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DEVELOPMENT OF A MULTI-FUNCTIONAL CHEMICAL VAPOR DEPOSITION SYSTEM FOR THE SYNTHESIS OF CARBON NANOSTRUCTURES

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Abstract: Carbon nanostructure synthesis has been extensively researched recently due to its potential applications in energy storage, drug delivery, imaging, composite materials, field emitters, Nano-electronic devices, EMI shielding, hydrogen storage Nano tanks, functional polymers, sensor materials, and material reinforcement. Vapor deposition is the most common technique used to synthesize carbon Nanostructures, including thin layers, nanotubes, and other Nano assemblies. Chemical vapor deposition is one of the most versatile methods among them due to its ability to scale up to an industrial scale. In this paper, a chemical vapor deposition (CVD) system was designed and developed in-house, and improvements were added. A separate reaction chamber, eternal solvent vapor introduction method, pre-heating of input gas, and the facile sample handling method can be listed as an improvement to the system. This system effectively synthesized carbon nanotubes (CNTs), carbon thin films, and other carbon structures.

Index Terms: vapor deposition technologies, chemical vapor deposition, carbon nanotubes, Spinnable carbon nanotubes

1 INTRODUCTION

Scientists throughout the world research Carbon nanotube (CNT) production processes due to the wide range of applications they can be used [1], [2]. However, the large-scale synthesis of CNT has many constraints [3]. Vapour deposition is the leading technology that is used to synthesize CNT [4].

Vapour deposition is classified as a surface coating method for generating thin layers. Vapour deposition techniques have a wide variety as atomistic deposition, specific deposition, and full thickness deposition. This atomic deposition can be divided into physical vapor deposition (PVD), ion beam-assisted vapor deposition (IBAD), and chemical vapor deposition (CVD) [4], [5], [6], [7], [8], [9]. The basic sketches of the different technologies described above are given in Fig. 1.



Fig. 1. Schematic diagrams of Atomistic deposition methods (a) physical vapor deposition (PVD), (b) ion beam assisted vapor deposition (IBAD), and (c) chemical vapor deposition (CVD).

In this research, we have used CVD to synthesize carbon nanotube structures. CVD's industrial application may be traced back to a patent published in 1893 by de Lodyguine, who deposited W onto carbon lamp filaments by reducing WCl₆ with H₂. As previously said, CVD technology has grown in importance in recent years. It is well suited for creating a wide range of innovative goods, including bulk materials, composites, coatings, and films [9].

CVD is the most versatile vapor deposition technology that can be developed on an industrial scale. For example, multi-walled carbon nanotube properties can be varied by adjusting the reaction conditions of the CVD system, like temperature, gas flows, and vacuum levels [5]. [6], [7], [8].

CVD methods can be classified according to operating conditions like atmospheric pressure CVD, low-pressure CVD, ultrahigh vacuum CVD, and sub-atmospheric CVD. This CVD method is also classified based on its physical characteristics of vapor and type of substrate heating (activated environment). Plasma methods, Atomic-layer CVD, Hot filament CVD, and Photo-initiated CVD are some other methods of CVD [6], [9], [10].

Default CVD design

The CVD system's default design comprises of feed gas unit, mass flow controllers (MFC), a furnace with a reaction chamber, and a vacuum pump (Fig.2).



Fig. 2. (a) Schematic diagram of a typical laboratory CVD equipment (i) chemical vapor precursor supply system, (ii) CVD reactor, and (iii) effluent gas handling system, and (b) process principles and deposition mechanism. This illustration depicts only the gaseous state precursor system. As described in the text, additional assemblies convert solid and liquid materials to the gas phase.

However, there is no universal setup, and the equipment can be tailored for synthesizing different coating materials and substrate geometry. In general, the items mentioned above can be included in the three main components of the CVD system. [9]

- i. Chemical vapor precursor supply system (feed gas unit, Mass flow controllers)
- ii. CVD reactor (furnace with a reaction chamber)
- iii. Effluent gas handling system (vacuum pumps, cold traps)

The chemical vapor precursor supply system produces and distributes vapor precursors to the reactor (Fig. 2.a.i). The CVD reactor's architecture is determined by whether the starting material is solid, liquid, or gas. The solid's surface area and contact time determine a solid precursor's sublimation. A bubbler is used to evaporate the reactants in liquid sources, and a carrier gas (reactive gases $[H_2]$ or inert gases [Ar]) is used to transfer the vaporized reactants into the reactor. The temperature of the source, the carrier gas flow rate, and the pressure above the source all influence reactant delivery. If the liquid precursor's vapor pressure is known, its partial pressure can be calculated and controlled by adjusting the carrier gas's volume and flow rate. The vapor precursor produced is frequently fed into the CVD reactor. Another method for vaporizing liquids is flash evaporation. The liquid precursor is delivered to the flash evaporator through a syringe pump or peristaltic pump, where it is instantly evaporated. When it comes to gas precursors, high purity (e.g., 99.99 %), gaseous reactants (C₂H₂), or carrier gases are introduced into the reactor using flow meters (e.g., mass flow meters) and control systems from high-

pressure gas cylinders. Before entering the reactor, these gases are frequently purified using purification columns to eliminate any oxygen, moisture, or pollutants picked up in the gas transfer lines.

A CVD reactor comprises a reaction chamber with a load-lock for transporting and placing the substrate into the chamber, a substrate holder, and a temperature-controlled heating system (Fig. 2.a.ii). The CVD reactor's principal duty is to heat the substrate to the desired deposition temperature. A hot-wall or cold-wall CVD reactor can be used. In a hot-wall reactor, the substrates are placed in a heated furnace for indirect heating. A three-zone resistively heated furnace is widely used to manage the outer zones while maintaining a consistent temperature profile in the middle deposition zone. The temperature is usually recorded using a thermocouple. The majority of CVD processes are endothermic. As a result, the deposition will take place on the heated substrate with minimal deposition on the reactor wall. Though these reactors are complex designs, they allow more control over the deposition process, reducing deposits on the reactor walls and reactant depletion. However, thermal convection can create a concentration gradient of reactive species in a cold wall reactor, resulting in non-uniform coatings. This constraint can be solved by using a lower pressure for CVD cold wall deposition. The size and shape of the substrate and whether it is conducting or non-conducting all influence the heating process.

The effluent gas handling system comprises an exhaust gas neutralizer and a vacuum system that provides the required low pressure for the CVD process, which operates at low pressure or high vacuum during deposition (Fig. 2.a.iii). Low-pressure CVD, in general, requires a less complicated vacuum system than the PVD process. A corrosion-resistant/chemical-resistant rotary or vane pump is used in low-pressure CVD. The effluent gas handling system's primary function is to securely remove hazardous by-products and toxic unreacted precursors. Unreacted precursors and corrosive by-products like HCl are neutralized or caught with a liquid nitrogen trap (cold trap) to prevent these gases from accessing the rotary or diffusion pump and causing damage. Hydrogen and other flammable gases are extinguished. Deposition efficiency impacts the amount of recycling and disposable methods used. Corrosive, toxic, poisonous, hygroscopic, inflammable, quickly oxidizing, and high vapor pressure, CVD reactants, and products are common. As a result, the reactor system's post-deposition portion must effectively render these compounds harmless before releasing them into the environment. Toxic gas sensors must be fitted, and special measures must be followed. These compounds' disposal and safety practices have been reported.

Solid particles that originate in the homogenous gas-phase process (Fig. 2.b.8) and are subsequently transported in the exhaust system may potentially cause abrasion difficulties. Particle filtration systems are frequently applied to prevent particle emissions. Different CVD systems may use different chemical precursors and reactions, which is a crucial point to remember. As a result, the effluent gas management system must be tailored to the specific CVD system [9], [21], [22], [23].

PROCESS PRINCIPLES CHEMISTRY OF CVD

The following main steps are involved in the CVD process in general.

(1) Providing gas reactants (feed gas) or producing active gaseous reactant species (catalyst vapor) from solids and liquids (Fig. 2.b.1).

(2) Gaseous species transport into the reaction chamber. However, when the catalyst powder is deposited in the reactor vessel, this step occurs in the same reactor vessel (Fig. 2.b.2).

(3) Gaseous reactants create intermediate species as a result of gas-phase reactions (Fig. 2.b.3).

(a) A homogeneous gas-phase reaction can occur at a high temperature above the decomposition temperatures of intermediate species inside the reactor. Intermediate species experience subsequent decomposition and chemical reactions, generating powders and volatile by-products in the gas phase. The powder will accumulate on the substrate surface and act as crystallization centers, while the by-

products will be carried out of the deposition chamber. The deposited film, on the other hand, may have weak adherence.

(b) Diffusion/convection of the intermediate species over the boundary layer (a thin layer close to the substrate surface) occurs at temperatures below the dissociation of the intermediate phase. Following that, these intermediate species progress through stages 4 through 8.

(4) Gaseous reactants are absorbed onto the heated substrate, and a heterogeneous reaction takes place at the gas-solid interface (i.e., heated substrate), resulting in the deposit and by-product species (Fig. 2.b.4).

(5) The deposits diffuse across the heated substrate surface, generating a crystallization core and film growth, followed by creating additional structures (Fig. 2.b.5).

(6) Gaseous by-products are diffused or convectively eliminated from the boundary layer (Fig. 2.b.6).

(7) Gaseous by-products of the 3a and 5 processes (Fig. 2.b.7).

(8) Unreacted gaseous precursors, catalyst vapor, and by-products are removed from the deposition chamber (Fig. 2.b.8).

Both catalyst materials and feed gas proceed through stages 1-8, as previously stated. Initially, the catalyst particles are subjected to the procedures mentioned above, followed by the input gas. For example, FeCl₂ catalyst deposition occurs first, followed by C_2H_2 breakdown on the catalyzed substrate to form CNTs. The major CVD processes during the deposition are schematically illustrated in Fig. 2. The process parameters for the deposition of thick films and coatings are adjusted to favor the heterogeneous reaction. A combination of heterogeneous and homogeneous gas-phase processes is preferable for the deposition of porous coatings.

Metals, metal hydrides, halides, and halo hydrides are common precursors (e.g., FeCl₂) used in the CVD process. These materials, on the other hand, must be stable at room temperature. The precursor material selection requirements include low vaporization temperature, high saturation vapor pressure, steady vapor generation, and appropriate deposition rates. In addition, low toxicity, cost-effectiveness, and availability are considered in industrial applications. Thermal decomposition (pyrolysis), reduction, oxidation, hydrolysis, nitridation, disproportionation, and synthesis are the most common chemical reactions in the CVD process [9], [21] to [23].

Applications of chemical vapor deposition

In a nutshell, CVD technology's current applications can be divided into four categories: extraction and pyrometallurgy, electronic and optoelectronic materials, surface modification coatings, ceramic fibers, and ceramic matrix composites (CMCs).

Semiconductors (e.g., Si, Ge, III–V, II-VI), integrated circuits (I.C.s) for microelectronics, optoelectronics, energy conversion devices (e.g., solar cells), dielectrics (e.g., SiO₂, AlN, Si₃N₄) for microelectronics, metallic films (e.g., W, Pt, Mo, Al, Cu, Ru) for electronics, refractory ceramic materials (e.g., TiB₂, SiC, B₄C, B.N., TiN, Al₂O₃, ZrO₂, MoSi₂) used for hard coatings, wear and corrosion resistance, and ceramic fibers (e.g., SiC) are typical examples of CVD applications [11], [24]-[27]. Due to their unique electrical, mechanical, chemical, and optical properties, 2D semiconductor materials, such as transition metal dichalcogenides (TMDs), have attracted significant attention as core materials for next-generation semiconductor device reality due to the advancement of CVD technologies [28].

CVD variations such as pulsed injection CVD, Electrostatic spray-assisted vapor deposition (ESAVD), and combustion chemical vapor deposition (CCVD) have recently been developed, allowing for the deposition of multi-component films with well-controlled stoichiometry and microstructure.[35] These modifications can potentially broaden the CVD applications for ferroelectrics (e.g., PbTiO₃,

PbZrTiO₃), superconductors (e.g., YaBa₂Cu₃O₇, B doped diamond), and perovskite (e.g., LaSrCoO₃, LaSrMnO3, LaNiO₃), perovskite/graphene materials, and surface treatments [30]-[37].

Apart from that, CVD can be used to synthesize carbon nanostructures such as graphene films and CNTs. Many applications can be found related to CNTs and graphene synthesized under CVD. CVD Synthesis of graphene on metal surfaces can be given as a typical example for such applications. The growth of graphene on the polycrystalline Ni films can be given as an example. Graphene photovoltaic cells, which are transparent, conductive, and highly flexible, considered great candidates, have also been developed using CVD. CVD Synthesis of Graphene on Cu substrates has also been reported. Besides nickel and copper, people tried various transition metal substrates such as Ru, Ir, Pt, Co, Pd, Rh, and Re, showing different carbon solubility, catalytic effects, and high quality. Large-grain graphene and graphene transistors, graphene-based polymeric composites for electronics, electrodes for supercapacitors, sensors, applications of two-dimensional materials and their heterostructures, fabrication, and application of diamond-coated drawing dies, applications of atomic layer deposition for perovskite solar cells, tuning of electronic properties of chemical vapor deposition grown graphene via self-assembled monolayer doping, high selective gas sensors are other remarkable application of CVD [38]-[42].

CVD can also be used to synthesize CNTs, which is known to synthesize high-purity CNTs where industrial applications are possible. Thus, it has been advanced from the laboratory level to the pilot level and the industrial level. Until now, chemical vapor deposition (CVD) has been the most widely used and preferred method for growing carbon nanotubes due to its potential advantages of high purity, high yield, ease of scaling up, and low setup cost [43], [44], [45], [46].

2 MATERIALS AND METHODS

2.1 Materials and components

The standard chemical vapor deposition system is mainly equipped with a feed gas section, a furnace, and vacuum-related accessories that can be categorized under vacuum components for tube furnaces flanges. Stainless Steel vacuum fittings, O-rings, processing tubes and blocks, digital vacuum gauges, pressure controller and flow meters, and vacuum pumps & high vacuum stations are included in the vacuum components section with the central furnace.

2.1.1. Furnace

A furnace (OTF-1200X, MTI-USA) was configured with vacuum components to make a complete CVD unit. The furnace can be fixed with a 60, 70, 80, or 100 mm O.D. quartz tube and a pair of stainless steel vacuum sealing flanges. A high-precision digital controller manages this tube furnace's temperature with 30 programmable segments and a +/- 1 °C accuracy. Without a vacuum, the maximum working temperature is 1200 °C, while with a vacuum, it is 1000 °C. Max positive tube pressure is given as 0.02 Mpa (3 psi).

2.1.2. Vacuum pumps

Three vacuum pumps (RZ-16-Vacuubrand-Germany, VD 201-ULVAC-Japan, Chemstar-1376N-Welch-USA) were used during the research work. R.Z. 16 is an oil-sealed rotary vane pump with a 16.6-19.1 m³/h of pumping speed where the ultimate vacuum (partial) without gas ballast is 2×10^{-4} mbar, ultimate vacuum (total) without gas ballast is 2×10^{-3} mbar, and with gas ballast is 1×10^{-2} mbar. (Fig. 3.a.)



Fig. 3. (a) Z-16-Vacuubrand-Germany, (b) V.D. 201-ULVAC-Japan, (c) Chemstar-1376N-Welch-USA, (d)-(f) vacuum oils, (g) particle filters, (h) oil mist filters, (i) cold traps, and (f) gas leak testing solution.

VD 201 is also an oil-sealed rotary vane pump with 20.2 m³/h of pumping speed (Fig. 3.b). However, the ultimate pressure is given as 6.7×10^{-3} mbar, which is a slightly higher vacuum than R.Z. 16. Chemstar 1376N is a belt-driven rotary vane vacuum pump. This pump is a corrosion-resistant pump that has a 20 m³/h of pumping speed and an ultimate pressure of 1 x 10⁻⁴ mbar without the gas ballast (Fig. 3.c). All pumps are air-cooled. Different types of vacuum oils have been used in different vacuum pumps (Fig. 3.d-f). For example, non-flammable, chemically inert, and thermally stable, Fomblin® PFPE vacuum pump fluids are used in corrosion-resistant vacuum pumps. Fomblin PFPE vacuum pump oils have a very long service life when used with suitable pump filtration. (Fig. 3.f)

2.1.3. Stainless steel vacuum fittings

Valves (KF25 vacuum right-angle valve, on-off valve with both side 1/4 tube fitting connector, screwed bonnet needle valve, high-pressure valve, needle, and metering valve), fittings (Tee type with two 1/4" tube fittings, 316 ss union 90 elbows with both 1/4" tube fitting, 316 ss 1/4" O.D. tube connector, 316 ss ferrule set 1 front ferrule/1 back ferrule), quick clamps and adaptors (Blank flange of K.F. fittings, KF25 to KF16 reducer union, KF40 to KF25 reducer union, KF50 to KF25 reducer union, KF25/40/50 centering rings with mesh, KF25/40/50 quick clamps, KF25/50 tee and bend flanges, K.F. 50 connector flange set), bellow and tubes (KF25 flexible ss vacuum hose, seamless ss tube: 1/4" O.D., adjustable flange support for KF25), cylinder regulators (Two-stage gas regulators for C₂H₂, Ar, Cl₂, N₂, and NH₃), and mixing chambers can be listed under stainless steel vacuum fittings Apart from that some assemblies that were made in-house have been used in the making of CVD. (Fig. 4.a-aa)



Fig. 4. (a),(b) right-angle valves, (c) screwed bonnete needle valve, (d) on-off valve/ball valve, (e) high pressure valve, (f) metered needle valve, (g) Tee, (h) 90 elbow, (i) connector, (j) front/back ferrule system, (k) end cap, (l) blank flange, (m) reducer, (n) centering ring, (o)-(s) clamps, (t) tee flange, (u) bend flange, (v) connector flange set with silicon o-rings, (w) connector flane with valve, (x) ss flexible hose/bellow, (y) flexible hose/bellow, (z) ss tubes, (aa) flange support.

2.1.4. O-rings

Different types of O-rings have been used while developing the system. Those are general purpose rubber O-ring for 25/50 D, high-temperature silicone rubber O-ring for 50 D, and chemical resistant O-ring for 25 D. Chemical resistant O-rings were mainly used when handling special solvent systems such as acetone. (Fig. 5.a)

2.1.5. Processing tubes and thermal blocks

Different types of processing tubes (Quartz tube: 25 OD x 20.5 ID x 450 L mm; 40 OD x 36 ID x 450 L mm; 50 OD x 44 ID x1200 L mm; 60 OD x 44 ID x1200 L mm, alumina tubes) have been utilized in the system. However, quartz tubes with suitable thermal blocks have been used throughout the experiments. (Fig. 5.b&c)

2.1.6. Digital vacuum gauges, pressure controller and flow meters

Manual flow meter (Compact direct read flow meter, 100-1000 cc/min), MFCs (C_2H_2 0-500 SCCM, C_2H_6O 0-500 SCCM, 10% Cl_2 +90% Ar 0-200 SCCM, NH₃ 0-500 SCCM, C_3H_6O), vacuum and pressure gauges (Capacitance diaphragm gauge, Up to 3.8×10^{-5} torr) and controllers have been used to develop the system. (Fig. 5.d-f)



Fig. 5. (a) O-rings, (b) quartz tubes, (c) thermal blocks, (d) pressure gauges, and (e) Mass flow controllers, and (f) MFCs readout box.

2.1.7. Gas regulators and other accessories

Different gas regulators with stainless steel 1/4" tube fittings have been used while developing the system. Some regulators are corrosion resistant, which can handle gases such as chlorine (Fig. 6.a-d). A separate mixing chamber (Fig. 6.e), heating coils (Fig. 6.f), and solvent containers (Fig. 6.g) are other accessories that have been used. Diamond cutters, tube benders, wrenches, and other general accessories have been utilized to set up the CVD system. (Fig. 6.h-j)



Fig. 6. (a)-(d) different types of gas regulators, (e) gas pre-mixing chamber, (f) external gas heating coil, (g) solvent container, (h) diamond cutter, (i) ss tube bender, and (j) a closer look of the tube bender.

2.2. ASSEMBLE A CONVENTIONAL CHEMICAL VAPOR DEPOSITION SYSTEM

A chemical vapor deposition (CVD) system was set up by modifying a conventional design. A tube furnace (single quartz tube) was connected with five types of gas inlets (C_2H_2 , Ar, Cl_2 , N_2 , NH_3) using gas regulators (R1-R5) through different mass flow controllers (MFCs from H1-H5). In order to secure the line isolations, each line was equipped with ball valves (V1-V2). This line is then connected to the quartz tube in the tube furnace (F1) via a mixing chamber (M1). Finally, the system was connected to a vacuum pump VP1 to obtain the high vacuum conditions inside the reaction chamber. A particle filter (F1) and a cold trap (CT1) are equipped before connecting to the vacuum pump. The vacuum is controlled via a right-angle vacuum regulator valve (VR1) and monitored via a pressure gauge (G1) (Fig. 7.) Components of the system were connected using leak-tight stainless tubing, which sustains at a vacuum below 1×10^{-3} torr where front and back ferrules are introduced to the system (Fig. 7.a). All the stainless steel tubings with the required length were cut using a diamond cutter and bent using a tube bender. A digital image of the completed CVD system is given in Fig. 7.b.



Fig. 7. (a) S.S. tubings with front and back ferrule system, (b) digital image of the CVD setup which is developed at Sri Lanka Institute of Nanotechnology, (c) external gas heating coil and solvent system, (d) MFC for solvent introduction, (e) introduction of the inner quartz tube, and (f) introduction of multiple quartz tubes.

This initial design was modified according to the requirements related to carbon nanotube growth, and the modifications are discussed in detail in the coming sections.

2.3. MODIFICATIONS TO CONVENTIONAL CVD DESIGN

2.3.1. Introduction of solvent introduction unit

Synthesis of carbon structures may require different solvent compositions. For example, the synthesis of vertically aligned CNTs required different gas compositions other than the feedstock (C_2H_2) and combined gases (Ar, Cl_2 , NH_3) mainly due to the catalytic property enhancement and to dilute or as a carrier gas during the CNT growth process. However, the supply of solvents to the system other than the pre-connected gas inputs is a challenge. Here, we have introduced separate solvent introduction units (Fig. 8.a. (C2)) to the system where solvent compositions can be easily changed (Fig. 7.a). C_2H_5OH , CH_3OH , $(CH_3)_2CO$, and $CHCl_3$ have been introduced using the developed system. Solution flow was controlled using a manual flow controller (Fig. 8.a (MH1)), where the unit was separated using a ball valve (Fig. 8.a. (V13)). Later, a separate solvent introduction unit. (Fig. 8.a. (C1)) was connected to the system with solvent-resistant MFC (Fig. 8.a (H6) with isolation ball valves. (Fig. 8.a (V11&V12))

2.3.2. Introduction of an external gas heating coil

The heating of feedstock may provide additional support for the CNT growth. Therefore, an external gas heating coil was used to heat the gas flow before the reaction tube (Fig. 8.a. (HC1)). S.S. tubing has been converted to a spiral assembly where the external gas heating coil can provide the heat at its maximum efficiency. (Fig. 7.c)

2.3.3. Introduction of chemical resistant vacuum pump

Due to the emission of acidic fumes, the system was equipped with a chemical-resistant vacuum pump. (Fig. 7.e)

2.3.4. Introduction of the inner quartz tube and quick release NW50 vacuum flange to the system

As explained before, a conventional CVD design consists of a single quartz tube, usually a 50/60 mm O.D tube as the reaction vessel. However, this preliminary design can cause difficulties when cleaning the reaction chamber where several components (flanges, pressure gauges, valves) are engaged in this setup. In order to eliminate the reaction vessel contamination, we have used an inner quartz tube, a 40 mm O.D tube, when performing vapor deposition experiments which is depicted in Fig. 7.f.

Quick-release NW 50 vacuum flange is also a unique feature of the system. This design helps fast assembly and disassembly of the system where inner quartz tubes are inserted and removed from the reaction vessel.

2.3.5. Introduction of parallel multiple inner quartz tubes to the system

Instead of a single quartz tube, multiple parallel quartz tubes were introduced to the system. This modification was implemented to investigate the impact on CNT yield. (Fig. 7.g)

2.4. SCHEMATIC DIAGRAM OF THE CVD DESIGN

Since the developed system was made of a number of piping and components, a schematic diagram is depicted (Fig.8.a) to enhance the content's readability. Further to that, a component notation diagram is also included in Fig. 8.b.





Fig. 8. (a) Associated schematic diagram of the setup displays the piping, instrumentation, other modifications, and (b) the component notations.

3. RESULTS AND DISCUSSION

The developed system successfully synthesized carbon nanostructures (Fig. 9.a-d). Importantly, using the inner tube while synthesizing carbon structures provided easy handling and less contamination. The main or outer quartz tube was not affected during the growth process as the growth is a catalyst-based reaction (Fig. 9.b). However, the secondary or inner tube was contaminated with catalyst and carbon depositions throughout the tube. Here, unreacted catalyst depositions were observed at the edges of the tube, where carbon depositions can be seen in the middle (Fig. 9.c). After taking the reacting tube (secondary tube) out, the outer tube is not contaminated, as shown in Fig. 9.a. The contaminated inner tubes can be cleaned using an acid followed by a thermal treatment. Since the main tube is not affected, a continuous growth can be achieved. In the synthesis of spinnable CNTs, the insertion of parallel tubes has increased the yield of CNTs. This arrangement increased the CNT yield from ~300 to ~850 mg. Using a quick-release flange has also been considered an added advantage in combining the effect of the inner quartz tube.



Fig. 9. (a) Digital image of CNT arrays on quartz, (b) SEM image of CNT arrays on quartz, (C) carbon coatings on ceramic, (d) spaghetti type carbon structures, use of the inner tube for a facile operation which (e) clean main/outer quartz tube with no carbon/catalysts depositions, (f) contaminated secondary/inner quartz tube inside the main/outer quartz tube, and (g) contaminated secondary/inner tube with catalyst depositions at the edges and the carbon depositions in the middle.

4. CONCLUSION

A multi-functional chemical vapor deposition system was developed by modifying conventional CVD laboratory design with some vacuum parts made in Sri Lanka. This system was successfully used to synthesize CNTs, spinnable CNTs, carbon coatings, and other carbon nanostructures hence providing the maximum use of the chemical vapor deposition concept. Minor design changes such as the introduction of inner quartz tubes have provided scalability options for the existing CVD design. This exercise also provided scientific knowledge on the ability to construct advanced CVD systems locally. Hence, an improved design with a larger reaction vessel equipped with a large number of inner parallel quartz tubes with quick-release flange unit can improve the yield as well as the batch efficiency of the current process.

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