Nuclear Inst. and Methods in Physics Research, A 874 (2017) 43–49

Contents lists available at ScienceDirect

Nuclear Inst. and Methods in Physics Research, A

journal homepage: www.elsevier.com/locate/nima

Development and characterization of a Drop-on-Demand inkjet printing system for nuclear target fabrication

R. Haas [a,b,*], S. Lohse [a,b], Ch.E. Düllmann [a,b,c], K. Eberhardt [a,b], C. Mokry [a,b], J. Runke [a,c]

[a] Johannes Gutenberg-Universität Mainz, Institut für Kernchemie, Fritz Strassmann Weg 2, 55128 Mainz, Germany
[b] Helmholtz-Institut Mainz, Staudinger Weg 18, 55128 Mainz, Germany
[c] GSI Helmholtzzentrum für Schwerionenforschung GmbH, Planckstraße 1, 64291 Darmstadt, Germany

ARTICLE INFO

Keywords:
Drop-on-Demand
Inkjet printing
Thin layers
Storage phosphor imaging plates
Radiographic imaging
Targets

ABSTRACT

A novel target preparation method based on Drop-on-Demand (DoD) inkjet printing has been developed. Conventional preparation methods like the electrochemical method “Molecular Plating” or the “Polymer-Assisted Deposition Method” are often limited, e.g., concerning the dimensions and geometries of depositions or by the requirement for electrically conducting substrates. Here, we report on the development of a new technique, which overcomes such limits by using a commercially available DoD dispenser. A variety of solutions with volumes down to 5 nL can be dispensed onto every manageable substrate. The dispensed volumes were determined with a radioactive tracer and the deposits of evaporated salt solutions were investigated on titanium and graphene foils. Additionally, the high precision of the printing system with which individual drops can be positioned was used to determine the spatial resolution of storage phosphor imaging plates with three tracers of different β-decay energies. The new technique is able to produce new kinds of targets with improved spatial geometries and thin layer deposits.

1. Introduction

There is a high demand for well prepared thin actinide layers serving as targets in both nuclear chemistry and nuclear physics applications. These include accelerator-based experiments like the production of the heaviest elements [1], nuclear reaction data needs from basic and applied sciences [2,3], or sources of fission fragments [4] as well as α-daughter recoil nuclei [5]. Generally, targets have to meet high demands in terms of chemical purity, homogeneity, thickness, as well as adherence to the deposition substrate. One of the most successful production methods for such thin layer depositions for nuclear applications is the electrochemical deposition from organic solvents called Molecular Plating (MP) [6,7]. Alternative methods include manual pipetting on superhydrophobic surfaces [8] or polymer assisted deposition (PAD) [9,10], developed by Jia et al. [11], the latter being suitable for the preparation of thin films of metal oxides. Naturally, all methods have some inherent limitations, like the need for an electrically conductive substrate in MP, or the limited size and constrained geometric form the target can take in MP and PAD. Manual pipetting, on the other hand, suffers from limited accuracy. An automated pipetting system has recently been built by a group at the Paul Scherrer Institute (PSI), Villigen, Switzerland, to overcome the limits in accuracy of manual pipetting and has been used for the production of targets for experiments of the n_TOF collaboration at CERN [12]. The application of precision printing systems like DoD inkjet printing has recently spread increasingly in many branches of the natural sciences.

The DoD technique is widely used in commercial printers for producing droplets of small, well defined volumes and to dispense them in a defined area, e.g., for barcode labeling. Here, single droplet dosage is performed either piezoelectrically or thermally [13]. By using modified commercially available inkjet printers, the thermal DoD method and its produced deposits were investigated by Fittschen et al. [14]. Thermal DoD printers are able to produce droplets of solutions in the range of picoliters with a good reproducibility, which lead to deposits with diameters of 5 μm to 20 μm in a hemispherical shape. In contrast, simple droplet evaporation of larger droplets is known to produce ring-shaped residues [14], which can best be overcome by depositing on superhydrophobic surfaces [8], which, though, introduces new limitations. For radiochemical applications, thermal DoD printers have a big disadvantage because a part of the solution is volatilized inside the printing head and the internal reservoir gets contaminated and cannot be

---

[*] Corresponding author at: Johannes Gutenberg-Universität Mainz, Institut für Kernchemie, Fritz Strassmann Weg 2, 55128 Mainz, Germany.
E-mail address: raphhaas@uni-mainz.de (R. Haas).

http://dx.doi.org/10.1016/j.nima.2017.08.027
Received 29 June 2017; Received in revised form 14 July 2017; Accepted 15 August 2017
Available online 24 August 2017

exchanged. Hence, the printing head would have to be fully exchanged for a different application to avoid cross-contamination effects. As an alternative to thermal inkjet printing piezoelectric DoD dispensers have been investigated. One such system is the PipeJet® P9 Nanodispenser from Biofluidix. This features an external reservoir, which is in contrast to thermal inkjet printers. It was shown that the modified thermal inkjet printer has a smaller deposition volume as well as the ability to print bands, whereas the piezoelectric dispenser has a slightly better precision in printing spots [15]. Piezoelectric DoD printers are currently used, e.g., in energy research to produce inkjet-printed $Cu_2ZnSn(S, Se)_4$ solar cells [16]. Piezoelectric dispensers have an external reservoir, which is easily exchangeable and the solution is not in contact with internal parts of the dispenser. Therefore, in this work, a new method to produce targets based on piezoelectric DoD inkjet printing has been developed and basic features of the produced thin layer deposits have been characterized. The setup of the system is built up of a sample stage and a dispenser based on the DoD technology with an external reservoir for liquids. It is possible to dispense liquids over a wide range of viscosities, from water-based solutions to, e.g., glycerin-based ones. Droplet volumes between 5 nL and 60 nL can be produced, which is far below the limit of commercially available manual pipettes.

In this work, different droplet volumes for water-based solutions with a specific concentration were verified using radioactive tracers. Additionally, the residue after droplet evaporation was characterized via optical microscopy as well as scanning electron microscopy (SEM). The system was finally applied to produce well defined patterns of radioactive species to study the spatial resolution of an autoradiographic imaging system as a function of β-decay energy.

2. DoD inkjet printing system

2.1. Setup of the printing system

The inkjet printing system comprises a DoD dispenser (Biofluidix, PipeJet® P9 Nanodispenser) for the droplet production, and two compact motorized translation stages (Thorlabs) to move a substrate in two dimensions, see Fig. 1. The dispenser consists of a printing head with an electronically controlled piezo-driven piston inside its casing and a replaceable tip connected to an external reservoir. The tips are made of polyimide and polypropylene and have an inner diameter of 200 μm. To connect the tip and the reservoir, an 8 cm-long flexible tube (Proliquid GmbH, LMT-55) made from TYGON® is used. The reservoir is a commercially available pipette tip made of polypropylene and can be fixed to the holder of the dispenser. To adjust the height of the dispenser over the substrate, the former is mounted to a stand. Each translation stage has a travel range of 50 mm and a bidirectional repeatability of 1.6 μm [17]. To fix substrates in a well-defined position during movements, a holder for circular substrates with a diameter of 26 mm is mounted on the top stage. This can be easily replaced with holders for targets of different geometries, providing for the flexibility of the setup. The liquid is held in external, easily exchangeable and cheap parts and not in the casing of the dispenser, which is a significant advantage over normal inkjet cartridges and especially relevant for applications using a multitude of sample solutions. Thus the work with radioactive substances is simplified, because the contaminated parts can easily be exchanged.

For good printing results, a vertical distance of 1 mm to 2 mm between the tip and the substrate is recommended by the manufacturer. Liquids and solutions with a viscosity of 0.5 mPa·s to 500 mPa·s and a surface tension of 30 mN/m to 76 mN/m are suitable for dispensing. The manufacturer specifies a volume precision of <3% and accuracy of <10% for the dispenser [18]. Drop volumes of 5 nL to 60 nL can be generated in dependence of the used tip diameter. For each drop volume, concentration of a solution and solvent, the stroke velocity of the piston has to be calibrated via a manufacturer-delivered software. An optimal value is found when the dispenser can continuously print without generating satellite drops. The drop volume is set by the stroke range and the stroke velocity of the piezo-driven piston inside the dispenser. The schematic principle of the dispenser is illustrated in Fig. 2. Once filled, the liquid is kept inside the elastic polymer tip due to its surface tension. After a fast displacement of the piston with a specific stroke range and stroke velocity, the liquid is forced out of the respective part of the tip hit by the piston in both directions. At the bottom of the tip, a drop is generated and falls off. By a slow release, new liquid from the reservoir fills up the tip again. The stroke range influences the suppressed liquid volume inside the tip and hence is crucial for setting a drop volume. Additionally, the stroke velocity enables dispensing of a specific volume. If the stroke velocity is set too low, the kinetic energy is insufficient to let the formed drop fall off the tip. Too high values cause spraying or the generation of satellite drops. Therefore, the stroke velocity has to be calibrated for each solution and desired volume. Typical stroke velocity values are 60 μm/ms to 100 μm/ms for water-based samples, 50 μm/ms to 80 μm/ms for organic solvents and >150 μm/ms for viscous media. The maximum frequency of the dispenser to produce droplets lies at 100 Hz depending on the liquid used. After the dispensing process the drops evaporate on the substrate, leaving behind the residue of the dissolved species.

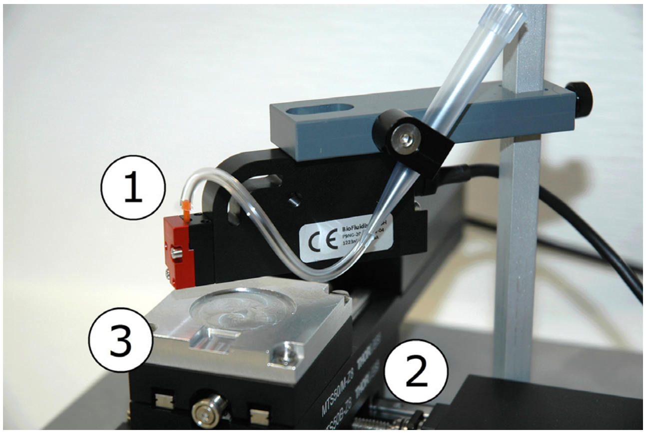

Fig. 1. View of the DoD printing system containing the piezo dispenser (1) and two compact motorized translation stages (2) with a holder for substrates with a diameter of 26 mm (3).

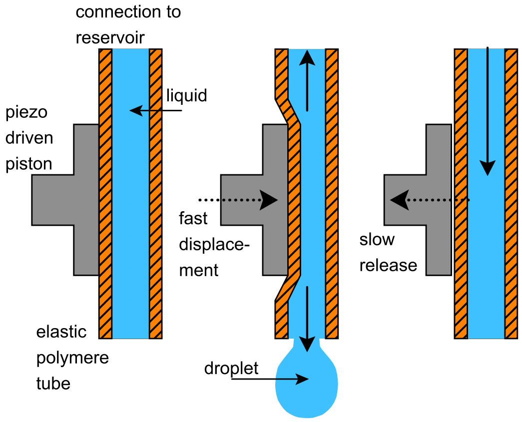

Fig. 2. Schematic representation of the dispensing process [18]. A liquid is kept inside the elastic polymer tip by its surface tension until a fast displacement of the piezo driven piston. The liquid is subsequently pressed to both directions, and a droplet forms at the lower end of the tip and is released. After a slow release of the piston, new liquid runs from the reservoir into the tip to fill up the tip again.