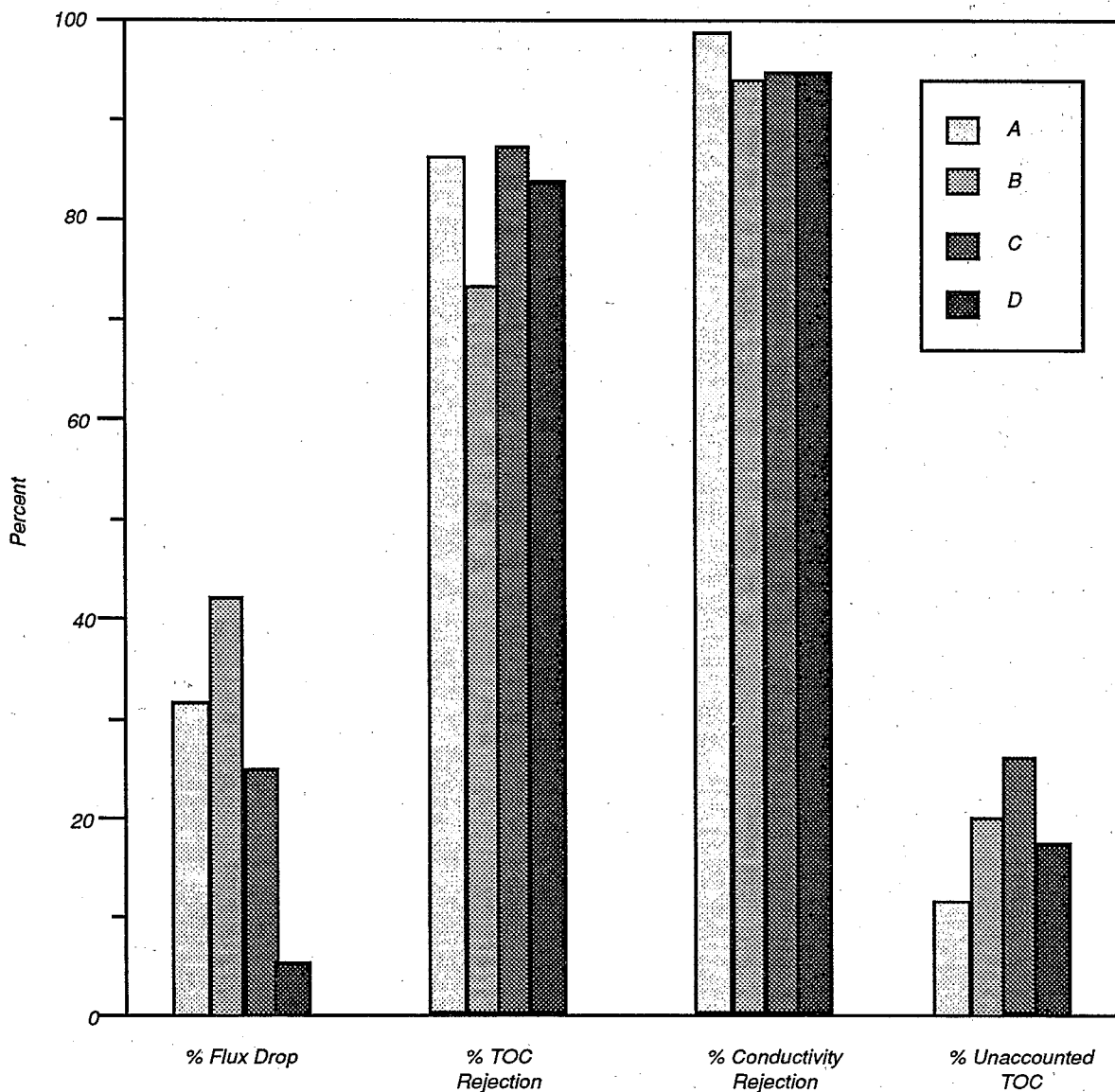


Figure 2. Summary of membrane performance with different leachates for SARM IV (A = EDTA wash solution; B = surfactant wash solution; C = distilled water wash; D = ozonation).

ments were in the range of 9 to 12×10^{-4} cm/s, and the pH of the membrane feeds were in the range of 5.8 to 6.8. Membrane runs recovery ranged between 17% and 80%. For the EDTA wash, 1.01 mol/L EDTA was used; for surfactant wash, 0.04% Triton X-100. For the ozonation experiment, ozonation time was 10 min. Figure 3 compares the typical water flux behavior of distilled water wash leachate and surfactant wash leachate. There was a 10% higher flux drop after 17% recovery for surfactant wash leachate, but as indicated in Figure 2, there was a 5% decrease in amount of organics adsorbed on the mem-

brane surface (% Unaccounted TOC). This could be because of the surfactant's ability to form micelles and bind hydrophobic organics. Figure 2 also shows that TOC rejections and conductivity rejections were high, which indicated good membrane performance. Table 1 shows the effect of EDTA wash on soil leachability of Cu, Ni, Pb, and Zn and their rejections by the membrane. EDTA washing enhanced leachability of metals. This could be explained by the higher stability of metal-EDTA complexes. For almost all the runs, the rejections of metals were 92% to 98%. Figure 4 shows the water flux behavior for

ozonated and nonozonated leachates. For the ozonated leachates there was only a 5% flux drop; for the nonozonated leachates, the flux drop was between 20% and 25%. This suggests that ozonated products do not interact strongly with the membrane.

Raising the pH of the leachate precipitated some heavy metals and also removed some TOC with the precipitate. When the pH of the leachate was lowered some white cloudiness formed. The presence of fine suspensions for a low recovery reduced organic adsorption on the membrane surface. At a high water recov-

ery (80%), total dissolved solids (primarily inorganics) contributed significant osmotic pressure. To predict water flux at different recoveries, the solution-diffusion transport model was modified to include an adsorption resistance term. Figure 5 shows that experimental and predicted values (at high water recoveries) agreed well. The adsorption resistance term was correlated with bulk concentration using the Freundlich isotherm.

Conclusions

This study has shown that thin-film, composite membranes can effectively treat soil-wash rinse-water leachates to produce permeates for reuse. The permeate can be recycled back to the soil washing step. If the permeate needs to be discharged, further treatment may be required. The treatment of the concentrated stream would be much easier and cost-effective because of the reduced volume to be treated. The advantages of this membrane process are that it is compact and modular and it has high solute separations at low pressures (< 2 MPa), high water flux, low energy and capital costs, and broad pH operating range. If EDTA recovery is also one of the objectives, then a loose RO membrane like a nanofiltration membrane may be used to recover EDTA with further treatment of the permeate. In addition, the ozonation-membrane process would effectively reduce the flux drop and increase the over-all rejections.

Membrane rejections were found to be high in terms of selected compounds: pentachlorophenol (> 98%), 4-aminobiphenyl (> 93%), ethylbenzene (> 97%), xylene (> 81%), 4-chloroaniline (> 90%), and 2,4-dinitrophenol (> 98%). At 80% water recovery, there was increased flux drop due to increased osmotic pressure of total dissolved solids and increased adsorption of sparingly soluble organics on the membrane because their solubility limit was exceeded. Feed preozonation, EDTA wash, and surfactant wash reduced adsorption of organics on the membrane. The modified solution-diffusion model was in good agreement with the experimental values in predicting water flux at different recoveries.

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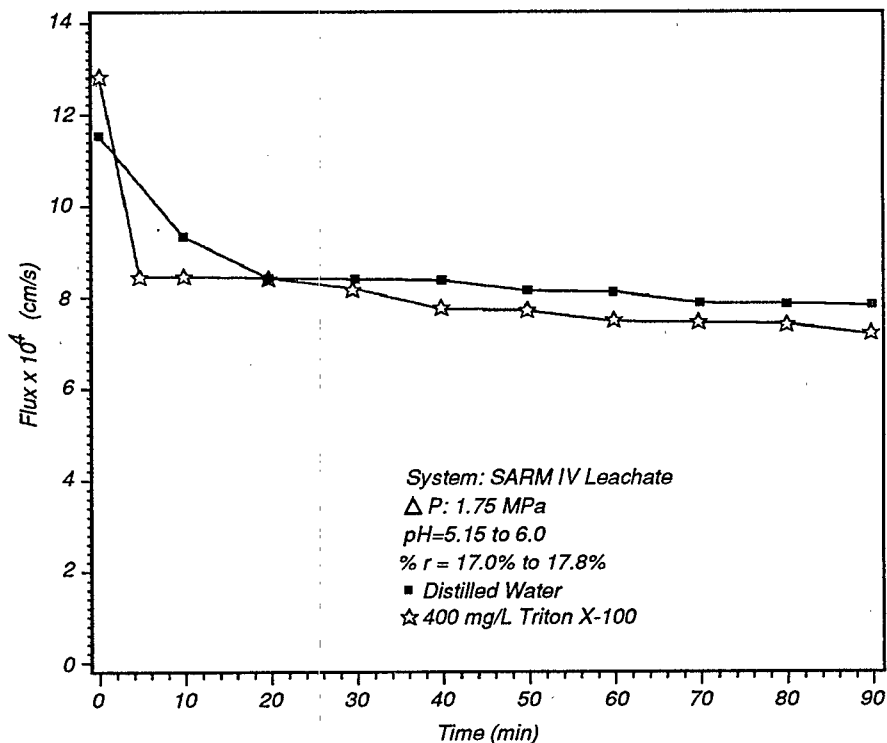


Figure 3. Permeate flux versus time for leachate after washing SARM IV with distilled water and surfactant (0.04% Triton X-100).

Table 1. Summary* of Effect of EDTA Washing on the Leachability and Rejection of Certain Metals

Active Ingredient mmoles/L	Feed, mg/L				% r	Rejection, %			
	Cu	Ni	Pb	Zn		Cu	Ni	Pb	Zn
0	1.58	10.5	1.72	224	17.8	96.2	93.1	96.5	93.3
0.10	20.7	14.4	8.19	372	18.7	>99.5	>99.3	>98.8	97.5

* Conditions:
 SARM IV:Wash Solution::1:10
 $\Delta P = 1.75$ MPa
 Chelant = Versene 100EP (TOC = 13.1% with 39% Active Ingredient)
 Active Ingredient = $\text{Na}_4\text{EDTA} \cdot 4\text{H}_2\text{O}$

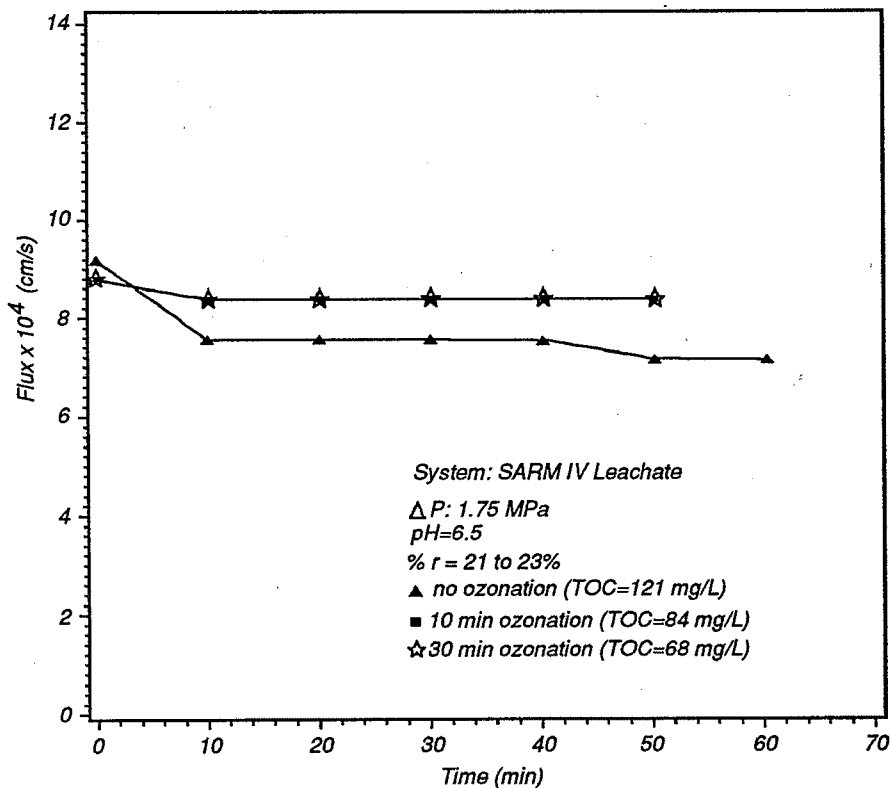


Figure 4. Permeate flux versus time for SARM IV leachate ozonated for different times.

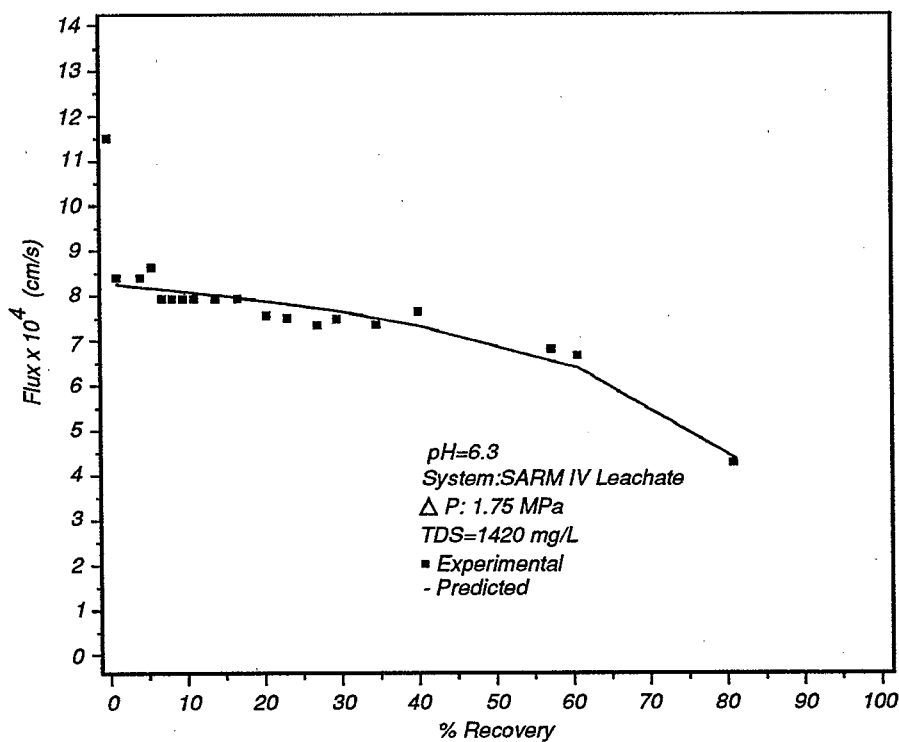
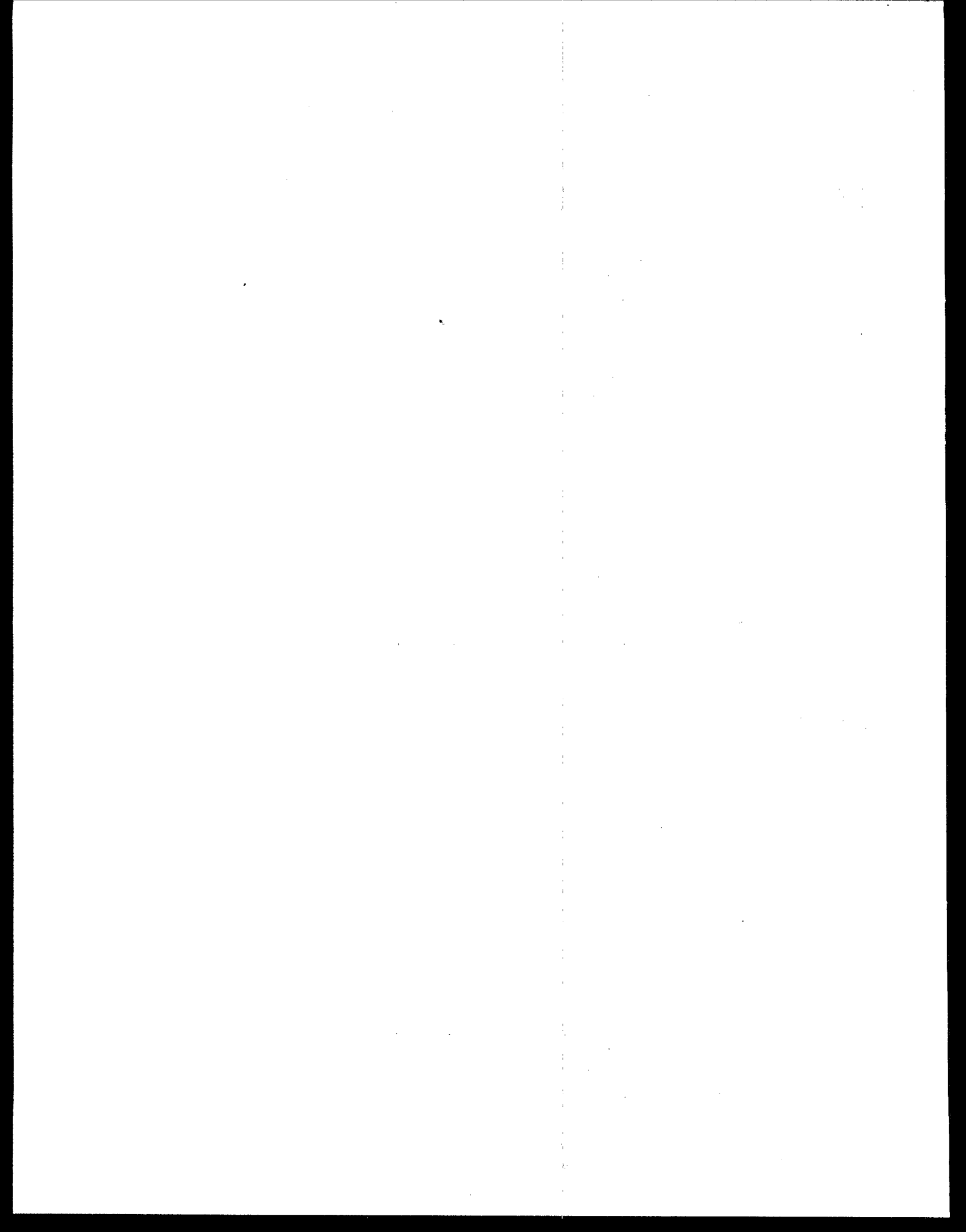
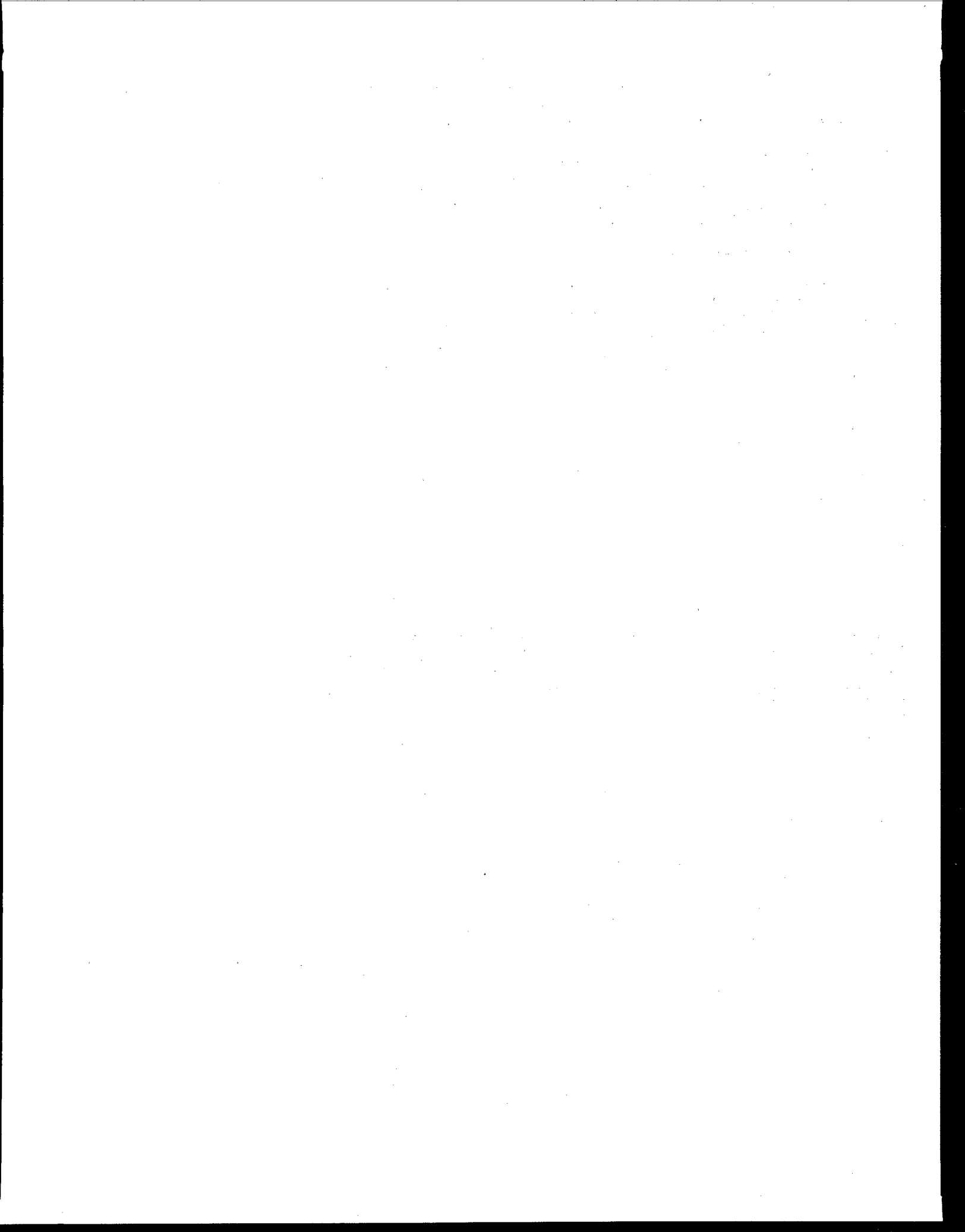


Figure 5. Experimental and predicted water flux with percent recovery.

References

1. Bhattacharyya, D., and Williams, M.E., "Separation of Hazardous Organics by Low Pressure Reverse Osmosis Membranes—Phase II, Final Report," EPA/600/2-91/045 (Jan. 1992)
2. Bhattacharyya, D., Barranger, T., Jevtitch, M., and Greenleaf, S., "Separation of Dilute Hazardous Organics by Low Pressure Composite Membranes," EPA/600/2-87/053 (1987).





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Richard Lauch is the EPA Project Officer (see below).

The complete report, entitled "Separation of Hazardous Organics by Low Pressure Membranes: Treatment of Soil-Wash Rinse-Water Leachates," (Order No. PB92-153436/AS; Cost: \$26.00, subject to change) will be available only from:

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