Additional Supporting Information for

Toward the Synthesis of Wafer-Scale Single-Crystal Graphene on Copper Foils

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Safety Design Considerations for Using Hydrogen at 2 Atmospheres of Pressure in a Glass CVD Apparatus. If a standard CVD tube furnace is not properly equipped for the 2 atm operation as described in this paper,¹ there is the risk of separation from the end-caps, quartz tube rupture, and/or hydrogen ignition. The authors therefore provide this additional safety information.

The CVD apparatus described¹ uses a 25 mm fused quartz tube with an O-ring compression seal on each end. Since the O-ring offers little resistance to sliding, the fitting must have additional support to resist the overpressure. The system is tested at 0.7 atm over the 2 atm absolute operating pressure; this corresponds to a maximum of 1.7 atm overpressure. The thrust on the end fittings will be slightly less than the equivalent of a 10 kg weight pulling on them in the axial direction.

Each of the two 25 mm O-ring compression fittings has a 3/4 inch (19 mm) diameter stainless steel tube welded to it at right angles, and this tube is mounted in a horizontal position relative to the sturdy steel optical table that supports the CVD furnace. The exhaust side is rigidly attached to the table. There are two 3/4 inch (19 mm) aluminum posts screwed to the table, which then use two standard chemistry rod clamps to firmly grip the welded tube (Figure S1).

Figure S1. Photo of the fixed gas exit support. The 25 mm quartz tube is at the right, covered by a brass wire screen. This fits into an O-ring sealed stainless steel adapter, with a $\frac{3}{4}$ inch (19 mm) tube welded onto the side. This is in turn held by two clamps on two $\frac{3}{4}$ inch (19 mm) aluminum posts that are screwed into the steel top of the optical table, which provides rigid mounting.

It is desirable that the gas input 25 mm O-ring compression fitting is movable so that the fused quartz tube can be readily removed for cleaning; hence the support at this site is secured to the table with a pair of magnetic bases (Figure S2).

Figure S2. A photo of the removable gas inlet support.

The 25 mm quartz tube enters the CVD furnace on the right, and it fits into a similar O-ring sealed adapter that also has a $\frac{3}{4}$ inch (19 mm) stainless tube welded to the adapter. A heavy-duty frame clamp is used to connect to the 1 inch (25.4mm) diameter black aluminum post. The aluminum post is anchored to an aluminum plate that is parallel to the quartz tube axis, and which in turn is attached to two switchable magnetic bases ca. 100 mm apart to provide stability against the outward thrust of the O-ring sealed end fitting when under pressure. The plate must

fit well into the tops of the magnetic bases to avoid even the slightest tilt of one of the bases, which would compromise the holding force. The magnetic bases are approximately 50 mm on a side, and are standard components available from companies that sell optical tables (Newport Corp. or Thorlabs, Inc.).² Once the two magnets are switched off, the post and compression fitting is easily removed as a unit.

The 25 mm OD tube has a standard 1.5 mm wall thickness. Although the tube is operated at 2 atm absolute, it was tested with to an overpressure of 1.7 atm with inert gas. The exposed portions of the tube that are not inside the furnace are covered with sturdy brass wire screen when it is used in an overpressure condition (Figure S1). A hazard is the potential for quartz tube fragments to project if an explosion occurs, hence the use of the safety screen.

If the tube failed at the end, ignition of the hydrogen is unlikely and it would rise to the ceiling and be exhausted by the lab ventilation. If it failed inside the furnace, the hydrogen would burn off in a few seconds, but the fuel value is small so that it would be little more than a momentary flash. Since there is no oxygen inside the quartz tube, it would just burn off as it exited into the furnace where it is already very hot. The system volume is about 1.3 L, or about 100 mg of hydrogen, which is the about the amount of hydrogen that would escape if the tube cracked. However, with our robust mounting system, there has never been a tube or end-joint failure.

Since some researchers use 50 mm OD quartz tubes in their CVD reactors, they need to be mindful that the forces are multiplied four-fold if the same hydrogen overpressure is used, and the design must be robust enough to resist these much greater forces, including the need for a thick wall quartz tube. Unless the table is very sturdy, an independent rigid support-frame is desirable that will not deform under applied forces equal to a mass of ~40 kg with ≤ 2 atm

overpressure (3 atm absolute). This should be checked by a competent device technician before use.

References

- (1) Yan, Z.; Lin, J.; Peng, Z.; Sun, Z.; Zhu, Y.; Li, L.; Xiang, C.; Samuel, E. L.; Kittrell, C.; Tour, J. M. Toward the Synthesis of Wafer-Scale Single-Crystal Graphene on Copper Foils. *ACS Nano* **2012**, 6, 9110–9117. DOI: 10.1021/nn303352k
- (2) Vendors for magnetic bases: Model MB-2 Magnetic Base from Newport Corporation, 1791 Deere Avenue, Irvine, CA 92606. Model MB175 Magnetic Base from Thorlabs Inc, 56 Sparta Avenue, Newton, New Jersey 07860.