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ABSTRACT TITLE: Catalytic DNA Biosensors for Radionuclides and Metal Ions

ABSTRACT: We are developing novel field-portable catalytic DNA biosensors to detect and quantify bioavailable radionuclides such as uranium, technetium, strontium, and plutonium as well as metal contaminants such as mercury and chromium. The sensors will be highly sensitive and selective, not only for different metal ions, but also for different oxidation states of the same metal ion (such as U(IV) vs. U(VI), or Cr(III) vs. Cr(VI)). To achieve the goals, we are using the combinatorial biological technique called *in vitro* selection to obtain catalytic DNAs that are highly specific for a given radionuclide or metal ion. We also use state-of-the-art biochemical and biophysical techniques to elucidate the structural elements responsible for high selectivity. These DNA have been transformed into fluorescent or colorimetric sensors by labeling the DNA with fluorophores or gold nanoparticles through catalytic beacon and nanoparticle assembly technologies developed in the PI's group, with detection limit down to 11 ppt and over millions-of-fold selectivity of uranyl over other radionuclides and metal ions. These sensors have been developed into commercially available sensor products that allow detection and quantification of radionuclides and metal ions in less than 2 min.

In the past year, we have made significant progresses in three areas. First, we have developed label-free catalytic and molecular beacon containing an abasic site for sensitive fluorescent detection metal ions such as Pb^{2+}.[1] The label-free method can significantly simplify the sensing method and lower its costs. Second, having obtained highly sensitive and selective sensors for radionuclides and metal ions, we have elucidated structural features and mechanism for the high sensitivity and selectivity, such as the importance of peripheral sequences in determining the metal selectivity of a catalytic DNA.[2] This knowledge will be quite valuable for designing new sensors and chelating agents for radionuclides and metal ions. Finally, for the first time, we have now extended the catalytic DNA sensors in detecting uranyl in living cells using catalytic DNA gold nanoparticles probe that can readily enter cells and can serve as a metal ion sensor within a cellular environment.[3] Such a method can be generally applied to the detection of other radionuclides and metal ions using other catalytic DNA selected through in vitro selection.

Detecting and quantifying radionuclides and metal contaminants onsite and in real-time in a simple and cost-effective way will impact many other areas of research under the SBR program. For example, it will enhance geochemistry/biogeochemistry research by lowering the costs of characterization and by providing more accurate information of the radionuclides and metal contaminants at the DOE sites. This information will strengthen the correlations between results obtained from microbial ecology and community dynamic analyses and DOE site properties, improve the understanding of the mechanisms of biotransformation, and provide deeper insight into biomolecular science and engineering. Practical applications of these sensors will not only help assess the effectiveness of science-based solutions for remediation performed by researchers and engineers, but will also contribute to the long-term monitoring of DOE contaminated sites by DOE staff members, state and local regulation agents, and concerned citizens around the sites.

1. Panshu Song, Yu Xiang, Hang Xing, Zhaojuan Zhou, Aijun Tong and Yi Lu, Analytical Chemistry 84, 2916-2922 (2012)).
2. Kevin E. Nelson, Hannah E. Ihms, Debapriya Mazumdar, Peter J. Bruesehoff and Yi Lu, ChemBioChem 13, 381-391 (2012).
3. Peiwen Wu, Kevin Hwang, Tian Lan and Yi Lu, *J. Am. Chem. Soc.*, 135, 5254–5257 (2013).