

Development of a Method for Radioactive Nickel Removal

Dae Hyuk Jang^{a, c}, Sun Young Lee^{a, c}, Hyuncheol Kim^b and Miyong Yun^{a, c, *}

^aLab of Functional Aptamer, Department of Bioindustry and Bioresource Engineering, College of Life Sciences, Sejong University, Seoul, South Korea; ^bEnvironmental Radioactivity Assessment Team, Korea Atomic Energy Research Institute, Daejeon, South Korea; ^cResource Upcycling and Discovery Research Institute, Sejong University, Seoul, South Korea

*Corresponding author: myyun91@sejong.ac.kr

1. Introduction

The development of technology to remove radioactive particles from liquid nuclear waste is essential for maintaining nuclear power plants. Nickel is one of radioactive molecules in liquid nuclear waste. In this study, an aptamer able to trap small molecules such as metal ions was used to capture nickel in solutions. We describe the identification of DNA aptamers for nickel by classical in vitro selection and comparison of the removal rates of radioactive and non-radioactive nickel ions by aptamers. The development of a nickel-binding aptamer provides a new possibility of reducing the presence of radioactive materials in wastewater generated during operation of nuclear power plants.

2. Methods and Results

2.1 Selection for nickel-specific aptamer by SELEX

Nickel-specific aptamer selection based on target-immobilized SELEX [1, 2] was used in this study. Briefly, a single-stranded DNA library containing 40 random nucleotides in addition to two flanking sequences was prepared and loaded onto a column filled with activated Ni-NTA resin. A portion of the ssDNA library that strongly interacted with nickel was enriched in the last SELEX round. To reduce accumulation of unwanted ssDNA sequences, a negative selection step during round 6 was performed to remove oligonucleotides that bound NTA resin regardless of the metal ion bound. The progress of SELEX was monitored by measuring and comparing the concentration of eluted ssDNA from each round. Since a high concentration of eluted ssDNA pool was observed in round 8, this DNA was used for cloning and sequencing to identify aptamer candidates (Table 1).

Table 1. Sequence and identification of selected aptamers for nickel

Aptamer name	Aptamer sequence	Random sequence size	KD value (nM)	ΔG value (kcal/mol)
FA-N1	TAGAAGCCCCAGTGCATTCCCCTCCGCTCGACCCCGCATG	40	6.9 ± 9.5	-9.45
FA-N2	TCCGCTGCGGTCCGACGATACGCTCGGCTCTGTAGTATCG	40	12.3 ± 8.5	-10.74
FA-N3	TCCGGTCACGGAGGTAGACTACTGTCGTTGGCGGTGGTCG	40	13.3 ± 11.9	-12.97

2.2 Determination of most effective aptamer against of nickel

The binding affinity between nickel and the selected aptamer candidates was measured by surface plasmon

resonance (SPR) to determine the KD values (Fig. 2A). Since it is known that aptamer properties are affected not only by sequence but also by structure, we predicted the structure of the aptamers using the Zuker's algorithm, Mfold [3]. In the structures of the three aptamers shown in Figure 2B, we expect nickel binding to occur either through the hairpin structure itself (base 40) or through the interaction with nearby base pairs.

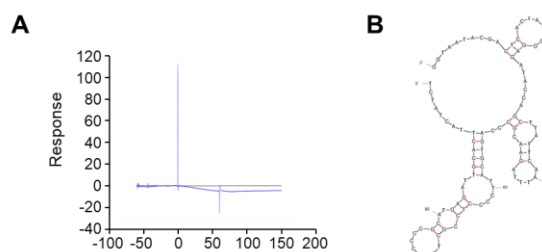


Fig. 1. Selection of aptamers with high affinity for nickel ion. (A) Measurement of Surface plasmon resonance (SPR) measurements for the selected aptamer. (B) Predicted secondary structure model of the selected aptamer

2.3 Elimination of radioactive Nickel from aqueous solution using the nickel binding aptamer

To determine how much nickel remained after the interaction with the aptamer-bead complex, we passed nickel solution through the column, collected the eluate and calculated the removal rate (%) by dividing nickel concentration in the eluate by its initial concentration. After injecting 10 μ M nickel into the column containing the aptamer-bead complex, the flow was stopped for 10 min to allow sufficient for binding. The amount of nickel remaining bound to the aptamer was about 80% (Fig. 2). The removal rate of both isotopes (Ni-59 as surrogate, Ni-63 as radioactive material) was the same and tended to be slightly higher for of Ni-63. This result confirmed that the aptamer could remove both the surrogate and the radioactive material similarly.

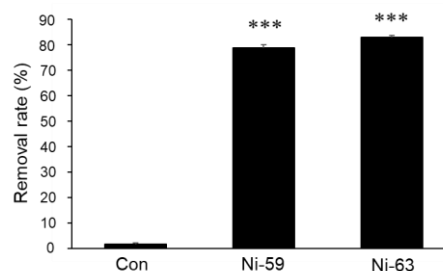


Fig. 2. Removal rates of nickel ions by aptamer. The concentration of non-radioactive nickel ion measured by ICP/OES and that of radioactive nickel ion measured by OOOO were compared to check for any differences in binding of the two isotopes. The experiment was performed three times. Control, only a column with the aptamer-bead complex; Ni-59 solution containing non-radioactive nickel ion was passed through the column; Ni-63, solution containing radioactive nickel ion was passed through the column. In all panels, error bars represent standard deviations of three replicates. *** $P < 0.001$ vs. control.

2.4 Recovery of Nickel from column containing aptamer/bead/Nickel

Regeneration is an important index in evaluating adsorbents for practical use. It has been reported that an aptamer maintains 90% performance even after being reused at least 20 times [4]. To determine the recovery rate of the aptamer, an aptamer-bead mixture was used to bind nickel; then, bound nickel was extracted twice with the same amount of buffer at 95°C, and the amount of nickel in the extract was measured (recovery rate (%) = $\frac{\text{extracted Ni}}{[\text{initial Ni}] - [\text{eluate Ni}]} \times 100$). The amount of extracted nickel was 99.95% or more of the initially bound nickel, and both aptamers (FA-N1, FA-N2) were yielded the same results [Fig. 3].

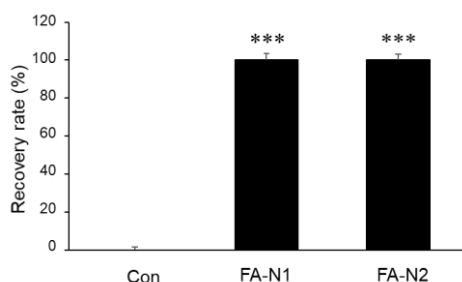


Fig. 3. Recovery rate of Cobalt throughout aptamer/bead column. After reacting with the aptamer-bead complex in a column, nickel bound to the aptamer was eluted by heating to denature of the aptamer. Nickel concentration in the supernatant was measured by ICP/OES and compared to the concentration of nickel initially bound to the aptamer. Recovery rate (%) = $100 - (\text{concentration of nickel eluted from the complex} / \text{concentration of nickel bound to complex})$. In all panels, error bars represent standard deviations of three replicates. *** $P < 0.001$ vs. control.

3. Conclusions

Radioactive materials are handled with care, and radioactive waste disposal is always an important issue. Here, a new treatment method for removal of nickel from radioactive waste is proposed. In this study, we focused on finding an alternative to ion-exchange resins, which are widely used for the removal of radioactive metal ions in nuclear power plants. FA-N1, an aptamer with high affinity for nickel, can capture about 80% of target ions and release more than 99.9% of bound nickel after a simple recovery procedure. The method is equally applicable to non-radioactive and radioactive nickel. The

results of this study provide a new resin that could supplement the existing ion-exchange resins that capture radioactive metal ions or even replace them.

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