A Closed-Cycle Immersion Cell Platform for Noble Triple Points

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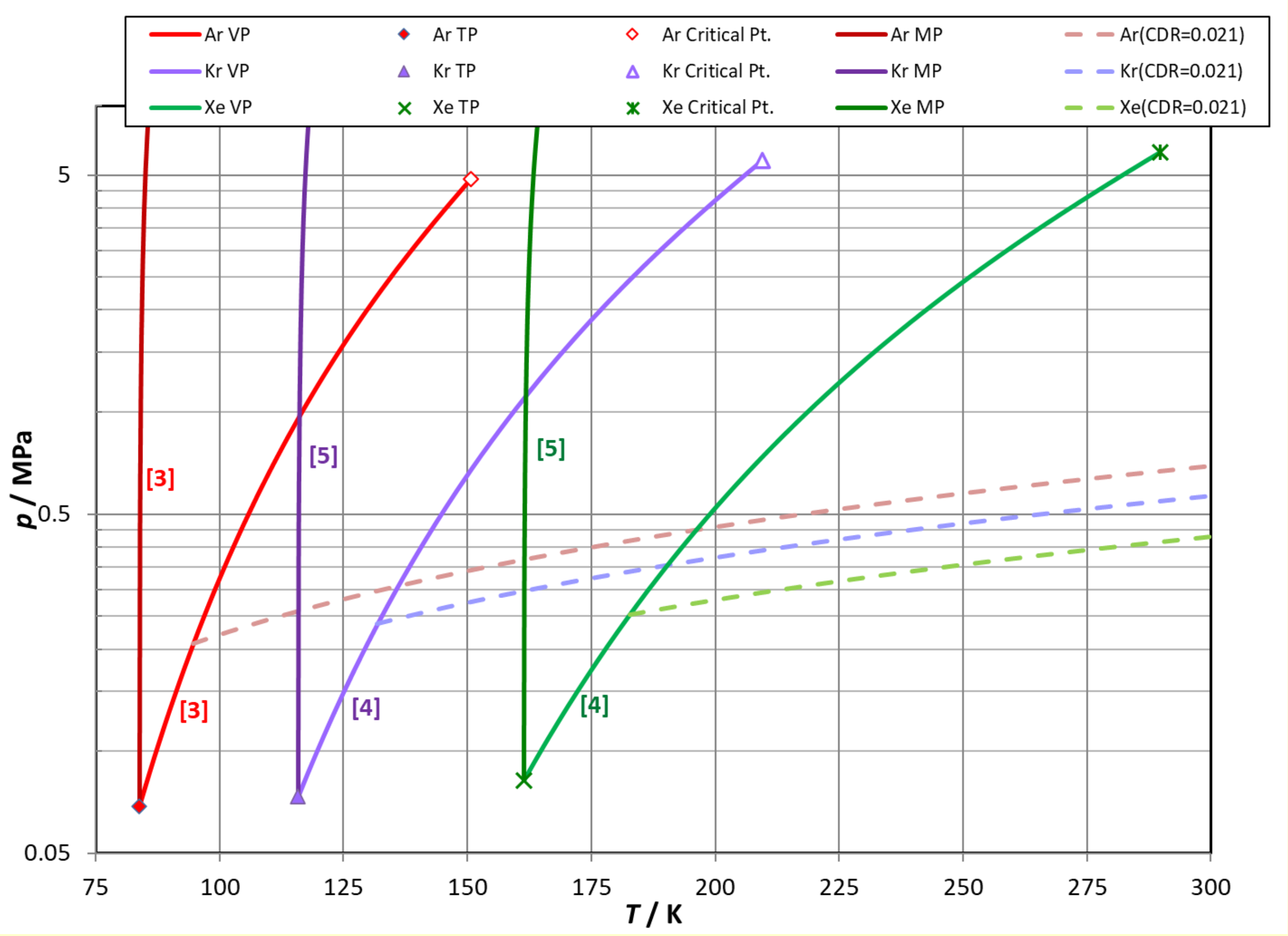
Abstract / Introduction

New technologies are needed for maintenance systems to support non-metal fixed points below 200 K using immersion-type cells. In addition, we are unaware of any immersion-type Xe triple point (TP) cell in the scientific literature. We have developed a new closed-cycle refrigeration platform for the storage, condensation, freezing and controlled melting of noble gas solids for direct realization of their TP temperatures in an immersion-type cell. The system accommodates long-stem standard platinum resistance thermometers (SPRTs) through a single 7.75 mm inner diameter thermowell that is continuously purged with helium gas and thermally coupled to four separate control zones and strongly coupled to the noble-gas condensate. Three other capsule-type SPRT thermowells are accessible from the interior vacuum space.

The system design incorporates several innovative features as well as some derived from a legacy argon system designed by Furukawa [1] which was used at NIST for more than 30 years prior to it's decommissioning. This new patented design [2] is based on two compact single-stage Stirling cryocoolers and does not utilize liquid nitrogen for maintaining the melt conditions. A four-zone control arrangement is implemented with active cooling applied to the first two outer zones via the two cryocoolers while two other inner zones are passively cooled and actively controlled via heating only. The Stirling coolers are soft-mounted using bellows connections and thermally coupled to the two outermost zones via flexible copper wire-rope straps which serve to decouple vibration transmission from the Stirling's linear drive motors operating at 60 Hz.

The cell holds approximately 210 mL of noble condensate, either argon, krypton or xenon, and the integral expansion ballast volume of 21 L produces a critical density ratio of 0.021, sufficient to keep pressures below 6 bar at ambient conditions. The gas manifold provides both pressure and mass flow rate regulation and is capable of controlling the rate of gas condensation and monitoring the rate of gas expansion as the system changes temperature. A single vacuum chamber encloses the four nested control zones and the cell. Testing has been conducted using a set of diagnostic diode thermometers dispersed across various points in all zones. Initial results indicate the system is suitable for Xe and Kr TP realizations but that additional refinements in the thermal strap configurations are needed to fully support Ar TP realizations.

Noble Gas Thermophysical Properties



Phase Diagrams for Ar, Kr, and Xe. (above) Showing gas-phase isochors with critical density ratio CDR=0.021 **Calculated values** (REFPROP, below) for thermophysical properties at the TPs. The melting line equation for Ar from [3] predicts a value for the head correction coefficient that is 5.5 % larger than the ITS-90 value (3.3 mK/m). The melting line data from [5] are inconsistent with the Clapeyron Equation {d*p*/d*T*} for Kr (7 %) and Xe (~30 %) near their triple point temperatures. The molar enthalpy values are taken from [6].

	T_{tp}	p_{tp}	ρ_l	ρ_s	Δh_f	dp_m/dT	dT/dz
	(K)	(MPa)	(kg/m^3)	(kg/m^3)	(J/mol)	(MPa/K)	mK/m
Ar[3]	83.806	0.06889	1416.8	1622	1190	3.98	3.48
Kr[4,5]	115.78	0.10675	2447	2824	1640	3.32{3.10}	7.2 {7.7}
Xe[4,5]	161.41	0.0818	2961	3545	2315	2.57{1.96}	11.3{14.8}

Design and Construction

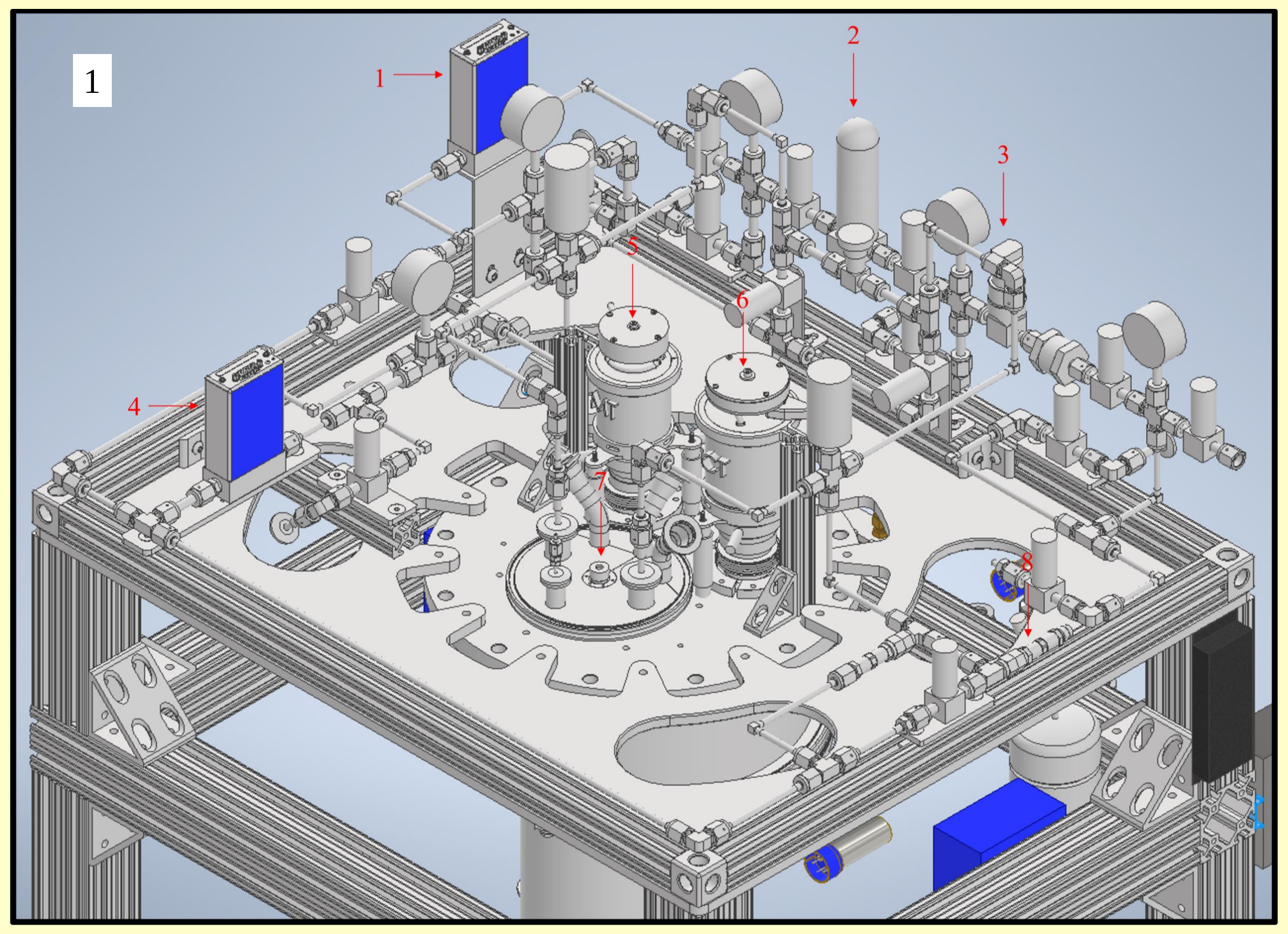


Fig. 1. (above) Key features of the platform. The top mounted high-purity gas manifold is constructed of orbital-welded 6.35mm stainless steel tubing and all-metal gasket fittings. The manifold (a schematic is shown in Fig. 7 below) is equipped with a mass flow controller (1), gas purifier (2), pressure regulator (3), mass flow meter (4), and two capacitance diaphragm gauges (CDGs) which are connected to two independent gas fill lines connecting the manifold to the cell. A series of 15 valves provide control and isolation, with check valves (8) for over-pressure relief. Continuous controlled cooling is provided via two Stirling-type cryocoolers, an 80 W (5), and a 160 W (6) (input power), operating in tandem. A central thermowell (7) provides direct access to the TP cell.

Fig. 2. An unmounted Stirling cryocooler.

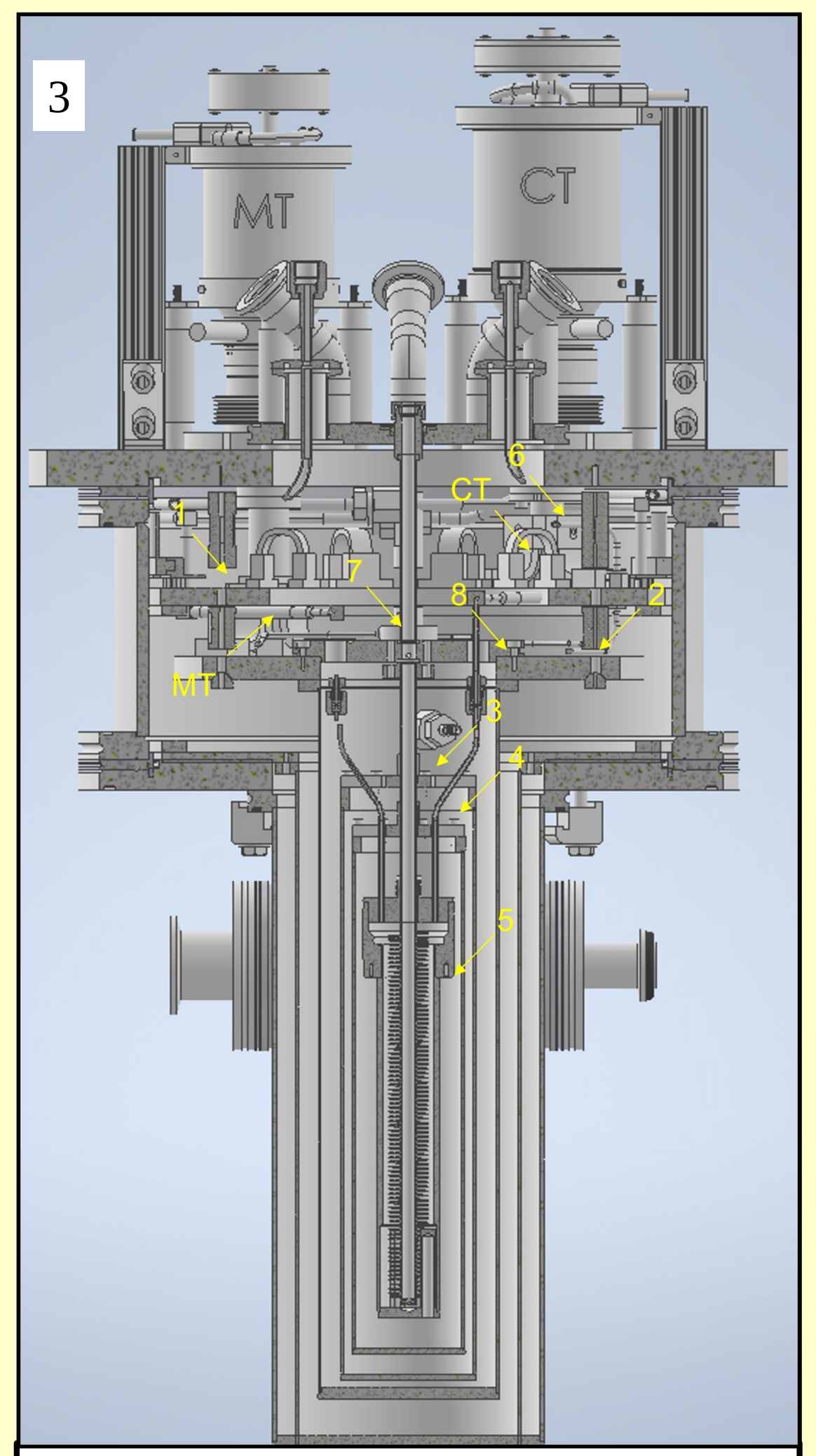
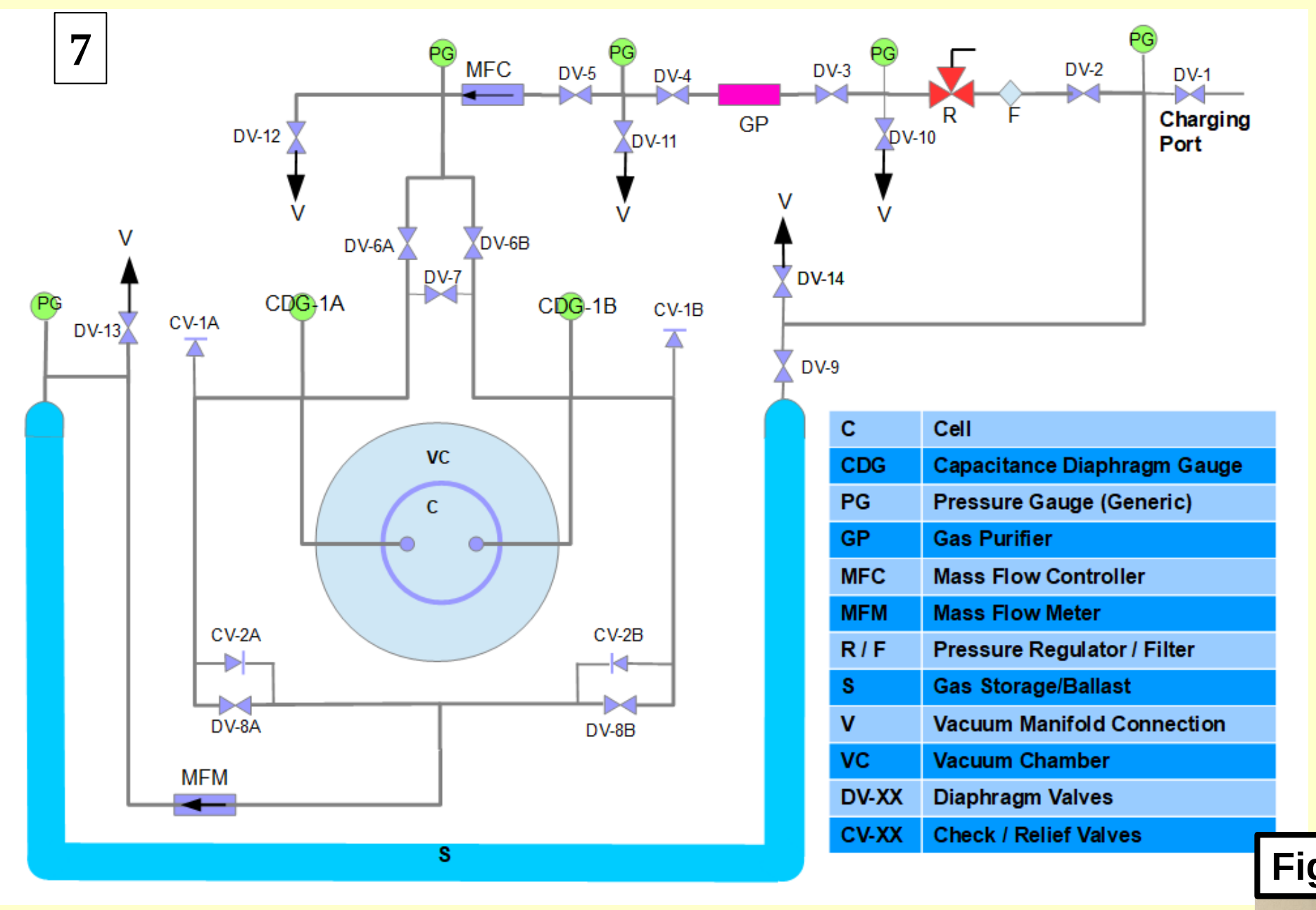


Fig. 3. (above) Cross-sectional diagram of the vacuum chamber showing locations of the diagnostic silicon diodes (1-8) and Pt100 thermometers (MT & CT).

Fig. 4. (Left) The TP cell. Shown are the two gas fill lines, 3rd zone and 4th zone anchor plates, thermowell, and burst disc.



C	Cell
CDG	Capacitance Diaphragm Gauge
PG	Pressure Gauge (Generic)
GP	Gas Purifier
MFC	Mass Flow Controller
MFM	Mass Flow Meter
R / F	Pressure Regulator / Filter
S	Gas Storage/Ballast
V	Vacuum Manifold Connection
VC	Vacuum Chamber
DV-XX	Diaphragm Valves
CV-XX	Check / Relief Valves

Fig. 5. (below) The complete assembled system as viewed from the front.

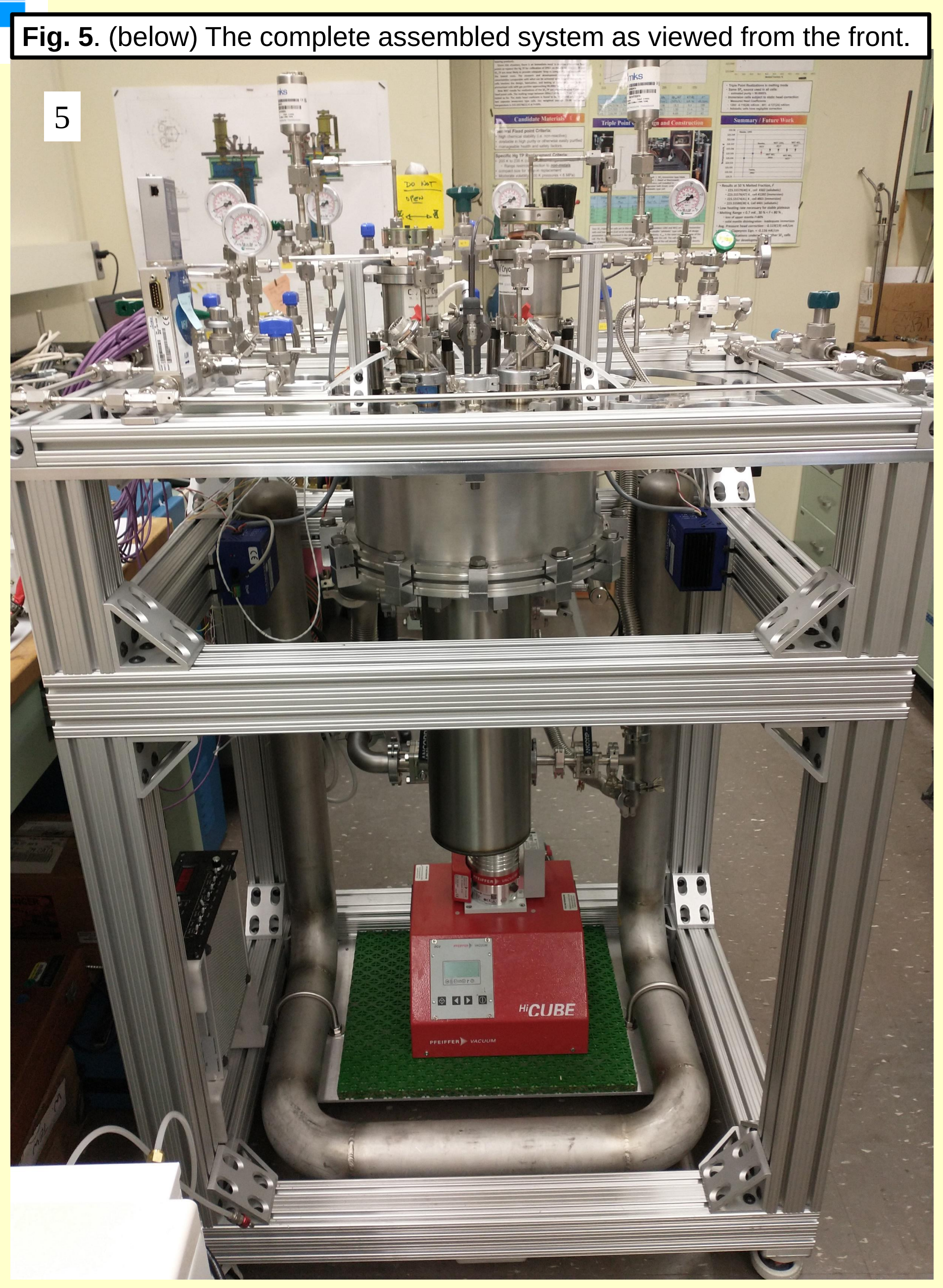
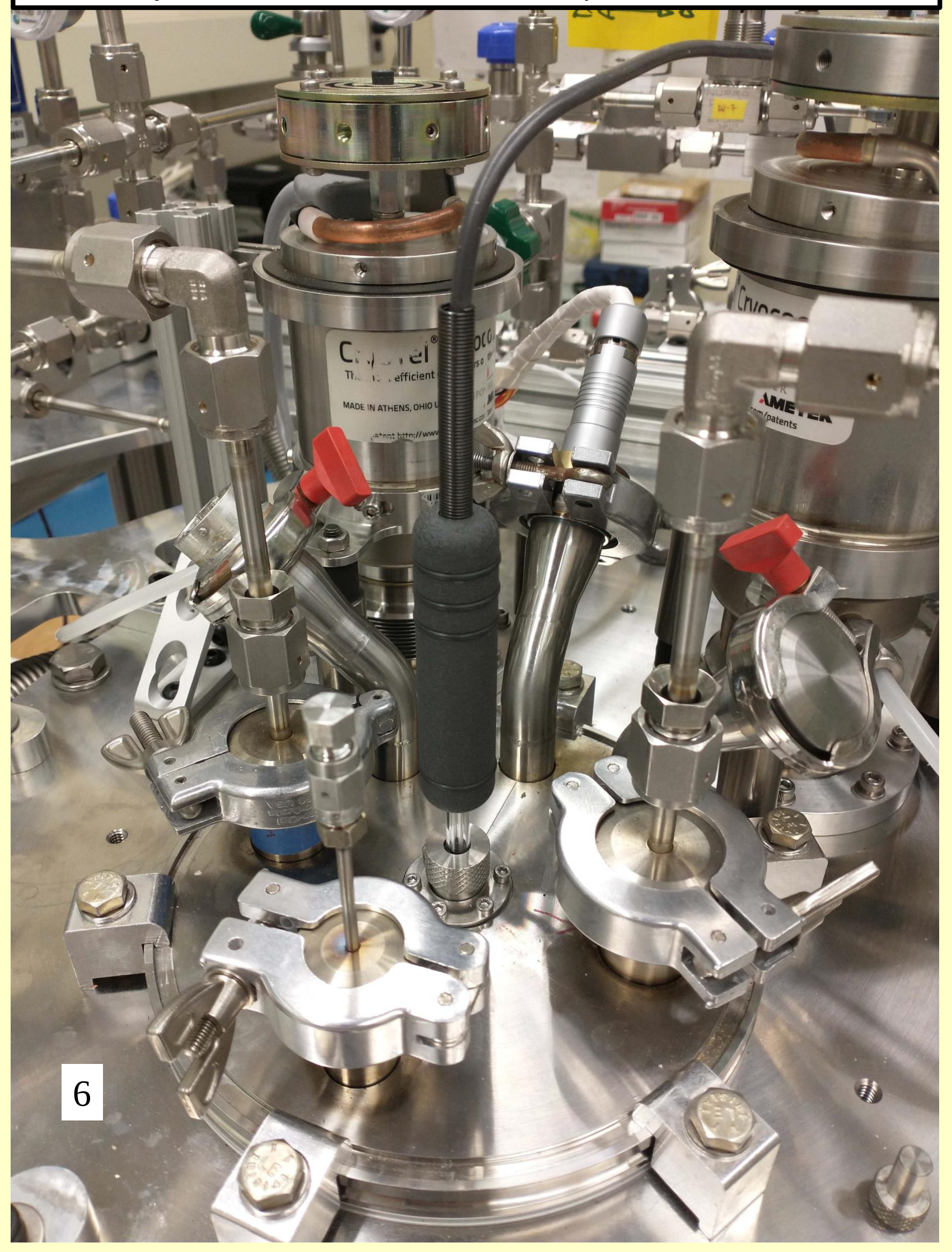
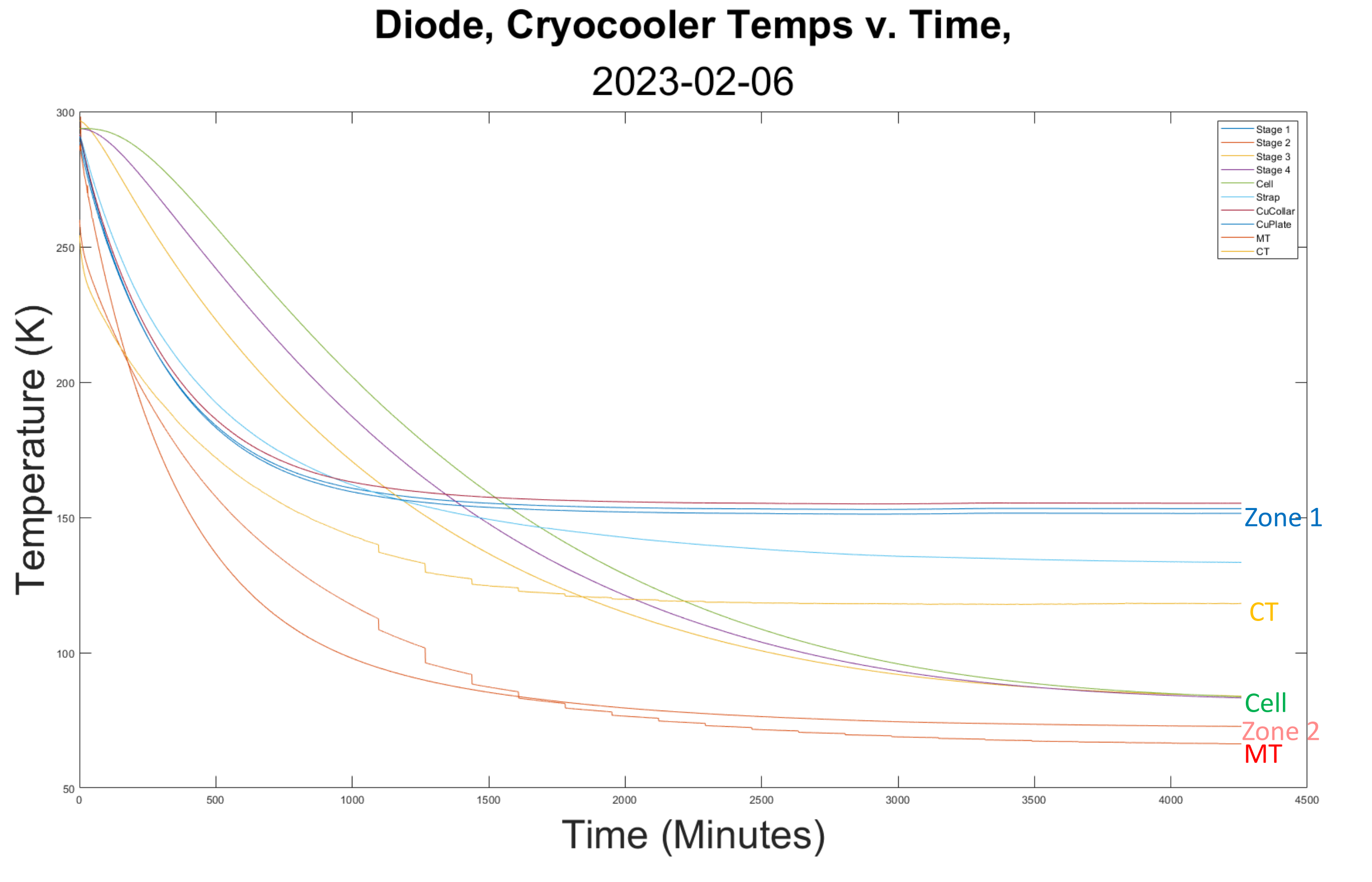


Fig. 6. (below) The view showing the thermowell and a long stem SPRT fully inserted at a total insertion depth of 50 cm.



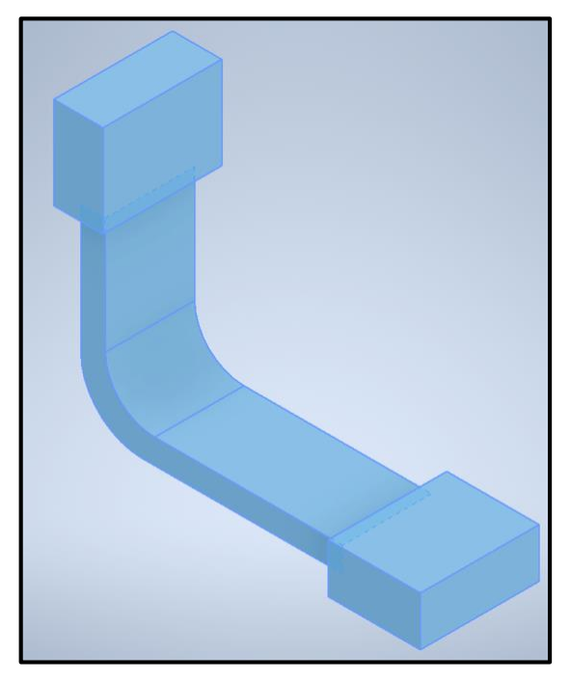
Testing Results

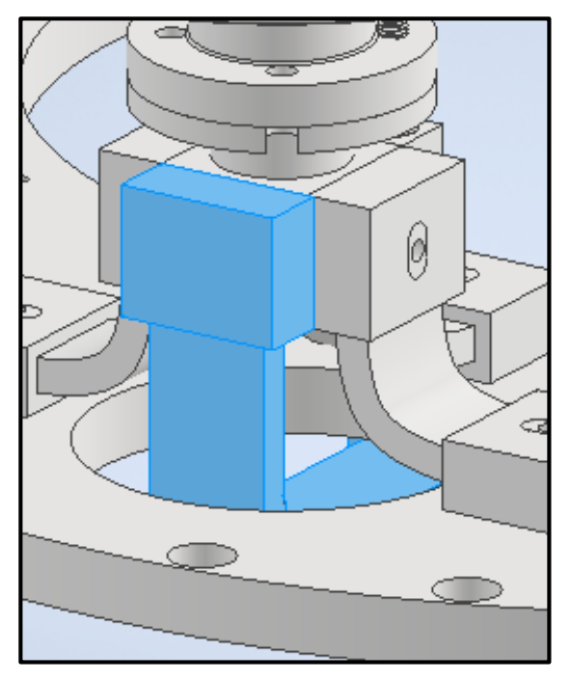


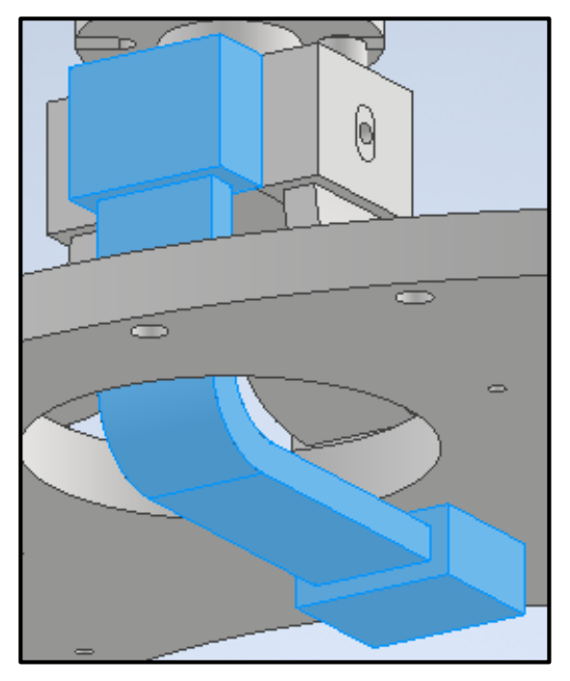
RTD/Diode	***Location***	***Temperature***
MT	**MT Stirling Cold Tip**	**63.3 (0.7) K**
CT	**CT Stirling Cold Tip**	**118.2 (0.5) K**
1	**Zone 1 Top Plate**	**153.3 (1.0) K**
2	**Zone 2 Base Plate**	**72.8 (1.0) K**
3	**Zone 3 anchor plate**	**83.9 (1.0) K**
4	**Zone 4 anchor plate**	**83.3 (1.0) K**
5	**TP Cell**	**83.7 (1.0) K**
6	**Gas fill line**	**133.4 (1.0) K**
7	**Thermowell Clamp**	**155.2 (1.0) K**
8	**Copper Plate**	**151.5 (1.0) K**

Summary / Future Work

Near-term modifications. Available cooling power to zone 1 is limited by copper wire-rope strap conductance, an additional thermal strap will be fabricated and installed to augment the effective thermal conductance between the CT absorber and zone 1 plate.







Summary

Compact Stirling cryocoolers are suitable for implementing multi-zone refrigeration platforms for the realization of noble gas or other non-metal triple points in the range 70 K to 200 K, below that of conventional refrigeration cycles. The approach conveys certain key advantages over systems based on liquid nitrogen pool-boiling cryostats:

- The cooling is continuous and controllable in closed-loop operation
- Avoids drift and perturbations associated with liq N_2 boil-off / refilling cycles
- Zones are cooled to any temperature as close to the TP temperatures as needed
- Zone settings are not constrained by liq N_2 vapor pressure
- Avoids the high heating or high pressure requirements of liq N_2 systems

Caveat: Improved melting line equations and/or data may be needed for Kr and Xe to enable accurate head corrections for those TPs via immersion cell realizations.

References

1. G.T. Furukawa, in Temperature: Its Measurement and Control in Science and Industry, vol. 6, ed. by J.F. Schooley (AIP, New York, 1992), pp. 265–269
2. Triple point immersion cell article, WL Tew, JR Nanninga - US Patent 11,204,288, 2021
3. Tegeler, Ch., Span, R., and Wagner, W., J. Phys. Chem. Ref. Data, 28(3):779-850, 1999.
4. Lemmon, E.W. and Span, R., J. Chem. Eng. Data, 51(3):785-850, 2006. doi: 10.1021/je050186n
5. Michels, A. and Prins, C., Physica, 28:101-116, 1962.
6. Pavese, F and Molinar, G, 1992, Springer, pp 471-475, https://doi.org/10.1007/978-1-4757-5869-6