Spatially Resolved Optical Emission Spectroscopy Monitoring of Trace Xenon in an Atmospheric-Pressure Helium Microdischarge

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Abstract

Molten-salt-reactor off-gas is a He stream carrying trace Xe and Kr. A compact DC He microdischarge sustained in the sampled stream converts Xe into line emission, and the Xe content is read from the intensity ratio R = I(Xe I 823.2 nm) / I(He I 706.5 nm). Within the 1.6 mm gap the emission is strongly inhomogeneous, so a ≈0.1 mm line of sight (LOS) was scanned along the axis in 0.25 mm steps for Xe 0–500 ppm and, separately, for pure He and He + 10 ppm N_2. The emitting species do not share one axial profile: the He I lines and the Xe I line peak at both electrodes, whereas the N_2 bands peak in the mid-gap column. Consequently the calibration quality varies with position. The sensitivity is highest in the mid-gap column (x = 0.50–0.75 mm, slope 71 ppm per unit ratio) but there the Xe line responds sub-proportionally and the intercept reaches −22 ppm. In the positive column at x = 1.25 mm the Xe response is proportional and the linearity is best (R^2 = 0.9989, LOD 1.0 ppm), while at the anode glow (x = 1.75 mm) the proportionality is lost again. Position-resolved calibration thus separates the most sensitive LOS from the most linear one and fixes the collection geometry for a low-cost fission-gas monitor.

Experimental set-up

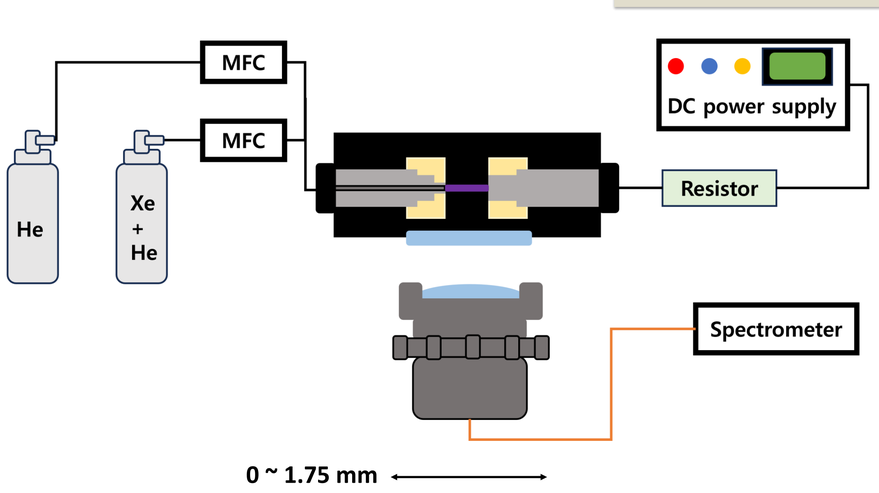


FIG. 1. Schematic of the He-microdischarge Xe monitoring system: hollow-cathode pin (gas inlet), rod anode, MFC-controlled He and Xe/He supply, DC source with ballast, and the side-on lens relay feeding the spectrometer.

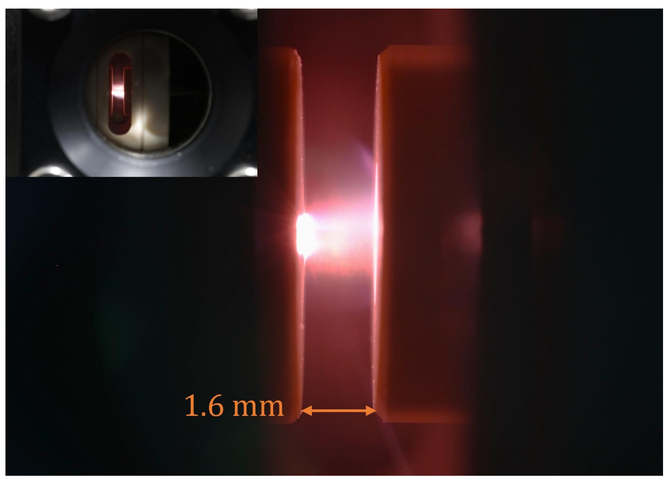


FIG. 2. Side-view photograph of the He microdischarge between the hollow-cathode pin and the anode (gap 1.6 mm); bright glows at both electrodes and a weaker mid-gap column.

* Discharge and gas

- Hollow W cathode pin (bore 0.5 mm = gas inlet) and W anode,
- discharge gap 1.6 mm, 600 sccm flow rate, 102.5 kPa
- He 99.999 % and certified Xe/He mixture: 0–500 ppm Xe in 100 ppm steps
- DC 1.5 mA through a 200 kΩ ballast, 500–600 V normal glow
- Gas-temperature measurement: He + 10 ppm N_2, same cell and lens-coupled optics

* Optics and spectrometer

- Side-on two-lens fiber relay, LOS width ≈ 0.1 mm, scanned in 0.25 mm steps (x = 0 at the cathode-bore end)
- Avantes AvaSpec-NXS2048CL-VA, 348–901 nm, instrumental FWHM 0.4–0.7 nm
- 0.8 s per spectrum, ~180 spectra per LOS position and concentration

* Analysis

- Line areas per frame: Voigt profile + constant background fitted in a ±5 nm window N_2 bands by spline integration
- Calibration ppm = a·R + b over 100–500 ppm at each position, R from frame-wise net areas; LOD = 3σ(R at 0 ppm) × a
- Gas temperature: rotational fit of the N_2 SPS (0,2) 380.5 nm band in He + 10 ppm N_2, 15-frame blocks

Axial distribution of the emitting species in pure He

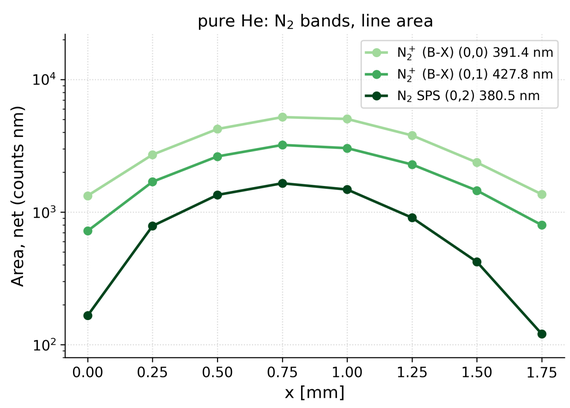


FIG. 3. N_2 bands in pure He (trace impurity): N_2^+ (B–X) 391.4 and 427.8 nm and N_2 SPS 380.5 nm line areas; maximum at x = 0.75 mm, ×4–10 lower at both electrodes.

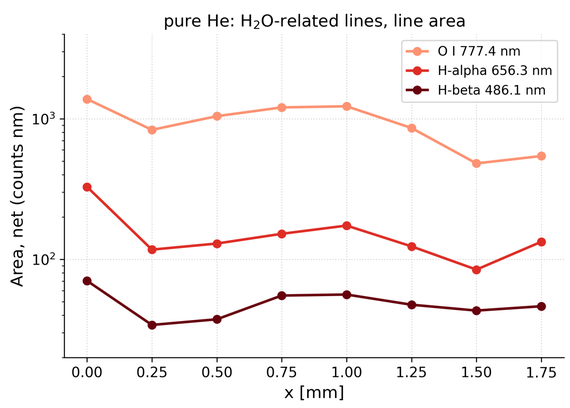


FIG. 4. H_2O-related lines in pure He: O I 777.4 nm, H-alpha and H-beta line areas; dip at x = 0.25 mm, broad maximum at 0.75–1.0 mm.

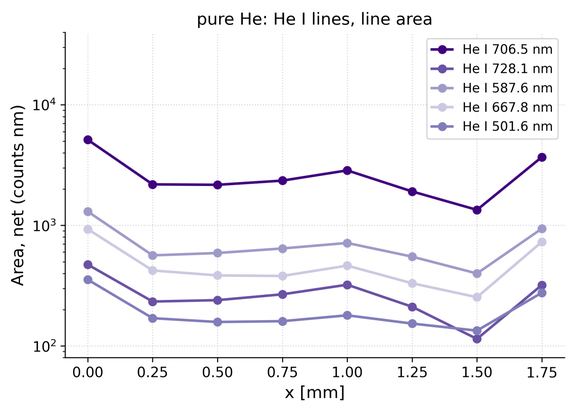


FIG. 5. He I lines in pure He: 706.5, 728.1, 587.6, 667.8 and 501.6 nm line areas; two maxima at the electrodes and a valley between.

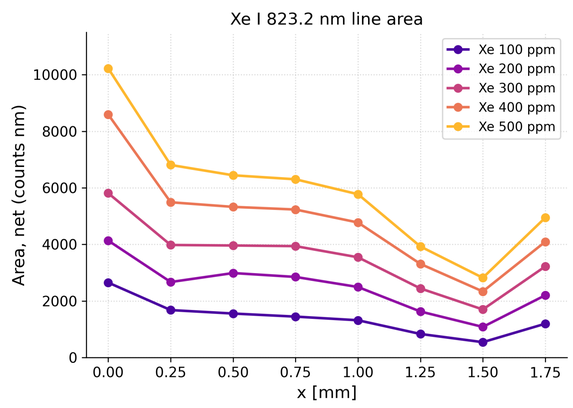


FIG. 6. Xe I 823.2 nm line area at 100–500 ppm Xe: the He I shape at every concentration.

Position-resolved calibration quality and discharge structure

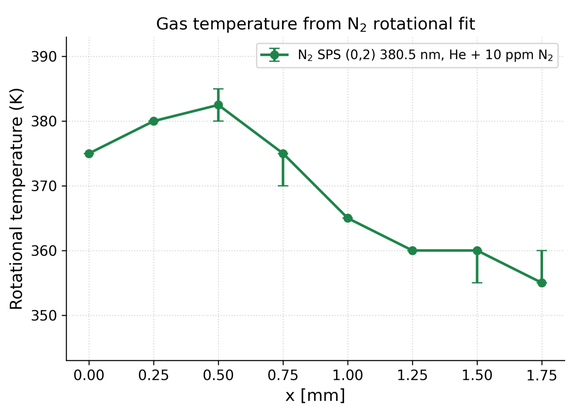


FIG. 7. Rotational temperature from the N_2 SPS (0,2) 380.5 nm band in He + 10 ppm N_2 (median and quartiles of 15-frame blocks): 355–383 K along the axis.

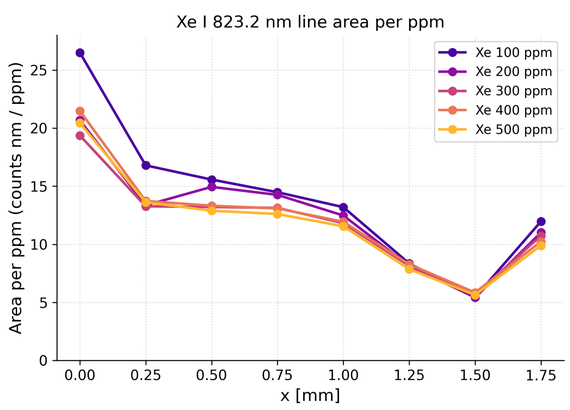


FIG. 8. Xe I 823.2 nm line area per ppm: the curves separate near the bore and at the anode, and coincide at x = 1.25–1.50 mm.

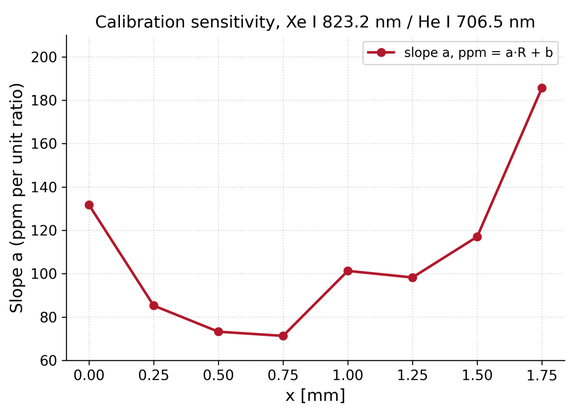


FIG. 9. Slope a of ppm = a·R + b, R = Xe I 823.2 / He I 706.5: minimum 71 at x = 0.50–0.75 mm, maximum 186 at 1.75 mm.

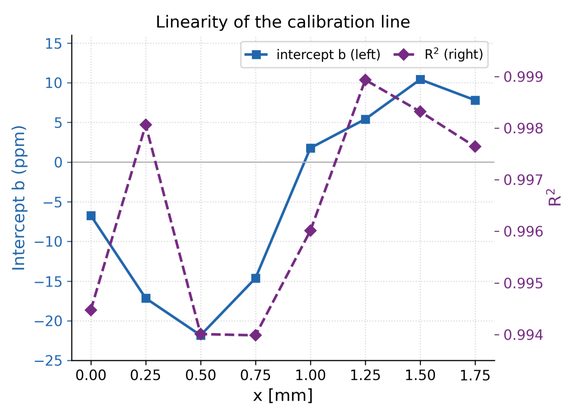


FIG. 10. Intercept b (left) and R^2 (right) of the calibration line at each position: b crosses 0 at x = 1.0 mm, R^2 is best at 1.25 mm.

Results

* Axial distribution of the emitting species (pure He)

- He I 706.5 nm: two maxima at the electrodes (x = 0 and 1.75 mm) and a valley in between (minimum at 1.5 mm, ×3.8 lower); the other He I lines follow the same shape.
- N_2^+ (B–X) 391.4 / 427.8 nm and N_2 SPS 380.5 nm: the opposite shape, a maximum at x = 0.75 mm and ×4–10 lower at both electrodes.
- O I 777.4 nm and H-alpha: intermediate, a dip at x = 0.25 mm, a broad maximum at 0.75–1.0 mm and a minimum at 1.5 mm.
- Xe I 823.2 nm (100–500 ppm) follows the He I shape at every concentration.

* Position-resolved calibration and discharge structure

- The Xe I 823.2 nm area per ppm separates by concentration near the bore (26 → 20 counts·nm/ppm from 100 to 500 ppm at x = 0) and coincides at x = 1.25–1.50 mm; at x = 1.75 mm it separates again.
- Slope a: minimum 71 ppm per unit ratio at x = 0.50–0.75 mm (highest sensitivity), maximum 186 at x = 1.75 mm.
- Intercept b: −22 ppm at x = 0.5 mm, crossing 0 at x = 1.0 mm, +10 ppm at 1.5 mm. R^2 is best at x = 1.25 mm (0.9989) and worst at 0.50–0.75 mm (0.9940).
- Gas temperature (N_2 rotational, He + 10 ppm N_2): 355–383 K along the whole axis; the position dependence is not thermal.
- Tentative assignment to the classical DC glow: x = 0 cathode glow / negative glow, 0.50–0.75 mm Faraday dark space, 1.0–1.5 mm positive column, 1.75 mm anode glow.

Conclusions and future work

- In the He microdischarge the emitting species are spatially separated: He I and Xe I are strongest at the electrode glows, the N_2 bands in the mid-gap column. A single LOS therefore samples a different excitation balance at each position.
- The calibration quality follows this structure. The Faraday-dark-space region gives the highest sensitivity but a sub-proportional Xe response (intercept −22 ppm, R^2 0.994); the positive column gives a proportional response and the best linearity; at the anode glow the proportionality is lost again.
- Sweet spot: x ≈ 1.25 mm, where the positive column ends and the anode glow has not yet begun. Sensor robustness is defined by calibration linearity (R^2 0.9989, LOD 1.0 ppm), not by constancy of the He I reference line, which itself responds to Xe and N_2 admixtures.
- Future work: confirm the sub-proportional first-100-ppm response near the inlet (randomized concentration order, 0.1 mm re-scan), a ≤ 1 ppm N_2 admixture with band-shape separation, and extension to Kr and mixed fission-gas streams.

References

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