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Microfluidic solvent extraction for an automated ^{68}Ga cyclotron production loop

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Since the introduction of the first commercial $^{68}\text{Ge}/^{68}\text{Ga}$ generator about 20 years ago, interest in ^{68}Ga -labelled radiopharmaceuticals is steadily increasing. Currently, over 400 clinical studies including ^{68}Ga 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 ^{68}Ga demand in the future. To date ^{68}Ga is still mostly produced by $^{68}\text{Ge}/^{68}\text{Ga}$ 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 ^{68}Ge and a limited supply of ^{68}Ga per day. To accommodate the increasing demand, alternative production routes are being investigated. Cyclotron-based production of ^{68}Ga , through proton irradiation of $^{68}\text{Zn}(\text{NO}_3)_2$ liquid targets, is therefore of increasing interest as an alternative. After irradiation, ^{68}Ga 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 ^{68}Zn target material necessitates target solution recycling. In the present study, we developed a new, two-step solvent extraction process to recover ^{68}Ga from zinc nitrate solutions in under 15 minutes. Solution compositions can range from 1 - 5 M $\text{natZn}(\text{NO}_3)_2$ in 0.01 - 1 M HNO_3 and the organic extracting phase consisted of 200 mM N-benzoyl-N-phenylhydroxylamine dissolved in chloroform. In conventional batch extraction, efficiencies of up to $99.6\% \pm 0.3\%$ were achieved within 10 minutes. Back-extraction into different HCl solutions resulted in efficiencies of up to $94.5\% \pm 0.6\%$ 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 $99.2\% \pm 0.3\%$ for extraction and $95.8\% \pm 0.8\%$ 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 ^{68}Ga .

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