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Cost and Systems Analysis of Innovative Fuel Resources Concepts

Fuel Cycle Research and Development

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Executive Summary

Economically recovered uranium from seawater can have a transformative effect on the way policy makers view the long-term viability of uranium based fuel cycles. Seawater uranium, even when estimated to cost more than terrestrially mined uranium, is integral in establishing an economic backstop, thus reducing uncertainty in future nuclear power costs. While a passive recovery scheme relying on a field of polymer adsorbents prepared via radiation induced grafting has long been considered the leading technology for full scale deployment, non-trivial cost and logistical barriers persist. Consequently, university partners of the nation-wide consortium for seawater uranium recovery have developed variants of this technology, each aiming to address a substantial weakness. The focus of this NEUP project is the economic impacts of the proposed variant technologies.

The team at University of Alabama has pursued an adsorbent synthesis method that replaces the synthetic fiber backbone with a natural waste product. Chitin fibers suitable for ligand grafting have been prepared from shrimp shell waste. These environmental benefits could be realized at a comparable cost to the reference fiber so long as the uptake can be increased or the chemical consumption cost decreased.

In an effort to reduce the number of input materials and process steps required for fabrication of the reference fibers, a team at the University of Maryland engineered novel adsorbents utilizing an oxylate ligand grafted on nylon fibers. The use of these materials allows for radiation induced grafting without any organic solvents, offering a "greener" fabrication process and lower chemical costs. If this novel adsorbent can achieve an uptake like that of the reference material, then a 10% cost savings could result.

Research at the University of Idaho has focused on the back end of the uranium recovery process. Traditionally adsorbent fibers have been treated with harsh acidic compounds to collet uranium off the braids after each deployment. The acidic elution process however requires a costly alkaline regeneration step prior to each recycle and has been linked to degradation of adsorbent capacity. Therefore the University of Idaho team developed a mild basic elution process utilizing bicarbonate to remove uranium off

adsorbents, resulting in reduced uranium recovery costs and a greater number of economical adsorbent recycles.

Although the adsorbent fabrication makes the most significant contribution to the seawater uranium production cost, the mooring and deployment cost effectively establishes a cost floor by governing the maximum number of economical adsorbent recycles. Therefore, researchers at the Massachusetts Institute of Technology designed a symbiotic deployment structure where uranium adsorbing braids are moored to the base of an offshore wind turbine. A belt and pulley system guides an adsorbent net through reaction tanks, allowing for the continuous off-shore elution and redeployment of the adsorbent. By circumventing labor and capital costs, this deployment system can offer up to a 30% cost savings as compared to the reference system.

The uranium capacity of adsorbent fibers has been shown to be a significant driver of the final uranium production cost. Partners at Hunter College of the City University of New York have attempted to increase capacity by enhancing the uranium affinity of the amidoxime ligand used in reference fibers. In addition to amidoxime, these novel fibers contain a second amine ligand that offers two mechanisms of facilitating amidoxime-uranium interaction. Although initial experimental data has indicated these novel adsorbents have lower uranium uptake, our study shows that if the uranium capacity can be increased 50% above the levels observed a 16% savings could result.

Introduction

Economically recovered uranium from seawater can have a transformative effect on the way policy makers view the long-term viability of uranium based fuel cycles. Seawater uranium, even when estimated to cost more than terrestrially mined uranium, is integral in establishing an economic backstop, thus reducing uncertainty in future nuclear power costs. While a passive recovery scheme relying on a field of polymer adsorbents prepared via radiation induced grafting has long been considered the leading technology for full scale deployment, non-trivial cost and logistical barriers persist. Consequently, university partners of the nation-wide consortium for seawater uranium recovery have developed variants of this technology, each aiming to address a substantial weakness. The focus of this paper is the economic impacts of the proposed variant technologies.

The remainder of the introduction enumerates our milestones in tabular form. The specific methodology applied to accomplish each task is described in the following section. All adsorbent technologies were evaluated using the same discounted cash flow techniques applied to the lifecycle of the reference ORNL adsorbent [1] [2]. Similarly, relevant input data regarding material and equipment costs, scaling factors, design parameters, and the like can be found in [2], while all new input data unique to novel technologies is displayed in the document. More detailed results of the cost analyses of select adsorbent technologies can be seen in the Papers section of this work.

Milestones	Deliverable
Task 1: Prepare Survey of Process Data and Performance to facilitate data collection from NEUP teams-**100% complete**	Finalized survey prepared and distributed; data collected
Task 2: Update cost and energy analysis to model developments by University of Alabama-**100% complete**	Presentation of resulting system analyses at working group meeting
Task 3: Update cost and energy analysis to model	Presentation of resulting system

Milestones	Deliverable
developments by University of Maryland-**100% complete**	analyses at working group meeting
Task 4: Update cost and energy analysis to model developments by University of Idaho-**100% complete**	Presentation of resulting system analyses at working group meeting
Task 5: Conduct System analyses in support of Massachusetts Institute of Technology-**100% complete**	Presentation of resulting system analyses at working group meeting
Task 6: Conduct System analyses in support of New York University-**100% complete**	Presentation of resulting system analyses at working group meeting

Cost and Systems Analysis methodology

Task 1: Survey

The task of preparing a *Survey of Process Data and Performance* was completed by July of 2013. The survey was designed to guide the transfer of information from an NEUP process to easy to implement data for the economic analysis. Basic high-level equipment and material templates from the NEUP teams will guide the initial conversations to generate the highest fidelity economic models.

The survey included explanation text, sections for materials necessary for completion of the NEUP process, descriptions of equipment, and space for process flow diagrams. Examples of equipment used, chemicals, process flow diagrams, and materials/descriptions are included in the document. The document explores opportunity for data collection throughout the systematic process: major equipment items and commodity, material and energetic inputs to the adsorbent fabrication, mooring and deployment, elution/regeneration and disposal steps. Detailed attention to known sensitive information topics is kept based on ORNL extensive spreadsheet quantifying and topical review.

The survey provides the structure for data transfer. It is simple in design and in-depth in topical coverage. From types of materials and equipment to steps in the uranium recovery from seawater process, the survey creates opportunities for a comprehensive and organized exchange of complex and potential known unknowns of data.

Task 2: University of Alabama

The team at University of Alabama has set out to address environmental concerns regarding the introduction of large quantities of plastic to marine ecosystems. In doing so they are attempting to eliminate the use of plastic altogether by pursuing an adsorbent synthesis method that replaces the synthetic fiber backbone with a natural waste product. Chitin nanomats suitable for ligand grafting have been prepared from shrimp shell waste. Through close collaboration with researchers at the University of Alabama the production process of chitin based adsorbents was modeled and analyzed for economic feasibility. While progress on these adsorbents is still ongoing, the remainder of this section will describe the production method and associated cost from the most recent collaboration.

The adsorbent synthesis process used in the economic model follows the chemistry described in [3] and can be seen in Figure 2.1. In its raw waste form, chitin exists simply as wet shucked crab and shrimp shells which must be converted into usable powder. Wet shells are pressed with a screw press, dried, and ground to result in a powder composed of chitin and proteins. The shell-derived powder is then dissolved in an ionic liquid, 1-ethyl-3-methylimidazolium acetate, to separate the chitin from proteins via an electrospinning process. The resulting fibers consist of high molecular weight chitin chains, providing ample binding sites for amidoxime ligands.

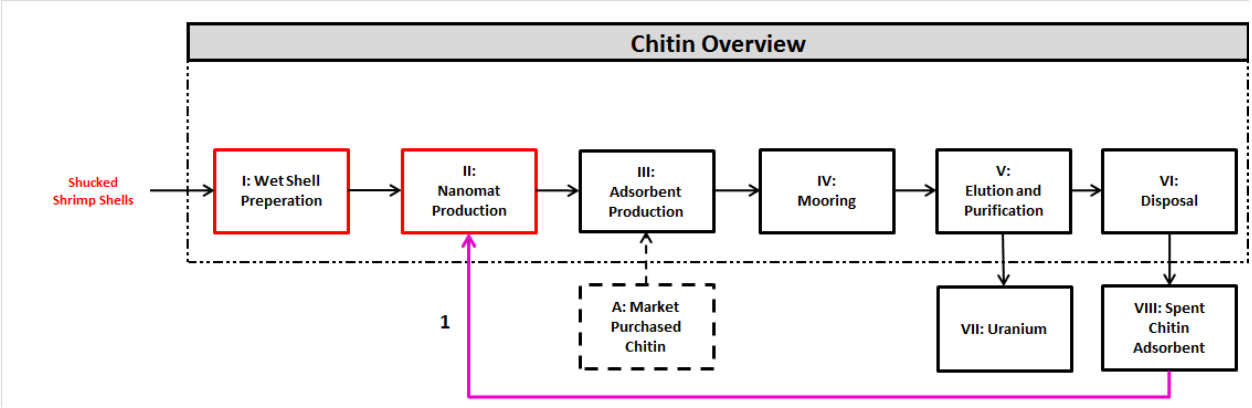

Figure 2.1 Adsorbent synthesis process for chitin nanomats

Although the amidoxime ligand used for the chitin based adsorbents is chemically identical to that of the reference fibers, the process of attaching it to the chitin nanomats is unique. Instead of relying on irradiation induced graft polymerization, the University of Alabama method [3] is a strictly chemical grafting process, pictured in Figure 2.2. The degree of ligand grafting achieved with the chitin substrate was seen to be on the order of 10%.

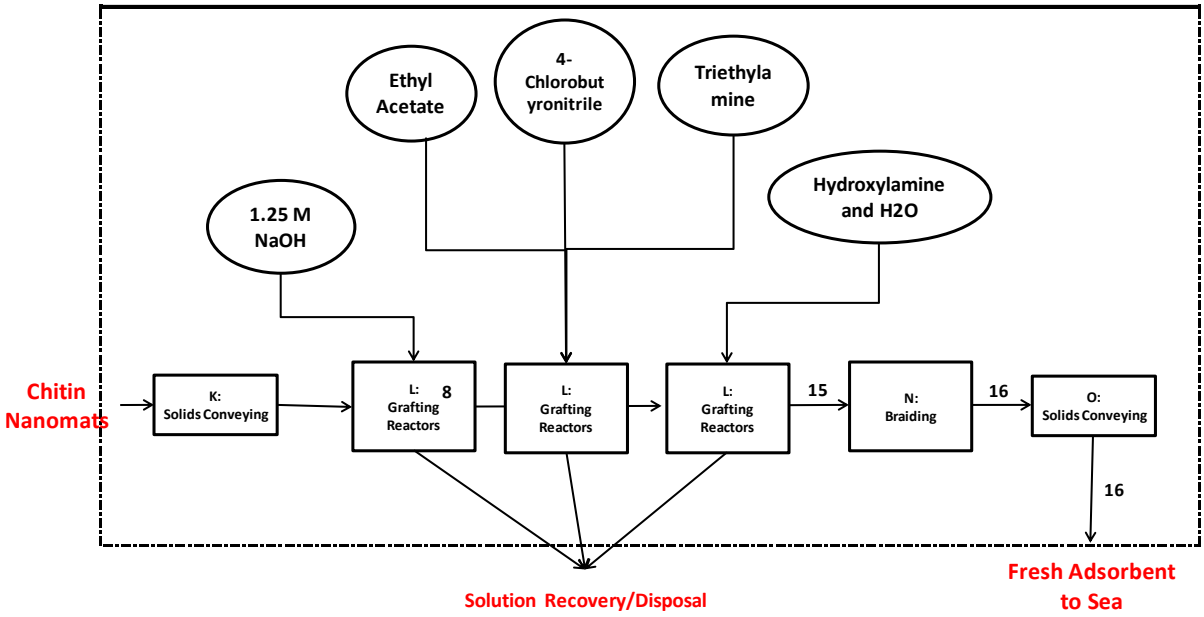

Figure 2.2 Chemical grafting process for University of Alabama fibers

Analogous to the reference ORNL adsorbents, stoichiometric relationships describing the grafting chemistry and the assumption of 100% efficiency were used to calculate chemical consumption rates and associated costs. Given the boutique-nature of some chemicals, the ionic liquid in particular, a market analysis was conducted to quantify the change in price, P resulting from increased demand for use in chitin nanomat production. The effect of economies of scale on growing global demand, M, ca be seen in eqn 1 and was used to arrive at the ionic liquid price seen in Table 2.1, along with all other chemical inputs.

$$P_2 = P_1 \left(\frac{M_2}{M_1}\right)^{0.6}$$

Table 2.1 Chemical requirements for chitin substrate

	Chemical	tonnes per tonne of chitin	Cost of chemical ($/tonne)
Chitin Substrate	Sodium Hydroxide	0.67	$480
	Ethyl acetate	21	$890
	Triethylamine	0.021	$2,000
	4-chlorobutyronitrile	0.029	$12,000
	Hydroxylamine	0.092	$1,500
	Ionic Liquid (1-ethyl-3-methylimidazolium acetate)	49	$360

These adsorbent production methods and associated input costs were used in conjunction with the same mooring and deployment and elution strategy used by the reference ORNL adsorbents so the resulting uranium production costs could be compared. Currently, the chitin based adsorbents have resulted in a low uranium uptake, on the order of micrograms per gram. To explore the economic viability of these adsorbents if the uptake were to be increased, the adsorption capacity is scaled up to 3.48 g U/kg ads to be of the same order of magnitude as the ORNL adsorbents. Using this uptake performance along with historical deployment parameters, notably a 5% degradation of adsorbent capacity upon recycle and a total of 6 adsorbent uses, the uranium production cost for these chitin based adsorbents is estimated to be about $3,000/kg U.

The breakdown of the uranium production cost by major process step can be seen in Figure 2.3. Much like other adsorbent varieties, adsorbent production is the most

expensive step in the lifecycle. Unique to this adsorbent however is the degree to which a single chemical, the ionic liquid contributes to the adsorbent production cost.

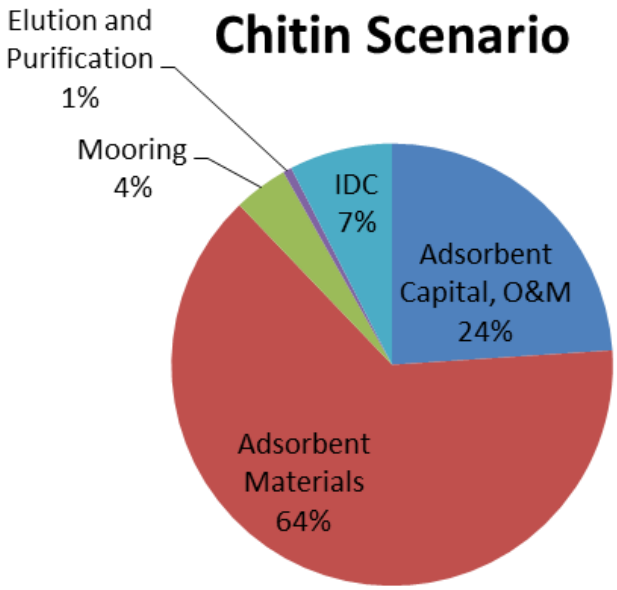

Figure 2.3 Cost breakdown by major process step for University of Alabama fibers

While the ionic liquid is not in itself prohibitively expensive, once adjusted for future economies of scale, the extremely large quantity required per unit mass of chitin drives up the cost significantly. This expense is exacerbated by the need to heat the large volume of ionic liquid, which is predominantly water, to high temperatures. Therefore it is worth exploring the cost savings that would result if the required mass of ionic liquid could be reduced via chemical recycle or if the weight percent of ionic liquid required to dissolve each unit mass of chitin were reduced. Figure 2.4 shows the effects of varying these two parameters.

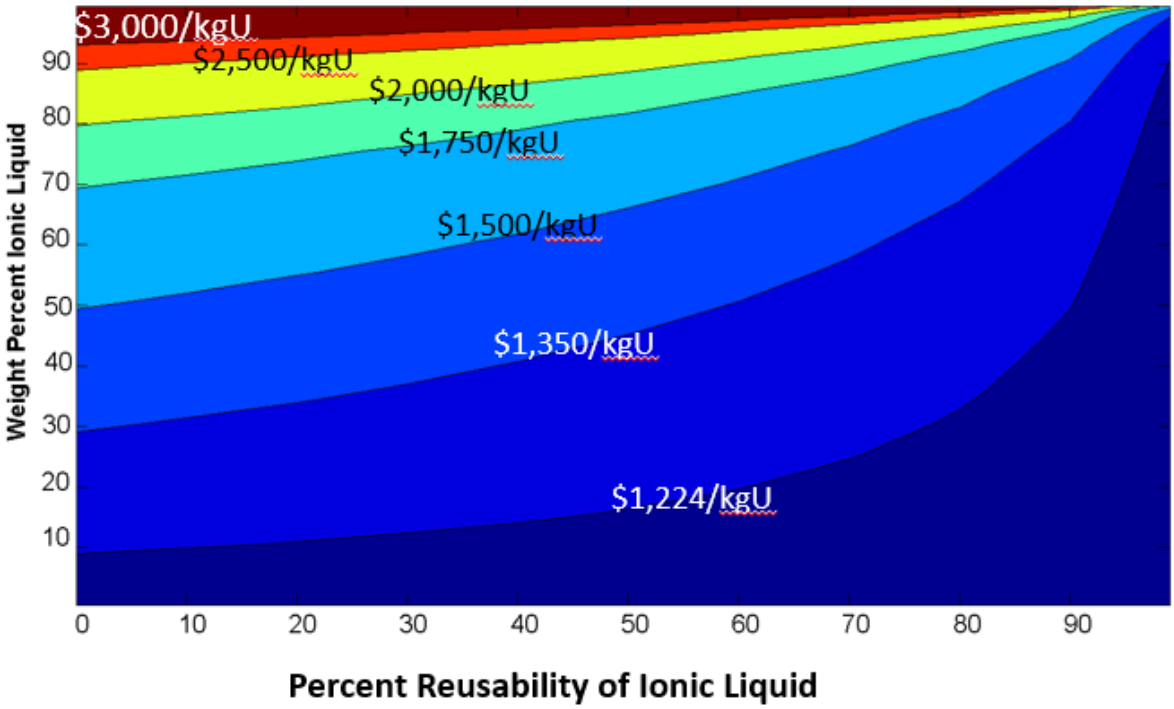

Figure 2.4 Uranium production cost as a function of ionic liquid consumption

Alternatively these sensitivities could be considered in terms of the adsorption capacity required to achieve a production cost similar to that of the ORNL adsorbents. Figure 2.5 shows the target capacities to achieve a uranium production cost of $640/kg ads as a function of ionic liquid recyclability and weight percent of ionic liquid in shell dissolving solution.

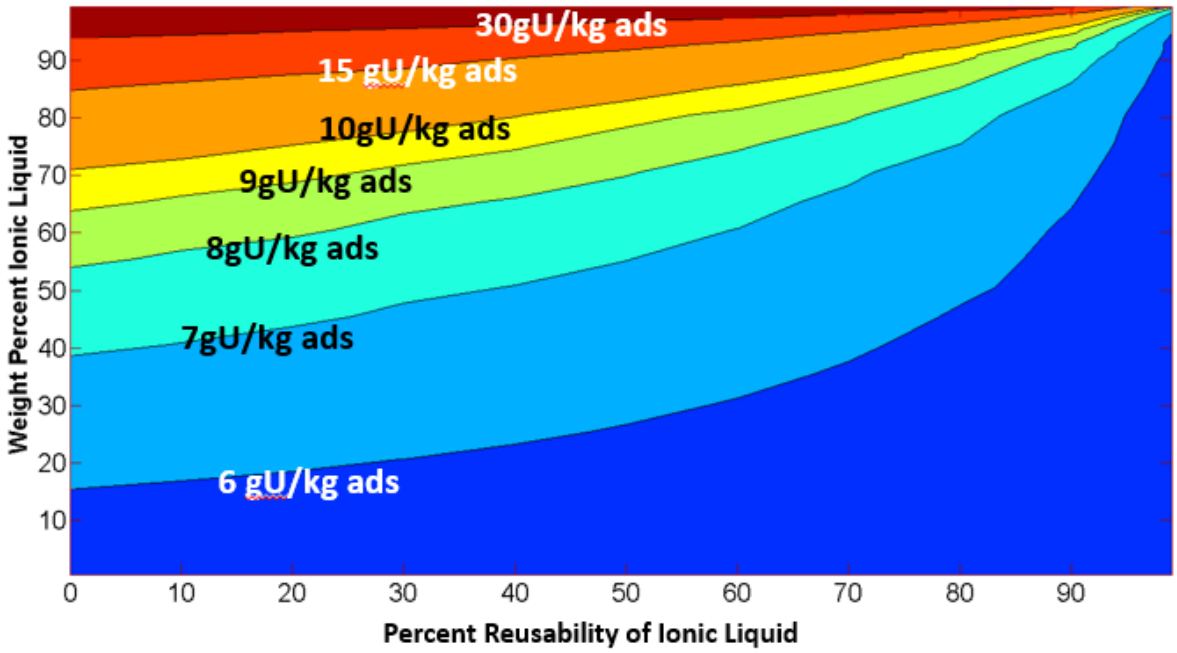

Figure 2.5 Breakeven required adsorbent uptake performance as a function of ionic liquid consumption

It is clear that in order to offer the significant environmental benefits of replacing plastic with an all-natural substrate, considerable improvements to the adsorbent production process or resulting performance must be achieved.

Task 3: University of Maryland

Recognizing the adsorbent production process as the most expensive step, researchers at the University of Maryland have created a novel adsorbent variety via a much simpler synthesis process. In addition to reducing equipment requirements, the Maryland team aimed to offer a "greener" production process by replacing organic solvents with simple water.

This particular adsorbent technology deviates quite significantly from the reference fibers with respect to the chemical composition of both the substrate and the

uranium chelating agent. The fibrous backbone is made of a high surface area Winged nylon fabric available for commercial purchase. An X-ray irradiation process is then used to graft the ligand, bis(2-methacryloxyethyl) phosphate (B2MP) onto the fibers in an aqueous environment. Sonication is then used to remove by-products of the irradiation process. This novel direct grafting procedure resulted in degrees of ligand grafting on the order of 100%, similar to that of the reference fibers. The process flow diagram for this simplified process can be seen in Figure 3.1.

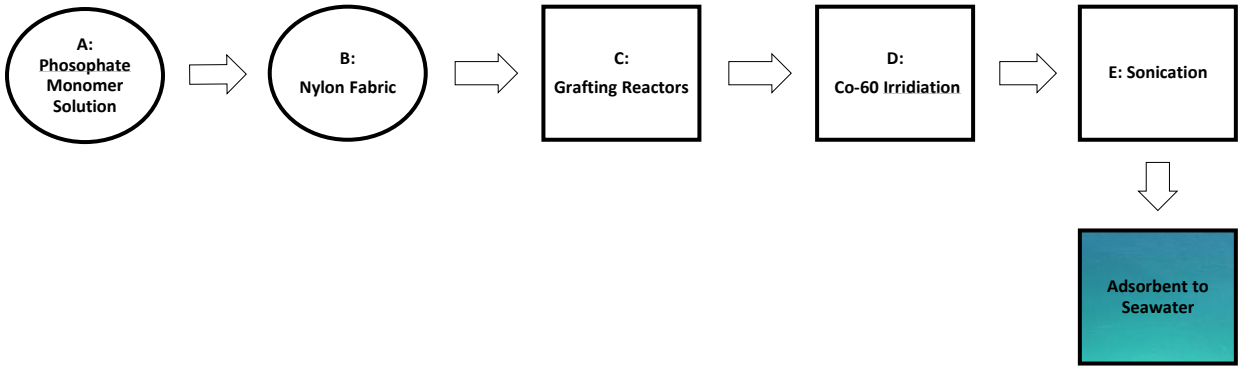

Figure 3.1 Adsorbent synthesis process for University of Maryland fibers

Further deviation from the reference adsorbents appears during the elution and recycle phases. Uranium captured by the nylon based adsorbents can be eluted off using saturated ammonium oxalate. This novel elution process was initially very economical for its ability to circumvent a costly alkaline regeneration procedure previously required by the reference adsorbents. Later developments however by the University of Idaho, which will be discussed in task 4 eliminated the need for alkaline regeneration. Enumeration of the required chemical inputs and associated prices for this adsorbent technology can be seen in Table 3.1

Table 3.1 Chemical requirements for University of Maryland fiber synthesis

Chemical	Input per Unit Finished Adsorbent (tonne/tonne)	Cost ($/tonne)
Nylon 6 Fiber	.50	$18,000
bis(2-methacryloxyethyl) phosphate (B2MP)	.49	$10,000
Ammonium Oxalate	1	$1,710

Given limited data regarding adsorbent performance in true marine conditions, the economic analysis considers adsorbent uptake capacity and kinetics similar to that of the reference fibers. Results of uptake experiments conducted using simulated seawater were notable in suggesting higher durability achieved by these adsorbents. Capacity losses on the order of 1% per re-use were observed and are thus used in the cost calculation in lieu of the 5% or higher degradation rate suffered by the reference fibers.

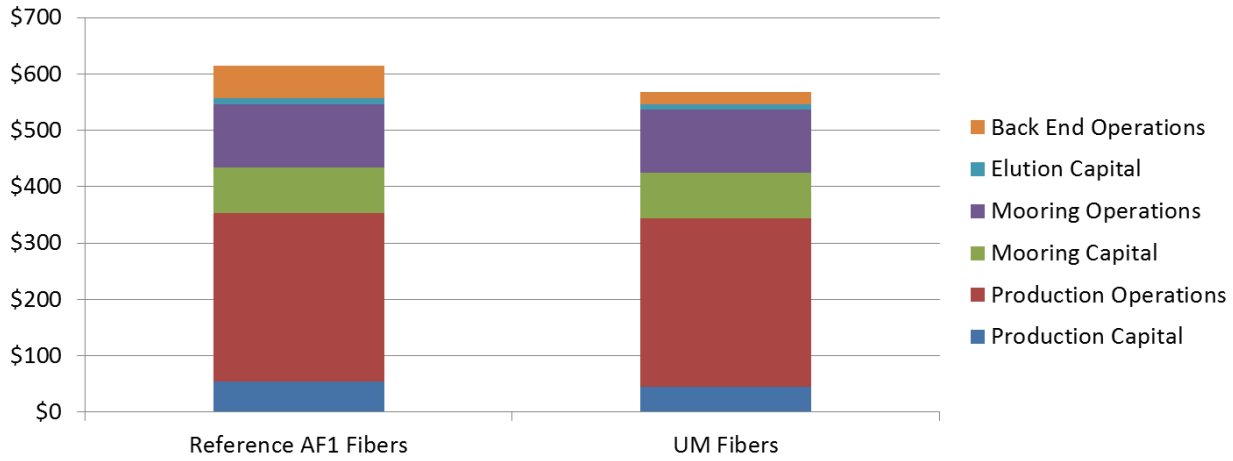

Figure 3.2 Cost breakdown for University of Maryland fibers as compared to ORNL fibers

The cost breakdown for these fibers as compared to that of the reference fibers can be seen in Figure 3.2. It is clear that these adsorbents can be considered a

competitive alternative to the amidoxime based fibers. In addition to reducing organic solvent consumption through the use of green chemistry, these adsorbents have the potential to offer a 10% cost savings if similar uptake performance can be achieved.

Task 4: University of Idaho

The acidic elution process had been identified as a significant cost contributor due to its effects on adsorbent durability and the requirement for subsequent adsorbent regeneration with an alkaline solution. Therefore we partnered with Dr. Chen Wai who replaced the acidic elution chemicals with mildly basic potassium bicarbonate. The main benefit of this replacement is removal of the costly re-conditioning step after each re-use of adsorbent. Although each individual regeneration is not prohibitively expensive, since the alkaline re-conditioning step is repeated multiple times over the adsorbent's lifetime and requires a high chemical input relative to adsorbent mass, this accumulates to a non-trivial cost.

Given that uranium occurs in seawater in the form of uranyl tricarbonate the novel elution process uses a high bicarbonate concentration to reverse the process according to the reaction pictured in Figure 4.1 The cost of the bicarbonate elution process is calculated analogously to the acidic elution process; chemical consumption values are calculated based on stoichiometry assuming 100% conversion efficiency. While there is no required alkaline regeneration, a hydroxide rinse is used in conjunction with this elution process. This is a notable difference as the NaOH is not consumed and it is therefore assumed to be reused with a 90% recovery rate.

$$UO_2(HA)A^- + 3HCO_3^- \rightarrow [UO_2(CO_3)_3]^{4-} + 2H_2A$$

Figure 4.1 Chemical reaction describing University of Idaho elution process

Experimental results have also shown that the bicarbonate elution process does not degrade adsorbent like the acidic process does, therefore reducing or potentially eliminating the drop in uptake with subsequent re-uses. This is significant not only in

increasing the lifetime recovery of uranium by a unit mass of adsorbent, but also allowing for a greater number of economically advantageous recycles.

The uranium production cost as a function of adsorbent uses for both the acidic and bicarbonate elution process can be seen in Figure 4.2. Both of the scenarios pictured assumes a 5% loss in uptake with each adsorbent reuse, although this may not accurately reflect the resulting performance of adsorbents eluted with the acidic process.

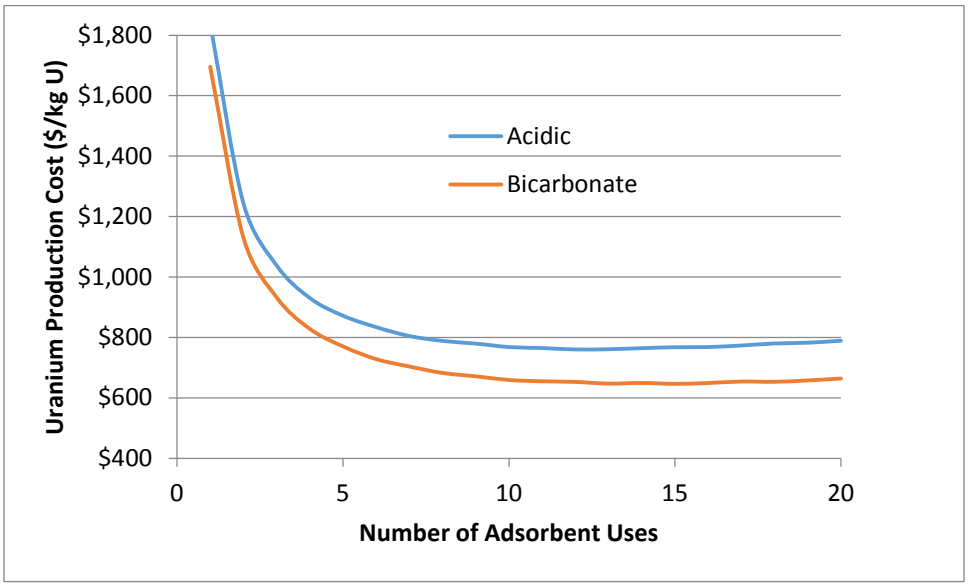

Figure 4.2 Uranium production cost for University of Idaho bicarbonate elution compared to reference acidic elution process

Later experimental results indicated however that adsorbent durability may be a function of not only uranium elution process but also length of soaking campaign and adsorbent recycle number. This relationship can be seen in Figure 4.3 where loss in uranium uptake is seen to be worse on the second reuse as compared to the first. All subsequent reuses are assumed to experience the same loss in uptake as the second adsorbent use.

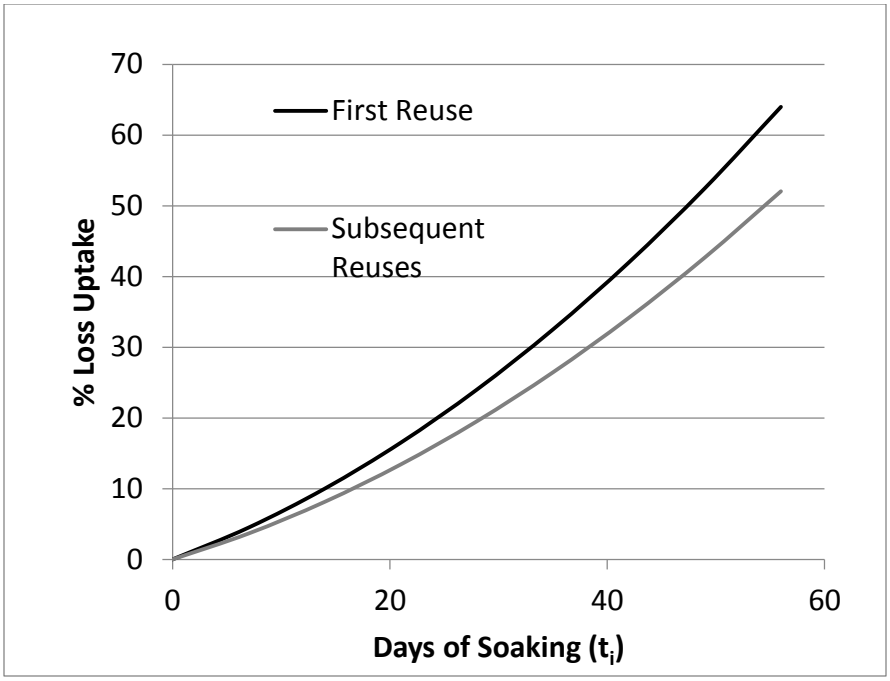

Figure 4.3 Degradation of adsorbent uptake upon reuse as a function of number of uses and days of soaking campaign

Both the adsorbent durability pictured here in Figure 4.2 and the constant 5% loss in uptake seen in previous experiments by Japanese researchers on chemically similar adsorbents are assumed to reflect realistic outcomes for adsorbent performance. Therefore these two scenarios are used to bound the best and worst case uranium production costs, in addition to the uncertainty surrounding the effects on uptake of marine biofouling as can be seen in Figure 4.4.

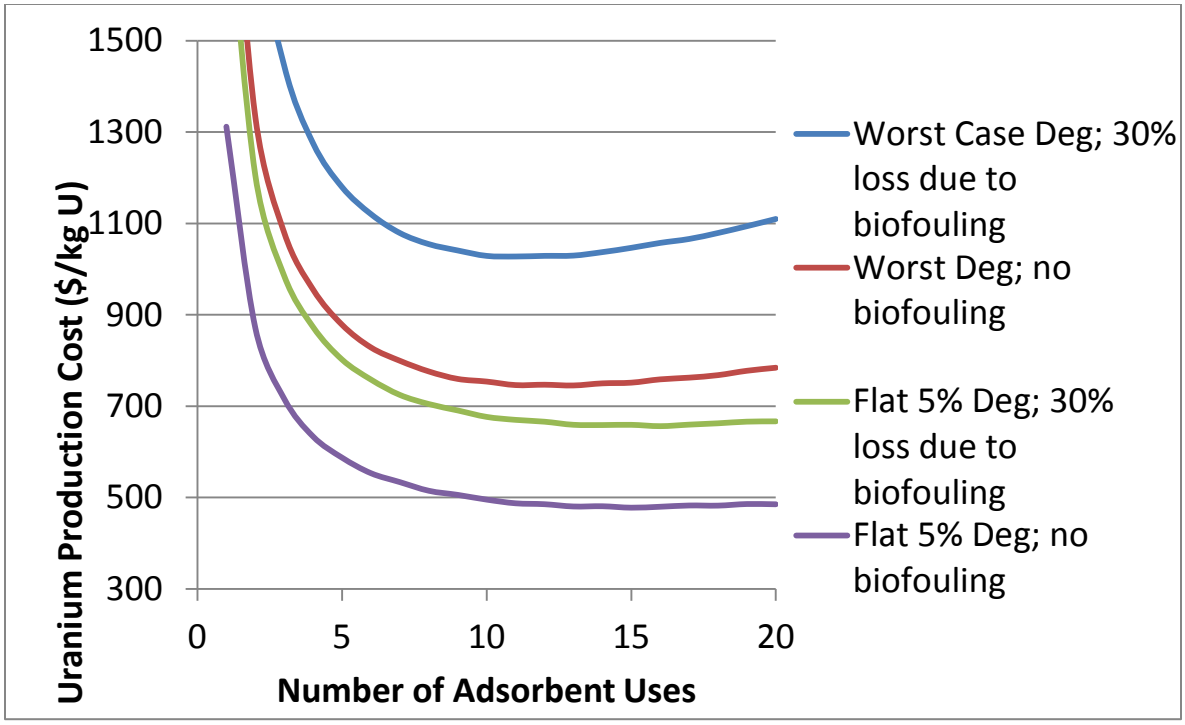

Figure 4.4 Uranium production cost range of reference AF1 adsorbents by ORNL showcasing the bounding effects of adsorbent degradation and marine biofouling

Task 5: Massachusetts Institute of Technology

Although the adsorbent fabrication makes the most significant contribution to the seawater uranium production cost, the mooring and deployment cost effectively establishes a cost floor by governing the maximum number of economical adsorbent recycles. Therefore, researchers at the Massachusetts Institute of Technology designed a symbiotic deployment structure where uranium adsorbing braids are moored on marine structures for wind based electricity generation.

The system first proposed by Picard et al. [4] allows for continuous uranium recovery by attaching a mobile adsorbent belt along with the necessary elution equipment to the base of off-shore wind turbines. The Wind and Uranium from Seawater Acquisition symBioitic Infrastructure can be seen in Figure 5.1, taken from the original Picard publication.

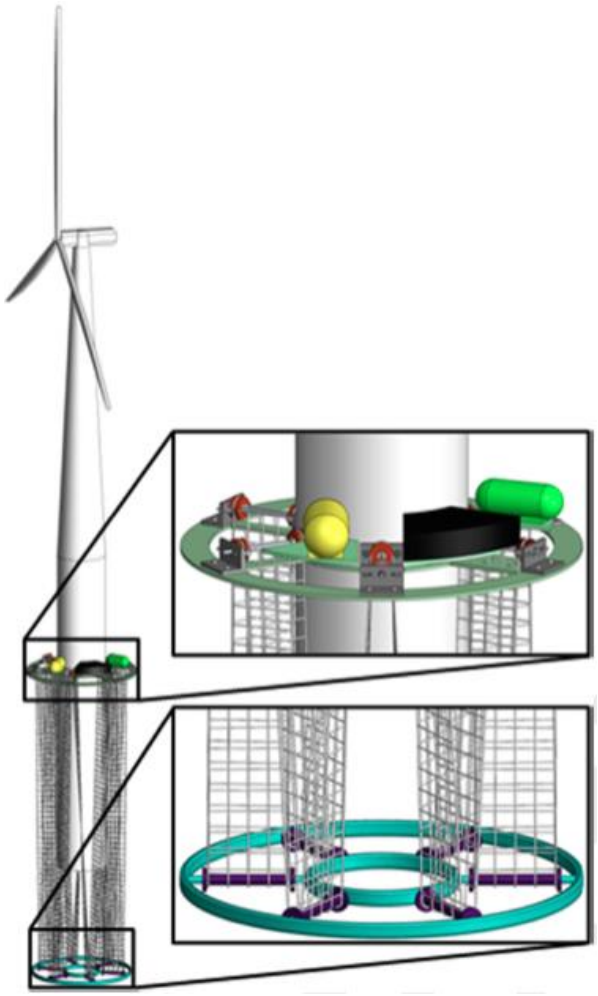

Figure 5.1 Depiction of the Wind and Uranium from Seawater Acquisition symBiotic Infrastructure (WUSABI) deployment platform, taken from [4]

This system was first analyzed for economic feasibility in the original publication. This zeroth order estimate was later expanded in an independent publication[5]. The higher fidelity cost analysis of the WUSABI system was significant in independently confirming the cost benefits of moving to this symbiotic system, especially when considered in light of some of the complexities and feedbacks left out of the initial cost

estimate. The uranium production cost for the WUSABI system as compared to the reference kelp-field deployment scheme as a function of number of adsorbent uses can be seen in Figure 5.2 as published in [5].

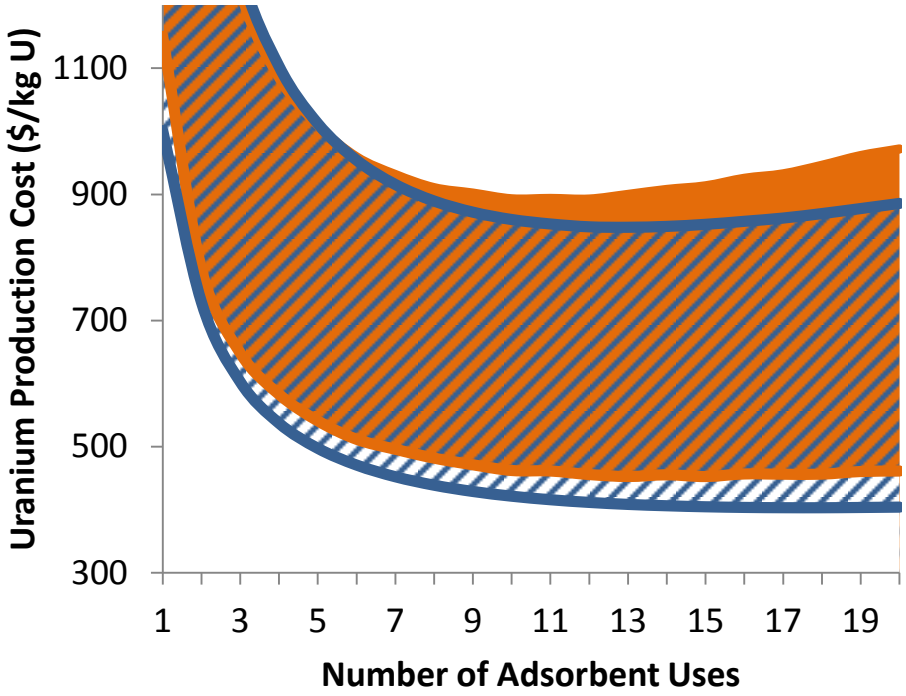

Figure 5.2 Uranium production cost as function of number of adsorbent uses for the WUSABI deployment scheme as compared to the reference kelp-field deployment scheme.

Perhaps more important, the application of methodologies used in cost analyses of other adsorbent technologies, including the reference ORNL adsorbents, allows for accurate comparisons even as technologies undergo continual developments. Incorporation of the WUSABI deployment scheme into the existing cost model also allowed for the conduction of sensitivity analyses in order to identify design parameters resulting in significant cost impacts. The previous publication determined that elution tanks and fleet of ships required to service them contribute a significant portion of the

mooring and deployment cost. Therefore work, in a publication pending submission, has aimed to alleviate some of these cost burdens by altering various design parameters.

In an attempt to further reduce the mooring and deployment cost the elution and chemical storage tank materials were changed from expensive 316 stainless steel, as suggested in the original design, to a cheaper, yet still robust, cross-linked polyethylene. Additionally the number of ships used to service the turbine field, to empty and refill chemical storage tanks, was optimized. The number of ships is an important factor in not only the capital and operating costs associated with the ships themselves, but also in sizing the chemical storage tanks. The contribution of these factors can be seen in reference to other mooring and deployment cost inputs for both schemes in Figure 5.3.

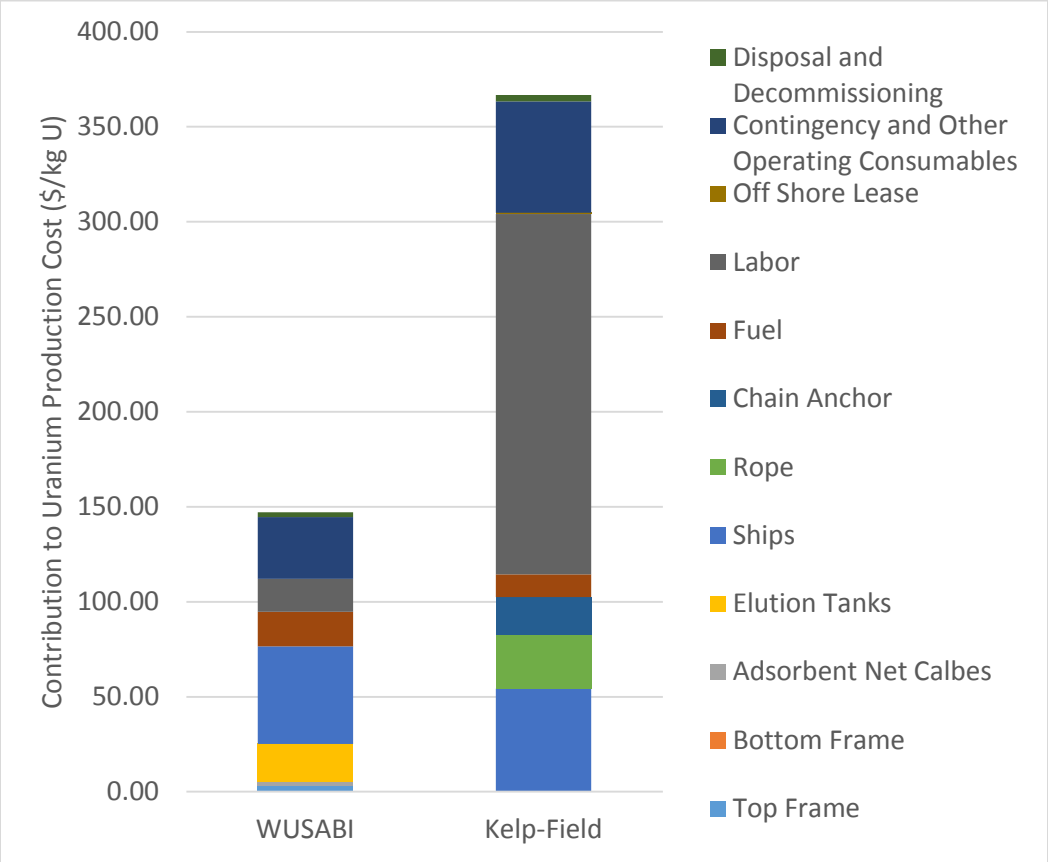

Figure 5.3 Breakdown of capital and operating cost for both deployment schemes

The uranium production cost resulting from the use of the WUSABI deployment scheme as compared to the reference kelp-field can be seen in Table 5.1 and Figure 5.4. An optimization framework established previously [6] was used to find the deployment parameters, specifically length of campaign and number of adsorbent uses, leading to the lowest uranium recovery cost by both schemes.

Table 5.1 Uranium production cost and optimal deployment parameters for both deployment schemes for the best and worst case adsorbent performance scenarios

		Kelp-field	WUSABI
Worst Case	Uses	11	20
	Days	15	12
	Cost per kg U	$870	$610
Best Case	Uses	14	20
	Days	41	60
	Cost per kg U	$430	$290

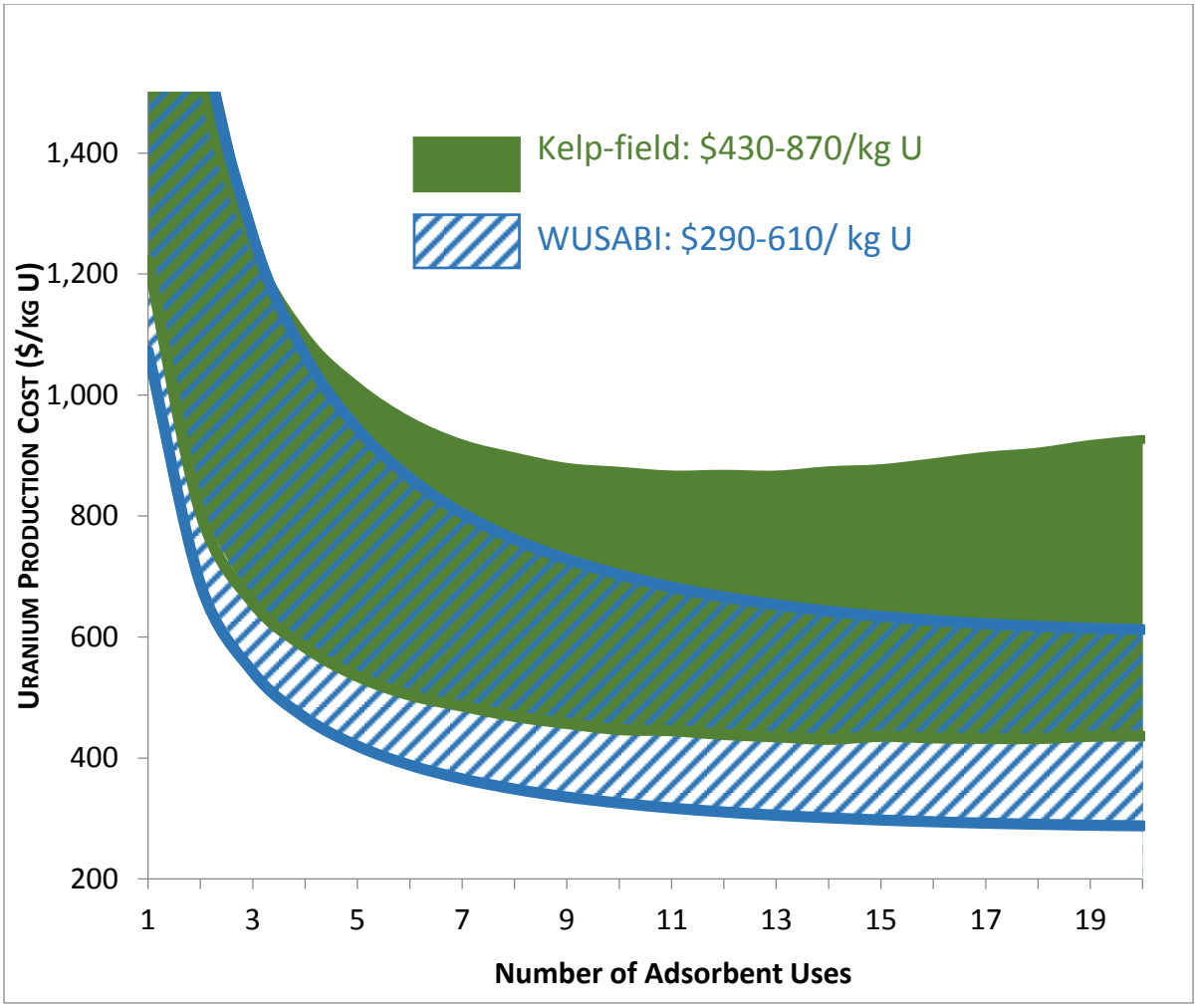

Figure 5.4 Uranium production cost for the best and worst case adsorbent performance parameters as a function of number of adsorbent uses for the improved WUSABI deployment scheme as compared to the reference scheme

It is clear that the symbiotic scheme is more economical. In addition to simply lowering the cost contribution from mooring and deployment, use of this system allows for a more favorable number of adsorbent recycles.

An additional noteworthy benefit of the WSUABI scheme is the ability to deploy adsorbents in shallower, often warmer, waters. Previous publications [7] [2]have shown

it is economically beneficial to deploy adsorbents in warmer waters due to the increase in uranium uptake by amidoxime at increased temperatures. The kelp-filed design in less apt to deploy in shallower waters due to the inclusion of a 40 meter clearance between the tops of the braids and the ocean surface to allow for ship traffic. In the WUSABI design however the pulley systems moves adsorbent belts up and down the height of the wind turbine platform, allowing adsorbents to experience a variety of temperatures.

The depth dependence of ocean temperature was incorporated to examine the sensitivity of uranium production cost to turbine height. In addition to allowing adsorbent to traverse varying ocean depths, and consequently water temperatures, trade-off exists structurally between a short turbine with a wide radius as compare to a taller narrower structure. A more detailed analysis of these feedbacks can be found in a manuscript currently under development. The resulting uranium production cost for two representative deployment locations can be seen for the best and worst case adsorbent performance scenarios.

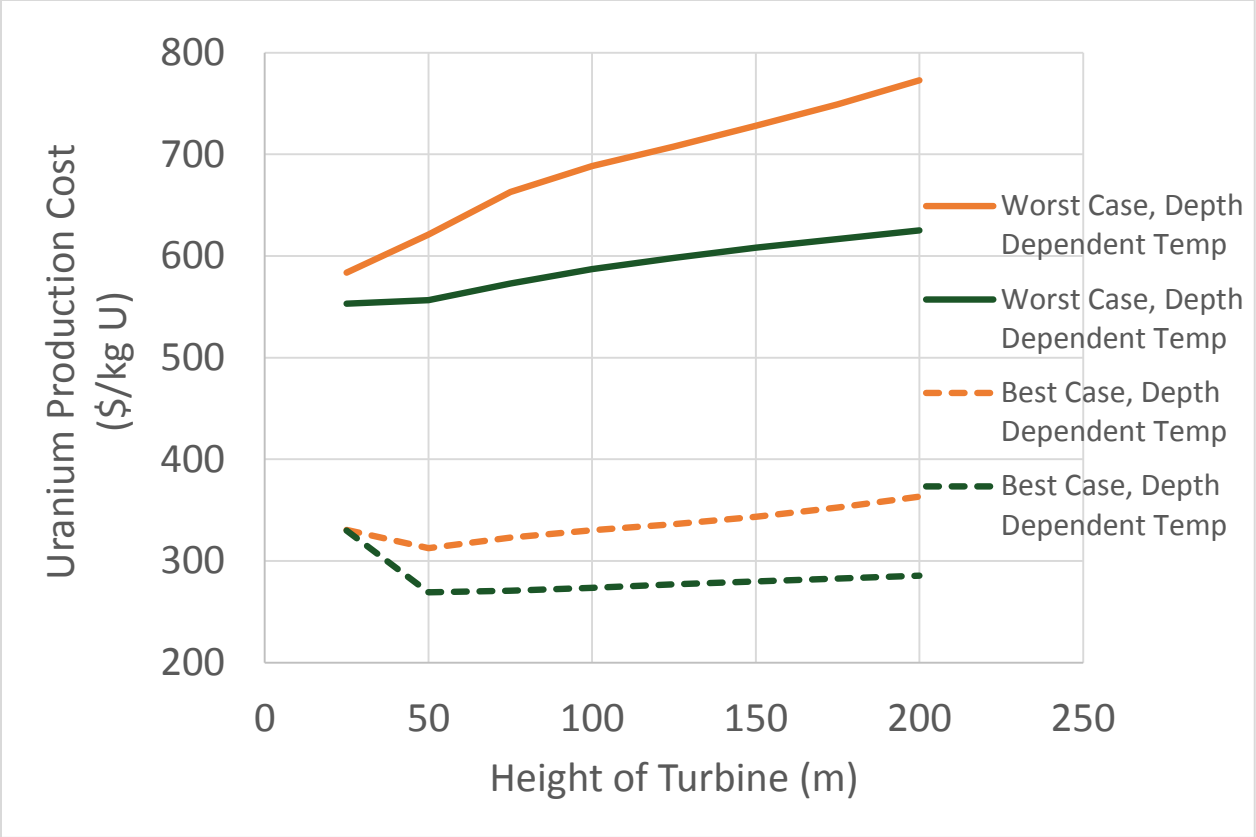

Figure 5.6 Uranium production cost as a function of turbine height for representative deployment locations (Orange represents New England and Green represents the Gulf of Mexico)

The WUSABI deployment scheme is unique from the other novel adsorbent technologies because it can be used in its current state to deliver a cost savings, while other technologies require additional data or process improvements. Beyond decreasing the uranium production cost, the WUSABI deployment schemes offers the benefit of a smaller environmental footprint, which is often cited as one of the motivating factors for pursing the recovery of uranium from seawater, given that utilized environment is already perturbed from its natural state due to the presence of the wind turbines.

Task 6: City University of New York

The uranium capacity of adsorbent fibers has been shown to be a significant driver of the final uranium production cost [8] [1]. Partners at Hunter College of the City University of New York (CUNY) have therefore attempted to increase capacity by enhancing the uranium affinity of the amidoxime ligand used in reference fibers [9]. In addition to amidoxime, these novel fibers contain a second amine ligand that offers two mechanisms of facilitating amidoxime-uranium interaction.

The adsorbent synthesis of the CUNY adsorbent differs from that of the reference scenario beginning with the fibrous backbone; commercially available polyacrylonitirle fiber is used in lieu of the high density polyethylene. Analogous to the reference adsorbents, acrylonitrile groups are converted to amidoxime using hydroxylamine hydrochloride. Additionally, a second ligand, Diethylenetriamine, is added with the hopes of enhancing the affinity of amidoxime for uranium. It is suspected that DETA can provide two possible mechanisms for increasing uranium uptake: direct action upon the existing amidoxime to increase ion exchange ability, or contribution of additional coordination sites for the uranyl ion. After functional group conversion the adsorbents are washed with ethylene glycol and sodium hydroxide before they are ready for marine deployment. The required chemical inputs and associated costs can be seen in Table 6.1

Table 6.1 Chemical consumption and cost for synthesis of University of New York fibers

Chemical	Input per Unit Finished Adsorbent (tonne/tonne)	Cost ($/tonne)
Polyacrylonitrile Fiber	0.44	$2,300
Diethylenetriamine	0.43	$2,900
Ethylene Glycol	1.8	$920

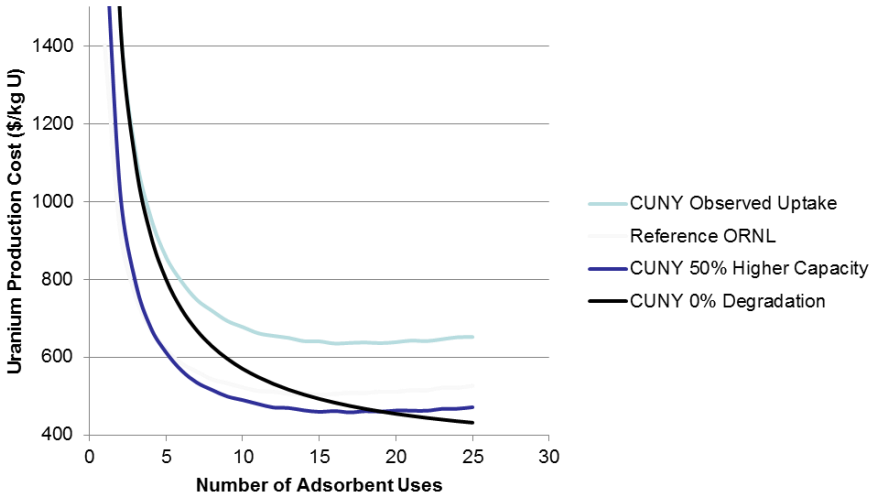

Figure 6.1 Uranium production costs as a function of number of adsorbent uses for CUNY fibers, subject to some sensitivity analyses

The cost to recover uranium using this adsorbent is calculated using the best and worst case adsorbent performance scenarios applied to other technologies and can be seen in Figure 6.1. Although initial experimental data has indicated these novel adsorbents have lower uranium uptake, our study shows that if the uranium capacity can be increased 50% above the levels observed a 16% savings could result. This sensitivity, along with other adsorbent performance parameters, is displayed in Figure 6.1.

Conclusions

The contribution of university partners has provided significant progress to the uranium from seawater program by providing a variety of alternative recovery technologies. While not all of the analyzed technologies are capable of decreasing the uranium recovery cost, all address their specific goal of targeting a particular weakness of the reference recovery system. If the recovery of uranium from seawater were to be adopted on a large scale, then some of the specified weaknesses could become increasingly important.

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