

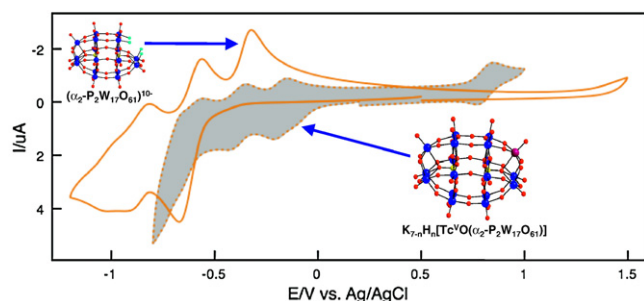
excesses of Sn(II), ligand, heat and low pH are usually employed for reduction. Polyoxometalates (POMs) are chemically robust metal oxide clusters that have unique and tunable electron transfer abilities. POMs can be reduced photochemically by multiple electrons, whilst maintaining their structure, and can, in turn, reduce high valent  $\text{Re}^{\text{VII}}$ . Irradiating a complexing ( $(\alpha_2\text{-P}_2\text{W}_{17}\text{O}_{61})^{10-}$ ) POM in  $\text{H}_2\text{O}$ /isopropanol in the presence of  $\text{ReO}_4^-$  results in reduction to  $\text{Re}^{\text{V}}$  and complexation into the defect of the POM. By supporting the POM on a solid support we envisage separation of the  $\text{Re}^{\text{V}}$  and transchelation to a suitable ligand for radiopharmaceutical purposes.

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### Redox-active ligands interrogate the complex oxidation state behavior of technetium

Benjamin P. Burton-Pye, Donna McGregor,  
Israel M. Mbomekalle, Lynn Carol Francesconi  
Hunter College of the City University of New York

Technetium possesses complex redox chemistry. Oxidation state stabilization of  $^{99\text{m}}\text{Tc}$  radiopharmaceuticals depends on the electronic environment set up by specific complexing ligands. We are attempting to understand oxidation state stability of Tc bound to ligands that undergo profound and rich redox chemistry. Polyoxometalates (POMs) are nanometer-sized metal oxides that can accept multiple electrons while maintaining their structural integrity. An electrochemical study of  $^{99\text{m}}\text{Tc}^{\text{V}}$  incorporated into POMs demonstrates that the  $\text{Tc}^{5/6}$  and  $\text{Tc}^{6/7}$  redox couples are not pH dependent (likely due to the unreactivity of the  $\text{Tc}=\text{O}$  moiety). The pH dependence of the  $\text{Tc}^{4/5}$  and  $\text{Tc}^{3/4}$  couples, however, suggests that reduction to lower valent Tc induces a structural change within the Tc-POM complex. We have employed multinuclear nuclear magnetic resonance and ultraviolet-visible spectro-electrochemistry to interrogate the low valent Tc coordination environment.



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### A pH-dependant structural study on Ga<sup>3+</sup> complexes with monoamide derivatives of 1,4,7-triazacyclononane-1,4,7-triacetic acid (NOTA) for PET applications

Dihesh Shetty, Jae Min Jeong, Soo-Young Choi, Lathika Hoigebazar,  
Yun-Sang Lee, Dong S. Lee, June-Key Chung, Myung C. Lee,  
Young K. Chung  
Department of Nuclear Medicine, Seoul National University, College of  
Medicine, 101 Daehangno Jongno-gu Seoul 110-744, Korea

All reported 1,4,7-triazacyclononane-1,4,7-triacetic acid (NOTA) derivatives for  $^{68}\text{Ga}$  labeling include a branched linker or an extra acetic acid, which require time-consuming multi-step syntheses. We synthesized two monoamide derivatives of NOTA conjugated with methylamine or benzylamine to see the possibility of stable  $\text{Ga}^{3+}$  complex formation.  $\text{Ga}^{3+}$  complexes were synthesized at different chelating pH values (3 and 5) and characterized by single crystal X-ray diffraction and multinuclear

nuclear magnetic resonance (NMR) spectroscopy. Crystals were obtained by slow evaporation of ethanol and water solution. X-ray crystal analysis showed the metal coordination to either nitrogen or oxygen atom of the amide bond at different chelation pH to form stable hexadentate complex. At pH 5, both ligands showed coordination to amide nitrogen. However, at pH 3, oxygen atom of amide bond was coordinated with gallium in case of benzylamine derivative, while methylamine derivative always showed nitrogen coordination. In solution chemistry,  $^{71}\text{Ga}$  NMR at range of pH showed possibility of two different coordination. Variable temperature  $^1\text{H}$  NMR spectroscopy showed the rigidity of these complexes. This finding could open up a wide range of applications for NOTA type bifunctional chelating agents by eliminating the multi-step synthesis required to introduce extra linkers.

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### A novel and nondestructive method for determining moisture in lyophilized reagents for labeling with $^{99\text{m}}\text{Tc}$

Bianca C.G. de Abreu, Ademar C. Filho,  
Neuza T.O. Fukumori, Margareth M.N. Matsuda  
Diretoria de Radiofarmácia (DIRF), Instituto de Pesquisas Energéticas e  
Nucleares (IPEN-CNEN-SP), Av. Professor Lineu Prestes,  
2242-05508-000, São Paulo/SP, Brasil

The presence of  $\text{SnCl}_2$  in the lyophilized reagents (LR) used in radiodiagnosis in nuclear medicine makes the determination of the moisture content an essential procedure to ensure the product quality because the decomposition of  $\text{Sn}^{2+}$  by hydrolysis and oxidation can reduce the yield of  $^{99\text{m}}\text{Tc}$  radiolabeling. The aim of this work was to determine the moisture content in ethylcysteinate dimer (ECD), methylene diphosphonate (MDP), methoxyisobutyl isonitrile (MIBI) and Sn (tin colloid) by a nondestructive method. A Computrac Vapor Pro RX equipment from Arizona Instrument was used. The heating temperature ( $100^\circ\text{C}$ ,  $140^\circ\text{C}$ ,  $170^\circ\text{C}$  and  $200^\circ\text{C}$ ) and the rate ending of moisture loss ( $0.01$ ;  $0.1$  and  $0.5 \mu\text{g s}^{-1}$ ) variation were evaluated. The bottles were weighed after withdrawing the residual moisture and before the introduction of the sample. The linear range for determination of water was  $y=228.36+1070.93x$ , and the product mass was in the range of 6–23 mg. The influence of the temperature and the rate ending of moisture loss were different for each LR. The percentage of water in ECD, MDP, MIBI and Sn were  $(1.60\pm 0.13)\%$ ,  $(4.89\pm 0.33)\%$ ,  $(3.20\pm 0.30)\%$  and  $(4.08\pm 0.06)\%$ , respectively, at optimized conditions:  $140^\circ\text{C}$  for ECD,  $100^\circ\text{C}$  for MDP and MIBI and  $200^\circ\text{C}$  for Sn. It showed to be an efficient method for moisture determination in LR.

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### Tumor imaging with $^{68}\text{Ga}$ -positron emission tomography: synthesis and characterization of macrocycle-amino acid derivatives

Dinesh Shetty, Jae Min Jeong\*, Chang Hwan Ju, Yun-Sang Lee,  
Dong Soo Lee, June-Key Chung, Myung Chul Lee  
Department of Nuclear Medicine, Seoul National University College of  
Medicine, South Korea

$^{68}\text{Ga}$  positron emission tomography (PET) imaging in clinical oncology is a noted development due to cyclotron independent availability. Labeled amino acid derivatives have been proved useful in imaging many kinds of tumors. We synthesized 3-aminoalanine, 4-aminohomoalanine and lysine derivatives of macrocyclic chelating agents by conjugating amino acids to mono carboxylic group of NOTA and DOTA using either EDC or DCC. Pure compounds were labeled with  $^{68}\text{Ga}$  (labeling efficiency  $>95\%$ , Specific Activity  $\sim 1.94\text{--}9.21 \text{ GBq}/\mu\text{mol}$ ). The preliminary evaluation was studied in Hep3B and CT-26 cancer cells, which showed high uptake of these derivatives. Highest in vivo tumor uptake was showed by  $^{68}\text{Ga}$ -NOTA-aminohomoalanine ( $2.40\pm 0.85\% \text{ ID/g}$ ) at 30 min. PET images in mice