Ph.D. Thesis Abstract for Jonathon A. Nye

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Production of Radiohalogens and [11C]-Methane at High Specific Activity

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At the University of Wisconsin-Madison
August 21, 2005

The halogens, occupying Group VII of the periodic table, play an important role in the biochemical processes underlying health and disease. A variety of positron emitters covering a broad range of half-lives permit the imaging of the body's physiochemical behavior using PET. Neutron deficient isotopes of the halogen group can be produced by (p,n) reactions from enriched targets with low energy (<13MeV) biomedical cyclotrons. These cyclotrons are distributed relatively evenly throughout the United States at research institutions and commercial distribution sites (i.e., 100+ CTI RDS 11MeV proton cyclotrons). However, these sites concentrate on the core group of positron emitters: ^{15}O, ^{13}N, ^{11}C, and primarily ^{18}F-fluoride. The simplicity of the production process insures their role in the clinical/research environment, labeling $H_2{}^{15}O$, $^{13}NH_3$, CH_3-compounds and ^{18}F-FDG. Halogens with half-lives longer than ^{18}F have been avoided due to a combination of several factors, such as complexity of the target systems, expense of the enriched substrate, low reaction yields, and extensive post-processing to reclaim the target material.

PET research over the last decade has forced a match between drug development and emerging small animal instrumentation, shifting focus to agents labeled with high specific activity $^{11}CH_3I$ and the long-lived radiohalogens, ^{76}Br and ^{124}I. A steady local supply of ^{18}F-fluoride, ^{11}C-methane, ^{76}B-bromide, and ^{124}I-iodide is essential to seize today's research opportunities or for limited distribution outside of our local area. To keep pace, new targetry developments are implemented to reliably produce these isotopes on a batch basis. The research presented details improvements on existing production methods for ^{18}F-fluoride intended for nucleophilic substitution and high specific activity ^{11}C-methane (CH_3I) for the N-methylation of a half-dozen neuroligands. A significant effort is placed on the novel use of low energy cyclotrons for the production of ^{76}Br and ^{124}I involved in labeling antibody and protein agents. Performance of these new designs and the success of the solid targetry development will be described. The ten-fold scale-up in yields at end-of-bombardment promises new hope for the synthesis of PET tracers, previously limited by access to the radio-halogen precursors.

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last updated: 08/01/2008