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Results of an Ocean Trial of the Symbiotic Machine for Ocean uRanium Extraction

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Abstract

Amidoxime-based adsorbents have become highly promising for seawater uranium extraction. However, current deployment schemes are stand-alone, intermittent operation systems that have significant practical and economic challenges. This paper presents two 1/10th scale prototypes of a Symbiotic Machine for Ocean uRanium Extraction (SMORE) that pairs with an existing offshore structure that reduces mooring and deployment costs while enabling continuous, autonomous uranium extraction. Utilizing a shell enclosure to decouple the mechanical and chemical requirements of the adsorbent, one design concept prototyped continuously moves the shells through the water while the other keeps them stationary. Water flow in the shells on each prototype was determined using the measurement of radium extracted onto MnO₂ impregnated acrylic fibers. The results from a nine-week ocean trial show that while movement of the shells through the water may not have an effect on uranium adsorption by the fibers
encased, it could help reduce biofouling if above a certain threshold speed (resulting in increased uptake), while also allowing for the incorporation of design elements to further mitigate biofouling such as bristle brushes and UV lamps. The trace metal uptake by the AI8 adsorbents in this trial also varied greatly from previous marine deployments, suggesting that uranium uptake may depend greatly upon the seawater concentrations of other elements such as vanadium and copper. This study’s findings will be used to determine the seawater uranium production cost from a full-scale SMORE system.

Introduction

With global conventional reserves of terrestrial uranium estimated to be depleted in a little over a century, extraction of uranium is expected to shift to lower quality sites, leading to higher extraction costs and greater environmental impacts. Fortunately, the ocean contains approximately 4.5 billion tonnes of uranium, nearly 500 times more than land and offer an alternative to land-based mining to meet nuclear fuel demand.

The system currently studied by a nation-wide consortium of national lab and university partners involves the passive recovery of uranium using polymer-based adsorbents. After initial marine deployment, the polymers are eluted to remove metal ions, including uranium. A number of elution cycles may be required before an alkali wash is used to regenerate the polymer, freeing its functional groups and allowing it to be redeployed in the ocean for reuse. The products undergo purification and precipitation, as for terrestrially mined uranium, to produce yellowcake.

Previous economic analyses have identified the adsorbent production and mooring as the most expensive components of the recovery process. Picard et al. designed a system which targeted cost reductions in the deployment, mooring, and recovery of the adsorbent by coupling the uranium harvester with an existing offshore structure, particularly an offshore wind turbine. A platform at the base of the 5 MW wind tower supports a belt of adsorbent that cycles through the seawater and through an elution plant located on the platform. The
system was sized to collect 1.2 tonnes of uranium per year, a sufficient amount to supply a 5 MW nuclear power plant. A recent independent cost-analysis compared this symbiotic deployment to a reference scheme in which the adsorbent polymer was braided into a buoyant net and deployed as a kelp-field across the ocean floor, serviced by boats for deployment, elution, and redeployment. It was found that the symbiotic deployment proposed by Picard et al. could reduce the seawater uranium production cost by up to 30%. 

Recent work, however, indicates that uranium-adsorbing materials with the optimal chemical properties for high adsorbent capacity have inherently low tensile strength and durability, suggesting that the adsorbent may not be strong enough to be woven into a belt in the design by Picard et al. Therefore, the Symbiotic Machine for Ocean uRanium Extraction (SMORE) was developed, utilizing shell enclosures to decouple the chemical and mechanical requirements of the machine. This paper discusses the results of an ocean trial of two SMORE prototypes. The designs tested are first presented, followed by a description of the experiment and test site. In addition to the mechanical testing of SMORE, uranium adsorbents were deployed and sampled, the procedure of which is also described in this paper. Furthermore, the trial included comprehensive ocean sensing, which is discussed in detail.

Symbiotic Machine for Ocean uRanium Extraction prototypes

Decoupling of the mechanical and chemical requirements of the offshore uranium harvesting machine was accomplished by a two part system, shown in Figure 1(a), comprised of a hard permeable outer shell that serves as the protective element for uranium adsorbent material in its interior. The outer shell has sufficient mechanical strength and durability for use in an offshore environment and chemical resilience against elution treatments, while the adsorbent material is designed to have high adsorbent capacity.

SMORE utilizes shell enclosures strung along high strength mooring rope, resembling
Figure 1: Design details of SMORE: (a) Hard permeable shell enclosure encapsulating the polymer adsorbent; (b) A 1/10th physical scale adsorbent ball-chain net as used in SMORE. White adsorbent enclosure shells were alternated with orange placeholder shells used for mechanical testing; (c) Side and (d) top views of SMORE, which uses rollers to move ball-chain lengths of adsorbent through the water column.

conventional ball-chain belts. The belts are strung together to create a net using cross-members which add rigidity and reduce the likelihood of tangling (Figure (b)). Large rollers are used to move the nets down the entire length of the turbine. Multiple subsystems are employed (Figures (c) and (d)) to achieve a higher device uptime given the lower probability that unforeseen circumstances or complications will cause simultaneous failure of all subsystems. Each subsystem is comprised of an adsorbent ball-chain net, rollers to actuate the net, and tanks, into which the ball-chain net can be rolled for the elution and
regeneration processes.

Two designs of SMORE were prototyped at a 1/10th physical scale for prolonged ocean testing (Figure 2(a)) to investigate if movement of the shells through the water column, inducing more seawater flow to the fiber adsorbents encased, would increase the uranium adsorbed. Previous work observed that flow velocities of > $5.52 \text{ cm/s}$ minimize mass-transfer resistances and maximize adsorbent capacities. These velocities occurred frequently at the ocean test site, suggesting that the shell net movement may not be worthwhile given the limited value of additional uranium adsorbed and the increased harvester cost due to its heightened complexity.

Haji et al. describes the design, fabrication, and assembly of the stationary and continuous systems. Aside from the bottom support, net, and motor assembly, the continuous system was analogous to the stationary system. In the case of the continuous system, the

![Diagram](image)

**Figure 2:** (a) Three-dimensional model of 1/10th physical scale prototypes for ocean testing of the SMORE design. Both a stationary and continuous version of the design were fabricated and mounted to a wooden float for ocean testing. (b) Layout of instruments used for measuring physical quantities at the ocean site. The instruments were attached to a piling at the end of the dock near the SMORE prototypes.
shell enclosure net moved in a complete loop at a rate of approximately 12 cm/s.

**Shell enclosure net**

For both of the prototypes, four lengths of shells were combined to make a single net. The majority of the shells did not contain uranium adsorbing fibers and were used primarily to test the mechanical components of the system. The stationary system comprised 508 shells and 852 shells were used for the continuous system net.

The effects of two shell enclosure designs on adsorbent uranium uptake were tested in this ocean trial (Figures 3(a) and (b)). Each net included nine of each type of design (Figure 1(b)). On the stationary net, the shells with the adsorbent fiber were placed about mid-depth in the water column. Adsorbents were also placed in two mesh bags on the stationary net to serve as controls. Results of recent flume studies suggests that the shell enclosure designs would have no effect on the encased fiber’s uranium uptake.\[^{18}\]

**Adsorbent deployment and sampling**

The prototypes utilized the AI8 adsorbent developed at Oak Ridge National Laboratory (ORNL) (renamed from the AI11 described in Das et al.\[^{19}\]). The uranium uptake, $y$, after a certain exposure time in days, $t$, can be predicted using the one-site ligand-saturation model,

(a)  (b)  (c)

Figure 3: Shell designs for the testing of uranium adsorption fibers using (a) slotted holes and (b) circular holes. (c) Pre-weighed adsorbent mini braid.
\[ y = \frac{\beta_{\text{max}} t}{K_D + t}, \]  
(1)

where \( \beta_{\text{max}} \) is the saturation capacity in g U/kg adsorbent, and \( K_D \) is the half-saturation time in days.\(^{[9]}\)

Each enclosure contained “mini braids”, pre-weighed, small masses (80-100 mg) of adsorbent fiber, were cut from a common braid prepared by ORNL (Figure 3(c)). The weight before deployment allowed for the determination of adsorption capacity as a function of the adsorbent mass. Weight after retrieval included biofouling (the growth of organisms on the fiber), however it did not account for any loss of fiber incurred during the deployment. To determine uranium adsorption as a function of time, samples were collected at 24 hours and subsequently every seven days after deployment for 56 days. Water samples were also collected for trace metal analysis.

**Ocean test measurements**

The prototypes were mounted to a wooden float at the Massachusetts Maritime Academy (MMA) in Buzzards Bay, MA in a low-tide water depth of \( \sim 7 \) m. Although fairly close to shore, the tides varied up to 1.8 m and the wind generated waves up to 0.9 m high. Additionally, the flow velocity could be extremely strong due to proximity to the Cape Cod Canal, which has currents of up to 2.6 m/s at peak tidal ebb and flow.

Physical ocean water properties were monitored between August 10, 2016 and December 18, 2016. Sensors measuring current velocity, temperature, conductivity, and light intensity were deployed on a nearby stationary piling (Figure 2(b)).

Temperature has been shown to have a significant impact on the uranium uptake by the adsorbent where decreasing temperature reduces uptake.\(^{[20][22]}\) For the A18 fiber, (2) governs

\[ y \]  
the uptake of uranium (g U/kg adsorbent).\(^{[21][23][24]}\)
\[ y = \frac{(0.323T - 1.253)t}{(0.111T + 19.673) + t}. \]  

(2)

By (2), a temperature difference of 5°C can result in a 50% change in uranium uptake.\(^{21-23}\)  

Temperature was measured at the piling and inside the shell enclosures using ONSET Tidbit Water temperature loggers.

Salinity directly indicates the amount of uranium present in the seawater by a well-defined relationship to \(^{238}\)U concentration.\(^{23}\) A Xylem EXO-2 Sonde collected salinity measurements from October 4, 2016 to December 13, 2016.

Light is observed to drastically affect biofouling, the growth of marine organisms, on the adsorbent, impeding uranium uptake by as much as 30%.\(^{23}\) To gather quantifiable data related to biofouling, HOBO Pendant Light/Temperature Loggers measured light at three depths on the piling.

Lastly, previous work has shown that the uranium adsorbed by the adsorbent fibers is dependent on the water flow rate for velocities less than 5.52 cm/s.\(^{17}\) For this reason, current was measured at three depths using Tilt Current Meters from Lowell Instruments.

**Water flow measurement**

One of the experimental objectives was to determine if increased water flow could be achieved by continuously moving the shell enclosures through the ocean and if that translated to an increase in uranium uptake of the fibers the shells encased. Following the method described in,\(^{15}\) the collection and measurement of radium extracted onto MnO\(_2\) impregnated acrylic fibers was utilized to quantify the volume of water passing through each of the different types of enclosures, including the control mesh bags, and the two shell designs on each of the two prototypes.
Results

The prototypes were deployed for a total of 56 days, from October 18, 2016 to December 13, 2016. The results described include comparison in the biofouling, water flow rate, and uranium uptake between the stationary and moving systems, as well as the physical properties and temporal changes the seawater at the test site.

Sensor data

Sensors monitored salinity, light, temperature, and currents for the majority of the prototype deployment. As expected, the light intensity dropped off significantly with depth (Figure 4(a)) and is most pronounced in the beginning of September with a difference of about 88% between the upper and lower light sensors. The difference is least pronounced in December with only about 59% disparity between the upper and lower light sensors.

Similar seasonal variations were seen in the temperature data (Figure 4(b)). The short term temperature differences were linked to the tidal and diurnal cycles. Given that the incremental adsorption of uranium decreases over time, it is likely that the colder temperatures, which occurred toward the end of the deployment, had minimal impact.

The salinity of the ocean test site also varied with tides (4(c)). Overall, the salinity during the experiment averaged 31.65 ± 0.15 psu, indicating an average 238U concentration by of 2.84 ± 0.076 ppb.

Current meter data does not span the entirety of the deployment due to premature battery failure and programming issues. As seen in Figure 4(d), while there were large short term variations in the currents (due to tides), overall little seasonal change was observed. In general, the currents peaked at approximately 5 cm/s, with stronger periods of over 10 cm/s. The currents measured are at times much larger than those used to in lab testing of adsorbents which have been tested in linear velocities up to 8.24 cm/s.[7]
Figure 4: (a) Light intensity as measured by the top, middle, and bottom light sensors. (b) Temperature as measured from the U24 conductivity logger. (c) Salinity as measured from the Xylem EXO-2 Sonde salinity meter with dashed lines indicating $\pm 2\hat{\sigma}$ where $\hat{\sigma}$ is the robust standard deviation. (d) Current as measured from the bottom current meter. The gray rectangle indicates the period of the ocean test, October 18, 2016 - December 13, 2016.

**Biofouling**

One striking result between the two systems was the difference in biofouling: at the end of the ocean trial, the stationary system’s shells were significantly more fouled than those on the continuous (Figure 5(a) and (b)).

The discrepancy may have been due to the rubbing of the continuously shells against various structural elements on the continuous system. Rubbing could have removed any growth that had already accumulated on the shells and inhibited further biofouling. Hence, future SMORE designs could incorporate bristle brushes to clean the shells as they pass,
Figure 5: Biofouling on the (a) stationary net and (b) continuously moving net at the end of the ocean test. (c) Percent weight gain or loss in the adsorbent fibers before and after deployment at each sampling.

reducing chances of organism growth. Additionally, adding UV LEDs to a point in the adsorbent net’s path could also prevent the formation of biofilm since UV light has been shown to have strong antibacterial properties.\(^{27}\)

The weight of the fibers after deployment, which account for any organism growth, was compared to the weight before deployment to determine if the reduced biofouling on the continuous system’s shells translated to reduced biofouling of the encased adsorbent fibers. The results (Figure 5(c)) show that for all enclosures and systems there was a trend toward weight loss in the beginning of the deployment and weight gain by the end of the deployment. The initial weight loss was likely due fibers shedding off in the harsh marine environment. There was little variation between the weight gained or lost between the different designs or systems. This may have been due to the fact that the continuous system was only moving for 37\% of the deployment, the majority of which occurred at the end of the trial. Given that biofouling begins within days of submergence in seawater, the lack of initial movement of the continuous system may have resulted in similar organism colonization on both systems. Additionally, the continuous system’s movement occurred during the last two-thirds of the
ocean trial, when the colder water temperature likely inhibited biofouling for all fibers.

There exist critical values of current speeds for different species of marine organisms above which fouling biomass is greatly reduced. In general, fouling is not possible at speeds greater than 150 cm/s, speeds that would likely damage the adsorbent. Future work should aim to determine adsorbent damage as a function of flow speed and to determine biofouling as a function of immersion time. The results of these studies can be used to determine the speed of movement of the adsorbent net as well as the frequency of other biofouling mitigation. The elution bath may also kill all organisms and hence reduce biofouling, a result that could also be used to determine the frequency of elution.

**Water flow rate**

Figure 6 shows the results of the water flow in the shells after correcting for the differences in the amount of radium fibers initially placed in each of the enclosures and adjusting for weight due to ash loss. The results indicate that there was a significant difference in the water passing within the enclosures on the different systems. In particular, the shells on the

![Figure 6: Total volume of seawater to come in contact with MnO$_2$ impregnated acrylic fibers in different enclosure types on different prototype systems as determined by $^{226}$Ra count using $\gamma$-spectrometry.](image-url)
continuous system had the most water flow, about 57% more water than the shells on the
stationary system and 35% more water than the nylon mesh bags.

However, as indicated by the error bars in Figure 6, there was no statistically significant
difference between the amount of water passing through shell designs on the same system,
suggesting that the design of the shell enclosure has little effect on the amount of water
reaching the interior. On the other hand, the difference in water flow to the shell enclosures
and the mesh bags on the stationary system varied drastically. Though neither was moving,
the bags had 33.8% more water flow than the stationary shells. Results from a recirculating
flume experiment showed that a statistically significant difference between the water flow to
six shell designs and a control in a recirculating flume did not affect the uranium adsorbed,\textsuperscript{18}
suggesting the uranium uptake of the ocean prototypes will be similarly unaffected by the
differences in water flow.

**Uranium uptake**

To correct for the varying salinity of natural seawater observed in different adsorption ex-
periments, all uranium adsorption capacity data was normalized to a salinity of 35 psu given
the conservative behavior of uranium in seawater.\textsuperscript{29} Fitting time-dependent measurements
of adsorption capacity using the one-site ligand saturation model allowed for the determina-
tion of adsorption kinetics and saturation capacity. As seen in Figure 7, uranium is not the
dominant metal adsorbed by the fiber.

Figure 8 shows the uranium adsorption (g U/kg adsorbent) of the AI8 fibers in all the
enclosures, with the lines indicating the one-site ligand saturation model fits for each en-
closure, along with the theoretical adsorption as predicted by \textsuperscript{2} for temperatures during
the ocean trial. Using a model detailed in Byers et al.\textsuperscript{9}, total uptake was modeled as the
incremental change in uptake over each day for each day’s average temperature. For cases
where the uptake at the current temperature exceeded the saturation capacity of the subse-
quent temperature, zero uptake was assumed due to the lack of empirical data. The fibers
deployed only adsorbed $\sim 1\ \text{g U/kg adsorbent}$ whereas the model predicted adsorption of over $2\ \text{g U/kg adsorbent}$. The predicted uptake comes closer to measurements when including the effects of biofouling using a 30% loss in total uptake\(^{26}\) (green dashed curve in Figure 8) but still does not accurately predict the uranium adsorbed.

The remaining discrepancy between the measured data and predicted uptake may be due to the other metal ions present in the water, specifically vanadium and copper. Vanadium exists at higher molar concentrations than uranium in seawater and has been observed to
Figure 8: Measurements of uranium adsorption (g U/kg adsorbent) for the AI8 adsorbent braids enclosed by the different shell designs on the two different systems, and enclosed only by a mesh bag (control). The uranium adsorption was normalized to a salinity of 35 psu. Curves drawn through the data represent fitting to a one-site ligand model by (1). The predicted uptake modeled by (2) using the temperature collected during the prototype deployment is shown in purple. The dashed green line shows the predicted uptake with a loss of 30% due to biofouling (as suggested by 26).

out-compete other ions, including uranium, from adsorption, though binding remains unclear. 30,31 Compared to marine deployments of the AI8 adsorbents at Broad Key Island, FL (BKI) (summarized in Table 1), while the vanadium concentration did not differ considerably between the deployments, the average V:U mass ratio adsorbed by the fibers in this study was almost 2.7 times higher than observed at BKI. 32 This may be due to the fact that vanadium has been found to load the adsorbent at much higher rates at colder temperatures, with vanadium saturation capacity being almost 14 times higher than the uranium saturation capacity for 8°C and only about three times higher for 31°C. 24 Given that the average temperature at the ocean site during adsorbent deployment was 10.9°C, as compared to 26.6°C at BKI, it is likely that the adsorbent saturated with vanadium, impeding uranium adsorption.

Marine deployment studies conducted at Woods Hole Oceanographic Institute suggest
that copper also greatly impedes uranium adsorption by as much as 54% \cite{33}. The copper concentration during this ocean trial averaged almost twice that measured during the BKI deployment and correlated to a five fold difference in average Cu:U mass ratio adsorbed by the fibers \cite{32}. These differences from previous marine deployments suggests that seawater concentrations of other ions may greatly impact uranium uptake and should be investigated further. There is no significant difference between the coefficients of the fits for each enclosure and system type \cite{34}, suggesting no significant difference in the uranium uptake by the adsorbent in different enclosures and on different systems. This indicates that the system movement, though increasing water flow to the adsorbent and decreasing biofouling on the shells, may not have increased the adsorbent uptake of uranium. These results agree with those reported by \cite{18} in which a temperature-controlled, recirculating flume study of six shell designs found no significant difference in uranium adsorbed by fibers enclosed, despite drastic differences in water flow rate between designs. The results are also in agreement with the suggestion by \cite{17} that for flow rates of > 5.52 cm/s the uptake of the adsorbent will no longer increase with increasing velocity.

Table 1: Mean and standard deviation in ambient seawater of vanadium (V, µg/L), V:U mass ratio, Mean and standard deviation in ambient seawater of copper (Cu, µg/L) and Cu:U mass ratio for the AI8 marine deployment summarized in this paper and reported for Broad Key Island (BKI) \cite{32}.

| Deployment        | V      | V:U    | Cu     | Cu:U   |
|-------------------|--------|--------|--------|--------|
| MMA (this paper)  | 1.78 ± 0.08 | 4.11 ± 0.25† | 0.61 ± 0.17 | 0.12 ± 0.01† |
| BKI               | 1.69 ± 0.15 | 1.55 ± 0.28‡ | 0.33 ± 0.27 | 0.02 ± 0.003‡ |

† Based on replicate determinations of the 56-day exposure time point for the mesh bag enclosure. Where more than two determinations were made the mean and standard deviation is reported.
‡ Based on replicate determinations of the 56-day exposure time point. Where more than two determinations were made the mean and standard deviation is reported.
Table 2: Uranium 56-day adsorption loadings (56-d, g U/kg adsorbent), saturation capacities ($\beta_{\text{max}}$, g U/kg adsorbent) and half-saturation time ($K_D$, days) for the AI8 adsorbents enclosed different shell designs on the two different, and enclosed only by a mesh bag (control). Adsorption loadings were normalized to a salinity of 35 psu.

| Prototype, Enclosure | 56-d$^\dagger$ | $\beta_{\text{max}}^\dagger$ | $K_D^\dagger$ |
|----------------------|----------------|-----------------|--------------|
| Stationary, Design 1 | 1.21 ± 2.04 | 2.17 ± 1.75 | 44.71 ± 66.18 |
| Stationary, Design 2 | 1.08 ± 0.76 | 1.84 ± 0.59 | 38.86 ± 24.11 |
| Continuous, Design 1 | 1.05 ± 0.39 | 1.35 ± 0.16 | 15.86 ± 5.57 |
| Continuous, Design 2 | 1.08 ± 0.57 | 1.61 ± 0.34 | 27.16 ± 13.15 |
| Mesh bag (control)   | 1.22 ± 0.33 | 1.56 ± 0.12 | 15.72 ± 4.04 |

$^\dagger$ Estimated using the one-site ligand saturation model [1].

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