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Microfluidic solvent extraction for an automated 68Ga cyclotron production loop

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Since the introduction of the first commercial 68Ge/68Ga generator about 20 years ago, interest in 68Ga-labelled radiopharmaceuticals is steadily increasing. Currently, over 400 clinical studies including 68Ga are being executed. Additionally, several PET imaging agents were approved for clinical use by the FDA, the latest being a new prostate-specific imaging agent, potentially leading to a significant increase in 68Ga demand in the future. To date 68Ga is still mostly produced by 68Ge/68Ga generators, but these generators show several drawbacks, such as a high amount of radioactive waste after the shelf-life of a generator is reached, the decreasing resources to produce 68Ge and a limited supply of 68Ga per day. To accommodate the increasing demand, alternative production routes are being investigated. Cyclotron-based production of 68Ga, through proton irradiation of 68Zn(NO3)2 liquid targets, is therefore of increasing interest as an alternative. After irradiation, 68Ga needs to be separated from the liquid target solutions, but lengthy, multi-step procedures involving ion exchange chromatography can lead to a significant loss of activity. Furthermore, the use of costly, enriched 68Zn target material necessitates target solution recycling. In the present study, we developed a new, two-step solvent extraction process to recover 68Ga from zinc nitrate solutions in under 15 minutes. Solution compositions can range from 1 - 5 M natZn(NO3)2 in 0.01 - 1 M HNO3 and the organic extracting phase consisted of 200 mM N-benzoyl-N-phenylhydroxylamine dissolved in chloroform. In conventional batch extraction, efficiencies of up to 99.6% ± 0.3% were achieved within 10 minutes. Back-extraction into different HCl solutions resulted in efficiencies of up to 94.5% ± 0.6% within 1 minute. Additionally, microfluidic solvent extraction was investigated, due to its ability to be integrated in an automated set-up, opposed to conventional batch extraction. By microfluidic solvent extraction using a membrane separator, comparable results of 99.2% ± 0.3% for extraction and 95.8% ± 0.8% for back-extraction could be achieved. Zinc contamination in the final HCl solution after back-extraction, was found to be below 3 ppm and is thus within hospital quality standards. Because all zinc nitrate solutions were used without any modification, this method should allow for direct recycling of the target solutions after the extraction process making it a suitable method to create a fast, efficient cyclotron irradiation loop, that enables a continuous supply of 68Ga.

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