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Citation vs. Self-Citation
Effect of wall material and shape on MeV ion focusing ability of tapered capillary optics

Sarawut Jaipen\textsuperscript{a,b,*}, Nares Chankow\textsuperscript{a}, Jun Hasegawa\textsuperscript{c}, Yoshiyuki Oguri\textsuperscript{b}

\textsuperscript{a} Department of Nuclear Engineering, Faculty of Engineering, Chulalongkorn University, Piyathal Road, Pathumwan, Bangkok 10330, Thailand
\textsuperscript{b} Research Laboratory for Nuclear Reactors, Tokyo Institute of Technology, Ookayama 2-12-1-5N-14, Meguro-ku, Tokyo 152-8550, Japan
\textsuperscript{c} Department of Energy Science, Tokyo Institute of Technology, 4259-G3-35, Nagatsuta-cho, Midori-ku, Yokohama 226-8503, Japan

1. Introduction

The elastic scattering of ions by atomic nuclei is one of the major interaction processes between energetic ions and a solid surface. The projectile ions approaching the nuclei in a solid target undergo conventional Coulomb or Rutherford scattering. Then, the projectiles lose their energy and change their direction of motion. In most scattering events, however, the scattering angle and the corresponding energy loss are very small because the scattering cross section is inversely proportional to the fourth power of the half scattering angle. Based on the idea that such glancing-angle scatterings may be useful for ion beam focusing, Nebiki et al. [1] demonstrated the ability to focus 2 MeV helium ions with tapered glass capillaries having outlet diameters of less than 1 µm. They successfully observed a beam focusing effect of the tapered glass capillaries and applied this new technique to in-air PIXE analysis [2]. Because the microbeam irradiation device based on this technique is compact and inexpensive compared to the conventional devices using a combination of a collimator and quadrupole magnets, this technique has been studied [3–5] and applied to various applications such as nuclear reaction analysis [6], high-contrast X-ray imaging [7], cell surgery [8], micro-PIXE analysis [9–10], in-air RBS [11], and in-air STEM [12].

An index of the ion focusing ability of the tapered capillary optics is the beam-focusing ratio, which is defined as the ratio of the averaged beam current density measured at the capillary outlet to that measured at the inlet. Because the beam heat load on the capillary glass limits the allowable incident beam current, the improvement of the beam-focusing ratio has been one of the crucial issues to be considered while developing applications of the tapered capillary optics. The strong dependence of the Rutherford scattering cross section on the target atomic number ($\sim Z^2$) indicates that the beam-focusing ratio might be improved by using capillaries made of heavier materials with higher atomic numbers. Moreover, because the scattering cross section is very sensitive to the scattering angle, the focusing ratio might be improved also by optimizing the shape of the capillary inner wall [13].

The purpose of this study was to reveal the effects of the capillary wall material and shape on the beam-focusing ability of the tapered capillary optics. To examine the wall material effect, we prepared tapered capillaries made of two different glass materials, borosilicate glass and lead glass, and compared the beam-focusing ratios evaluated from the energy spectra of capillary-focused protons. We also performed Monte Carlo (MC) simulations not only to support the experiment but also to investigate the effects of the wall material and shape in greater detail.

\textsuperscript{a} Corresponding author at: Department of Nuclear Engineering, Faculty of Engineering, Chulalongkorn University, Piyathal Road, Pathumwan, Bangkok 10330, Thailand.
\textsuperscript{b} E-mail addresses: jayjen.su@mu.titech.ac.jp (S. Jaipen), jun.hasegawa@es.titech.ac.jp (J. Hasegawa).

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Table 1
Specifications of glass tubes used in this study.

<table>
<thead>
<tr>
<th>Capillary glass</th>
<th>Elemental composition</th>
<th>Effective atomic number</th>
<th>Softening temperature (°C)</th>
<th>Outer/inner diameter (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Borosilicate glass (Premium corning 7059 Patch glass)</td>
<td>62.1% O, 21.5% Si, 9.8% B, 3.6% K, 1.1% Al, 1.3% Li, 0.6% Na</td>
<td>10.0</td>
<td>~718</td>
<td>1.65/1.20</td>
</tr>
<tr>
<td>Lead glass (Premium corning B161 Patch glass)</td>
<td>62.7% O, 8.9% Pb, 25.4% Si, 2.5% K, 0.5% Ba</td>
<td>26.7</td>
<td>~600</td>
<td>1.65/1.20</td>
</tr>
</tbody>
</table>

2. Fabrication of tapered glass capillaries

To extend a straight glass tube and form a gradually tapered glass capillary, we used a commercially available puller (PE-21, Narishige Co., Ltd.) consisting of a platinum heater and two types of electromagnets. Both ends of a straight tube were clamped tightly and pulled while the middle part of the tube was heated up to a temperature beyond its softening point. By controlling the amount of heat and the pulling force, we were able to fabricate tapered glass capillaries having various taper angles. After the pulling process, the tapered glass capillaries typically had an outlet diameter of less than 1 μm. To obtain a desired outlet diameter (30 μm in this study), a micro-force device (MF900, Narishige Co., Ltd.) was used to cut the tip of the capillary.

In this study, we fabricated the tapered glass capillaries from borosilicate glass and lead glass tubes having the same nominal inner and outer diameters. The specifications of the glass tubes are summarized in Table 1. Because borosilicate glass and lead glass have different softening temperatures, we carefully optimized the amount of heat and the pulling force separately for these two glass materials such that the tapered glass capillaries had the same inner wall shape. Fig. 1a shows the complete view of the tapered glass capillaries made of borosilicate glass (upper) and lead glass (lower). The outlet diameter and the total length were approximately 30 μm and 4 cm, respectively. Magnified images of the capillary outlet taken by a scanning electron microscope (SEM) are also shown in Fig. 1b and c. The outlet diameters of the borosilicate-glass and the lead-glass capillaries were evaluated from these images to be 31.5 μm and 32.5 μm, respectively.

To digitize the inner wall shape of the tapered capillary, we took magnified images of the capillary using a CCD camera connected to an optical microscope with changing the position of the capillary. The inner diameter of the capillary was precisely measured from these images every 0.2 mm along the center axis of the capillary.

Fig. 2a compares the inner wall shapes of the borosilicate-glass and the lead-glass capillaries. We see that the inner diameters at the inlet are not exactly the same, which is probably due to a manufacturing error of the original straight glass tubes. On the other hand, the wall shapes around the tip are identical to each other, which is advantageous for the comparison between the beam-focusing abilities of these two capillaries because the inner wall near the tip is considered to be the main contributor to the beam transmission through the tapered capillary. Fig. 2b and c compares the inner-wall profiles measured at a rotation angle of 0° with those observed at 90° for the borosilicate-glass and the lead-glass capillaries. We confirmed that the profiles measured at both angles coincide well with each other and the axial symmetry is high. The outlet diameters measured by the optical microscope well reproduced those determined from the SEM images (Fig. 1b and c), proving that the optical distortions due to the refraction of light are negligible in the measurement using the optical microscope.

3. Experimental setup

The beam-focusing experiments using the tapered glass capillaries were performed in a vacuum chamber at a pressure of
approximately 10⁻⁴ Pa. The configuration of the experimental apparatus in the chamber is schematically shown in Fig. 3. The tapered glass capillary was mounted on a stainless steel holder and its tilt angles were adjusted with respect to the beam axis using remote-controlled actuators. The incident proton beam from a 1.6 MV tandem accelerator was collimated by a 2-mm rectangular slit 2.2 m upstream of the capillary inlet and a 1.2 mm diameter aperture just in front of the capillary inlet. The beam energy was fixed at 2 MeV throughout this study.

Before every experiment with a newly installed capillary, we precisely aligned the capillary with respect to the beam axis as follows. First, we roughly adjusted the capillary tilt angles by maximizing the brightness of a beam spot on a plastic scintillator 65 cm downstream from the capillary outlet. Then, we changed the plastic scintillator to an imaging plate (BAS-TR2025, Fujifilm Corp.) to record the intensity distribution of the proton beam. After beam irradiation for a few seconds, the beam-intensity distribution on the imaging plate was read out using a scanner as an image of photostimulated luminescence (PSL) intensity. We finely adjusted the capillary tilt angles by repeating this procedure until we obtained an image with good axial symmetry.

The energy spectra of the protons focused by the tapered capillary were measured by using a silicon semiconductor detector (SSD) in two different experimental setups. First, the SSD was set exactly behind the capillary outlet and 5 mm from it. Because the detector solid angle was larger than the divergence angle of the proton beam, all protons exiting from the capillary were detected in this setup. In the other setup, the SSD was set 15 cm downstream from the capillary outlet. We placed a 50 μm thick tantalum plate with a 100 μm diameter aperture just in front of the SSD to decrease the detector solid angle. The SSD was mounted on the motorized stage, and the position of the SSD was changed every 0.125 mm perpendicular to the beam axis. The proton energy spectra were measured at various detector positions, and the beam intensities were evaluated from the total proton counts. To avoid the unwanted effect of the incident-beam current fluctuation, the beam intensity was normalized by the total proton charge deposited on the capillary holder.

4. Numerical simulation

We numerically simulated the transmission of 2 MeV protons in tapered capillary optics by using an originally developed MC code. In this code, the classical Rutherford scattering formula was used for the calculation of the scattering cross section. The stopping power of the wall material for the protons was imported from the SRIM database [14]. We examined not only the ion transmission in the borosilicate-glass and the lead-glass capillaries but also that in capillaries produced from single elements such as Be, C, Al, Fe, Ag, and Au to obtain a more general view of the effect of the wall atomic number. The data of the inner wall shapes shown in Fig. 2a were used in the calculation to simulate the experiment. In addition, the ion transmission in a tapered capillary having a constant taper angle was calculated to examine the effect of the capillary wall shape. In this paper, we call this capillary "conical capillary" to distinguish it from the tapered capillary actually used in the experiment. In the MC simulation, the conical capillary was defined as having the same outlet/inlet diameters and taper length as the tapered capillary (outlet diameter: 30 μm, inlet diameter: 1.2 mm, taper length: 15 mm).

5. Results and discussion

Fig. 4 shows the PSL intensity distributions on imaging plates irradiated by proton beams focused by the lead-glass capillary. As shown in the figure, the beam spot consists of an intense small spot called the "core" and a weak surrounding ring-shaped pattern called the "halo." We consider that the core and the halo are composed of the protons traveling straight through the capillary and those scattered by the capillary inner wall, respectively. When the capillary axis is misaligned with respect to the beam axis, the core overlaps the halo ring and the halo ring pattern is asymmetric.

![Fig. 3. Schematic of the experimental setup.](image)

![Fig. 4. Beam spot patterns recorded on imaging plates when the tapered capillary (lead glass) was (a) misaligned and (b) well aligned with respect to the incident beam axis. Imaging plates were located 65 cm downstream from the capillary tip.](image)

![Fig. 5. Energy spectra of protons focused by (a) borosilicate-glass capillary and (b) lead-glass capillary. The gray solid and dashed lines in Fig. 5a) show the core component and the proton incident energy, respectively.](image)
(Fig. 4a). After the optimization of the capillary tilt angle, the beam spot pattern becomes axially symmetric (Fig. 4b). However, even after the optimization, asymmetry is still observed particularly in the halo ring; this is probably due to the inhomogeneity of the incident beam profile.

Fig. 5 shows the energy spectra of protons focused by (a) the borosilicate-glass capillary and (b) the lead-glass capillary; the spectra were measured by the SSD 5 mm downstream from the capillary outlet. There seems to be no considerable difference between these two spectra. Then, we numerically divided the total proton count into two parts, corresponding to the core and the halo components, and evaluated a beam-focusing ratio from each energy spectrum. By assuming that the protons in the core lost no energy in the capillary and considering the finite energy resolution of the SSD, we used the following formulae to estimate the total counts of the core $N_{core}$ and the halo $N_{halo}$:

$$N_{core} = \int_{E_{min}}^{E_{max}} n(E)dE,$$

$$N_{halo} = N_{total} - N_{core}.$$  

Here, $E_0$ is the incident kinetic energy of protons (2 MeV), $E_{max}$ is the maximum energy of the proton in the spectrum, $n(E)dE$ is the count of protons having an energy between $E$ and $E + dE$, and $N_{total}$ is the total count in the spectrum. The gray solid line in the figure shows the core component defined by Eq. (1). The beam-focusing ratio of the tapered capillary was evaluated from the following formula:

$$\eta = \frac{N_{core} + N_{halo}}{N_{core}} = 1 + \frac{N_{halo}}{N_{core}}.$$  

From Fig. 5, we obtained $\eta = 1.32$ for the borosilicate glass capillary and $\eta = 1.37$ for the lead-glass capillary. On the other hand, the effective atomic number for the Rutherford scattering process was calculated as follows:

$$Z_{eff} = \left( \sum_i Z_i^2 f(i) \right)^{1/2}.$$  

Here, $Z_i$ and $f(i)$ are, respectively, the atomic number and the fraction of $i$-th element in the capillary wall. Although the effective atomic number of the lead glass is approximately thrice that of the borosilicate glass as listed in Table 1, the observed enhancement in the beam-focusing ratio was much lower than that expected from the dependence of the scattering cross section on the wall atomic number. This result implies that the scattering cross section was not a dominant factor determining the transmission and the focusing of MeV protons in the tapered capillary.

To discuss the effect of the wall atomic number on the beam-focusing process in greater detail, we investigated the core and the halo components separately. The intensity distributions of the proton beams, which were obtained by the spatially resolved energy spectrum measurement, are shown in Fig. 6 for the borosilicate-glass and the lead-glass capillaries. The intensity profiles were normalized by the respective intensities of the core components. As shown in the figure, the halo intensity peak for the lead-glass capillary is higher than that for the borosilicate-glass capillary. This result shows that the increase in the effective atomic number of the capillary wall actually leads to the increase in the scattered component intensity. However, according to the focusing ratios evaluated above, the increased wall atomic number seems not to contribute to the total beam transmission so much.

Fig. 6. Intensity distributions of the proton beams focused by borosilicate-glass and lead-glass capillaries. Beam intensities were evaluated along the dashed line in the beam spot image.

Fig. 7. Trajectories of protons focused by (a) a tapered capillary and (b) a conical capillary, and two-dimensional angular distribution of the protons on the detector of (c) a tapered capillary and (d) a conical capillary.
Fig. 7 shows the MC simulation results for proton trajectories in (a) the tapered capillary and (b) the conical capillary, and the angular distributions of protons focused by (c) the tapered capillary and (d) the conical capillary. Note that Fig. 7a and b shows only the trajectories of the protons that finally exit from the outlet. As shown in Fig. 7a, only the capillary inner wall near the outlet contributes to the beam focusing in the tapered glass capillary. This result justifies the use of the borosilicate-glass and the lead-glass capillaries having the same inner wall shape around the tip (see Fig. 2a) in the experiment. From the values of $N_{\text{cone}}$ and $N_{\text{cone}}$ obtained in the MC simulations, the focusing ratios were evaluated by Eq. (3) to be 1.60 and 1.63 for the borosilicate-glass and the lead-glass capillaries, respectively. These values are somewhat larger than those obtained from the experiment. This might be because the experimental focusing ratio was reduced by the inhomogeneity of the incident beam.

On the other hand, in the case of the conical capillary, almost all areas of the inner wall contribute to the beam focusing, as shown in Fig. 7b. Particularly, when the wall material is lead glass, the effective use of the capillary inner wall results in the focusing ratio for the conical capillary ($\gamma = 1.98$) being higher than that for the tapered capillary ($\gamma = 1.63$). However, as shown in Fig. 7d, the use of the conical capillary evidently makes the distribution of the halo component more diffuse because of its larger taper angle (∼28 mrad). Because the taper angle determines the minimum divergence angle of the halo particles, there is a trade-off between the beam-focusing ratio and the beam quality.

Fig. 8 plots the beam-focusing ratios obtained from the MC simulations as a function of the wall atomic number. When the wall atomic number is relatively low (Be and C), the focusing ratios for the conical capillary are smaller than those for the tapered capillary. This is probably because the cross section is very small for MeV proton scattering by such light atoms with a relatively larger scattering angle (>28 mrad). With increasing wall atomic number, the focusing ratio for the conical capillary increases gradually and reaches a value approximately twice that for the tapered capillary when the wall material is gold. In addition, the focusing ratio for the tapered capillary seems to be almost independent of the wall atomic number. To understand these trends, we need to consider the fact that most scattering events occur not on the surface of the capillary inner wall but in the body of the wall. After being scattered by an atom inside the glass wall, the proton needs to escape from the wall to finally exit from the capillary outlet. Here, we define the distance that the scattered proton travels in the glass body after the scattering as the "escape distance" ($a$ and $b$ in the schematic illustration in Fig. 8). As for the tapered capillary, because the escape distance is relatively large because of its convex inner wall, most scattered protons are stopped inside the capillary wall and are unable to escape from it. This is consistent with the results in Fig. 7a, which indicate that the beam focusing occurs only at the capillary wall near the outlet where the escape distance is relatively small. The escape distance for the conical capillary is, on average, shorter than that for the tapered capillary, which results in the effective use of the capillary inner wall for beam transmission and relatively high focusing ratios.

Even for the conical capillary, the dependence of the focusing ratio on the wall atomic number (Fig. 8) is much weaker than expected from the Rutherford scattering cross section. This result also supports the idea that the probability of the scattered proton escaping from the capillary wall predominantly determines the total transmission of MeV protons in the capillary. Because a heavier material has a higher stopping power for projectiles, the escape probability decreases with increasing wall atomic number even though the escape distance is constant. This effect probably reduces the contribution of the scattering cross section to the beam transmission and leads to the saturation of the focusing ratio for heavy-wall materials, as shown in Fig. 8. Note that the focusing ratios calculated for the borosilicate glass and the lead glass fit well on the curves predicted from the ratios for the single element materials when they are plotted with respect not to the effective atomic numbers $Z_{\text{eff}}$ defined in Eq. (4) but to the averaged atomic numbers of the glass materials $Z_{\text{ave}}$ which is calculated by:

$$Z_{\text{ave}} = \sum_i Z_i f(i) \quad \text{(5)}$$

A data point expressed by an open square in Fig. 8 shows the case when the focusing ratio of the lead-glass conical capillary is plotted with respect to the effective atomic number of the lead glass. Clearly, it deviates from the curve predicted by the single elements, showing that the use of the effective atomic number is not so adequate. Since the stopping power is known to be almost proportional to the atomic number of the stopping medium, this result does not contradict the hypothesis that the beam transmission through the capillary depends much more on the stopping power of the capillary wall for the projectile.

6. Conclusions

To investigate the effect of the wall material on the beam transmission through the tapered glass capillary, the energy spectrum and the intensity distributions of protons were measured for borosilicate glass and lead glass capillaries, whose average atomic numbers of the wall are considerably different. Although the increase in the scattered (halo) component intensity was observed for the lead glass capillary, we found that its contribution to the total beam...
transmission is very limited. On the other hand, the MC simulations indicated that the escape distance plays an important role in the beam transmission. Particularly, for the tapered glass capillary having a convex inner wall, the escape distance is relatively large and the probability of the scattered proton escaping from the wall reduces. The MC simulation predicted that when gold is used as the wall material, the use of the constant-taper-angle (conical) capillary improves the focusing ability by a factor of approximately 2. However, we must note that there is a trade-off between the focusing ratio and the beam quality. Therefore, to realize actual applications of the tapered capillary, the capillary wall shape should be carefully designed by taking this trade-off into account.

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