Chapter 2

High Temperature Aerosol Sampling Facility: Scheme and Instrumentation

This chapter deals with the design and fabrication of the experimental set-up that was employed to burn the graphite samples and characterize the aerosol produced. The graphite samples were burnt in a clean environment at elevated temperatures and various air flow rates. In the present study we have tried to analyze the particle generation behavior of graphite in an air ingress scenario. Hence we have not used dry zero air as that would not correspond to a realistic situation. The aerosol produced were collected through an isokinetic probe for accurate sampling. The high temperature and concentration of the aerosol laden exhaust gas necessitated the use of a cooling and dilution system to lower the temperature and concentration of the aerosol to a level which could be measured by the aerosol spectrometer. A special cooling arrangement was employed to maintain the mechanical integrity of the set-up against high temperature failure.

In the set-up (Figure 2.1), a recrystallized alumina tube of inner diameter of 70 mm was placed in the center of a split box-type furnace. The graphite sample to be burnt was placed in the center of this tube. Recrystallized alumina was chosen for this purpose because of its low thermal expansion, high thermal-shock resistance, high-compressive strength, excellent wear and abrasion resistance and outstanding chemical corrosion resistance under high temperature. The left end of this tube was connected through a water-cooled flange to the inlet air supply. The air flow rate was maintained by an air compressor (Model No. N035AVE 1920, Make-KNF, Germany) and monitored using a rotameter. Before entering the alumina tube, the supplied air was passed through a HEPA

filter (Grade 1602051, Make-TSI, USA). The clean air acts as the oxidant as well as the carrier gas and transports the aerosol coming out of the burning graphite.

The aerosol was sampled from a tube made of borosilicate glass, which was connected to the alumina tube by another water-cooled flange. Borosilicate glass has a very low thermal expansion coefficient, making it more resistance to thermal shock as compared to ordinary glass. It also has good optical clarity which provided easy access for arrangement of isokinetic probe. The probe, facing the air flow was placed in the center of the borosilicate tube. Since the temperature of the gas at the inlet of the isokinetic probe was measured to be in the range 120 - 150 °C, a counter-flow heat exchanger was connected downstream of the isokinetic probe to cool the aerosol to the maximum working temperature limit of the aerosol spectrometer (40 °C). After passing through the heat exchanger, the aerosol was led to a diluter to reduce the particle concentration within the measurable range of the spectrometer as well as to further cool the aerosol by dilution. After the diluter, the flow passes through a flow splitter (Model No. 3708, Make-TSI) which split the flow and provided two flow streams of flow rates 1.0 $Lmin^{-1}$ and 0.8 $Lmin^{-1}$. These streams were then led to the two spectrometers (OPS 3330 & Nanoscan 3910, Make-TSI, USA) which measured the aerosol size and concentration. The aerosol was led to a gas analyser by inserting a probe in borosilicate tube, close to the isokinetic probe, to measure the CO and CO_2 content. The remaining unutilized portion of the exhaust was passed through a filter paper held by a filter holder before being finally emitted into the atmosphere. Details of the various components of the setup are presented below in Figure 2.1 and Figure 2.2.

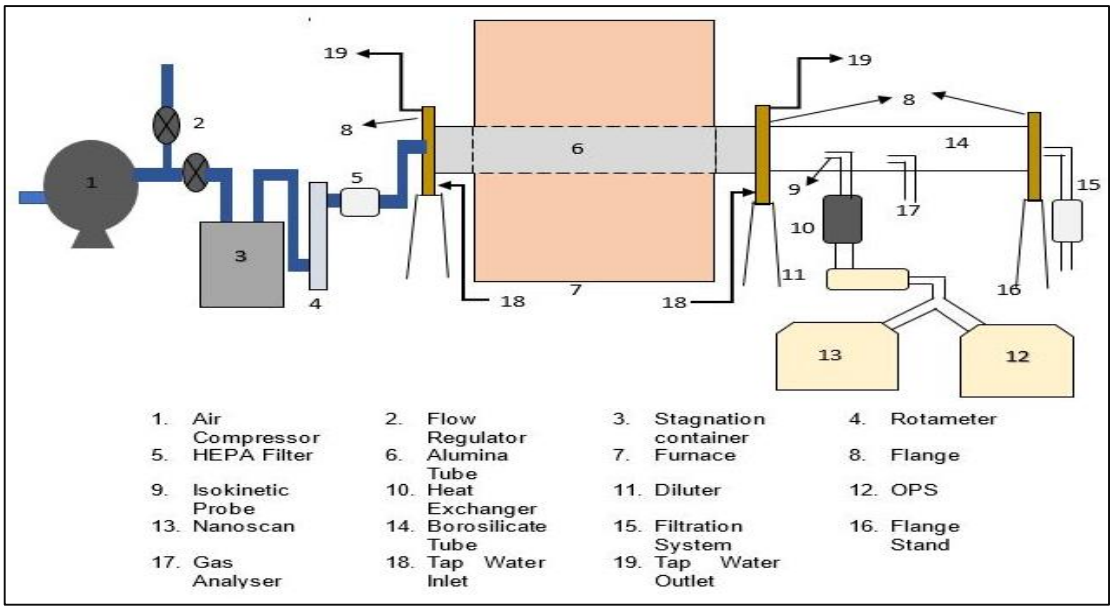

Figure 2.1: Schematic diagram of experimental set-up

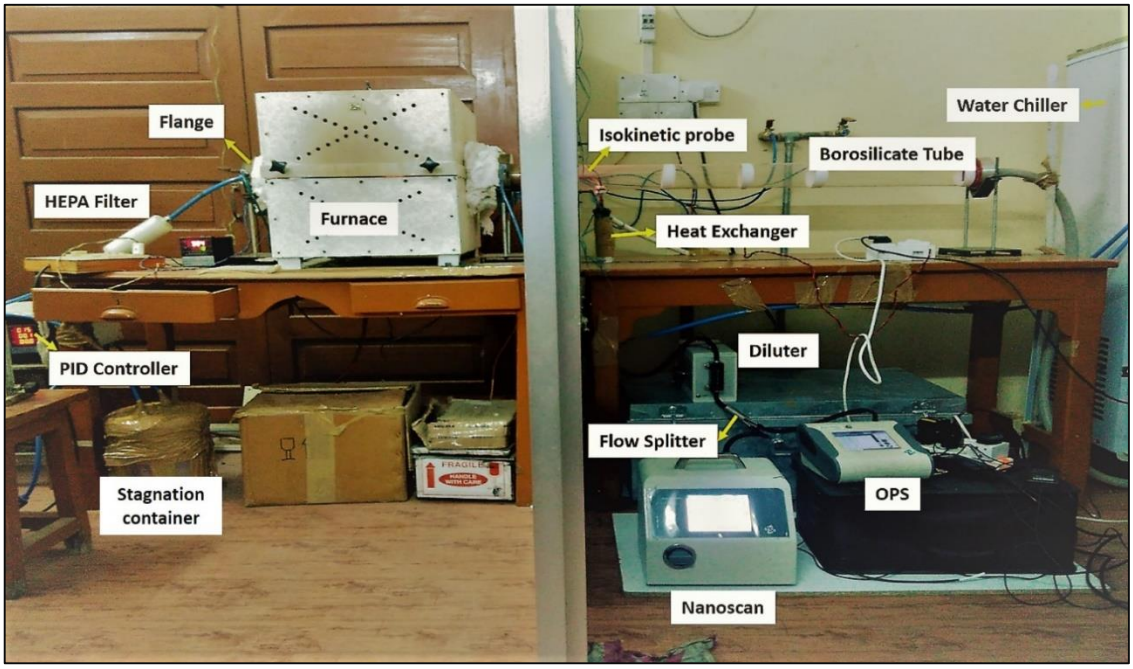

Figure 2.2: Picture of the experimental set-up

The experimental set-up can be broadly classified into the following units:

- **Heating unit**- Furnace, PID controller, Thermocouple, Flange, Flow homogenizer, Alumina tube, Quartz tube

- **Clean air supply unit**- Air compressor, Y-Joint, Flow regulator, Stagnation container, Rotameter, HEPA filter, PVC tubing
- **Aerosol sampling and gas measurement unit**- Isokinetic probe, Heat exchanger, Flow splitter, Diluter, OPS, Nanoscan, Gas analyser

Details of the various components of the experimental set-up are discussed below:

2.1 Heating unit

The heating unit consists of an electric resistance furnace in which a recrystallized alumina tube is placed and secured at both ends by water-cooled flanges resting on supports. Thermocouples and controller are used for temperature measurement and control.

2.1.1 Furnace, flow homogenizer, PID controller and thermocouples

For burning graphite, a box type split electric furnace (Make-Anil Scientico, India) (heating zone dimension: 205 × 130 × 220 mm^3) having 8 silicon carbide rod heating elements was used. The arrangement of the heating rods inside the furnace is shown in Figure 2.3. The reason for selecting a split furnace was due to its convenience in operating and cleaning the recrystallized alumina tube. A recrystallized alumina tube (I. D. = 70 mm, O.D. = 80 mm, L = 650 mm) was placed in the center of the furnace. A photograph and a schematic sketch of the furnace is given in Figure 2.3. A platinum-rhodium (R-type) thermocouple (measuring range: 0 - 1700 °C), was used to measure the temperature inside the furnace. The thermocouple tip touched the exterior surface of the alumina tube. The temperature measured by this thermocouple is called the ‘furnace temperature’ or ‘outside tube temperature’ throughout this work. To measure the temperature at the location where the graphite is placed for burning, another R-type thermocouple was inserted axially inside the alumina tube. The temperature measured by this thermocouple

is the ‘inside tube temperature’. Four J-type thermocouples (measuring range: 0 - 600 °C) were used for measuring the temperature of air inside the borosilicate tube at four different locations. A data acquisition system (Model No. DAQ-9178, Make-NI, USA) was used for acquiring the temperature at various locations in the set-up. The temperature and heating rate of the furnace was maintained and controlled using a proportional-integral-derivative (PID) controller which minimized the offset and oscillation in the corrective response (Figure 2.4).

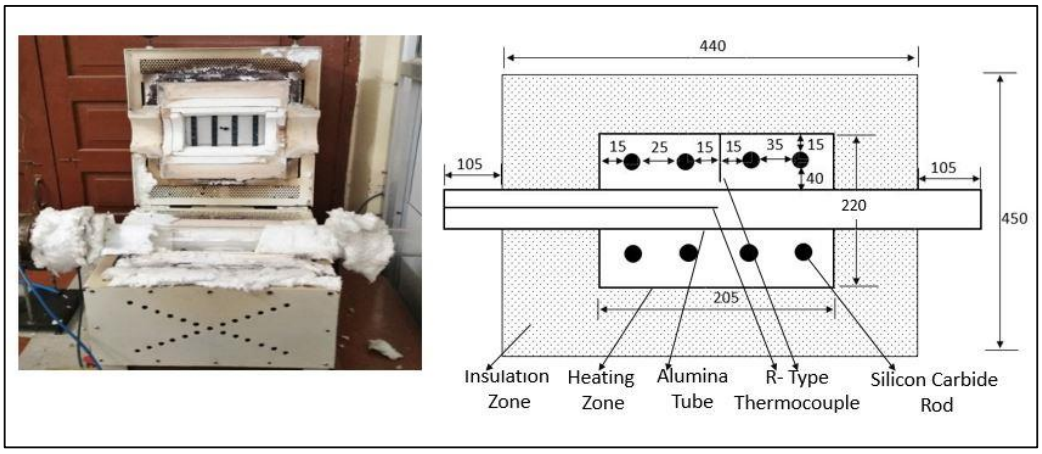

Figure 2.3: Furnace picture and schematic diagram (front view) (All dimensions are in mm)

Figure 2.4: PID controller

2.1.2 Flange and flow homogenizer

Two flanges made of gun metal (Copper-Tin-Zinc alloy) were fabricated because of its excellent wear resistance and sliding properties. Tap water was used for cooling both flanges through a 4 mm internal diameter pipe. The purpose of the water-cooled flanges was two-fold. The flange at the inlet of the alumina tube was used to prevent damage to the PVC tubing used for air supply. It was found that in the absence of this water-cooled flange, the temperature at the air inlet to the alumina tube during experiment shot up to a high value, sufficient to melt the PVC tubing. The other flange, located at the outlet of the alumina tube, provided a seamless and air-tight fastening of the alumina and borosilicate tubes.

Air supplied by the compressor, after passing through the HEPA filter was fed to the alumina tube by an 8 mm internal diameter PVC tubing. High air velocity inside the PVC tubing produced a jet of air into the alumina tube which resulted in a non-uniform burning of sample. To overcome this problem, a flow homogenizer device shown in Figure 2.5 was attached to the left flange to reduce the velocity of the incoming air. The incoming air was led to a small chamber in the device which had three holes at its periphery, spaced 120 degrees apart. The air escaped into the alumina tube through these three holes. This device resulted in a uniform air supply and consequently uniform burning. The specifications of the flange are shown in the Figure 2.6. A silicon sealant was used to ensure leak-proof air supply in the system.

Figure 2.5: Flow homogenizer device

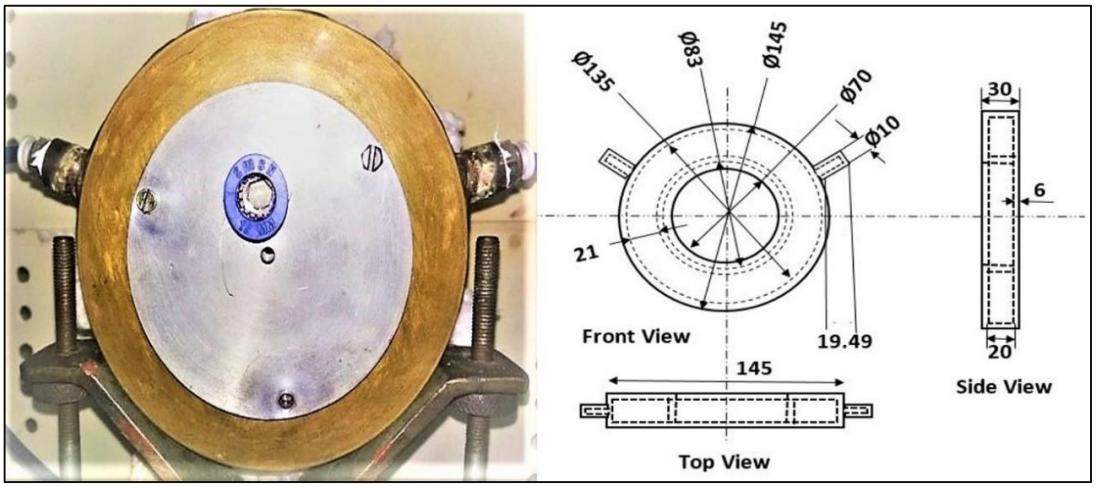

Figure 2.6: Picture of flange and its drawing (All dimension are in mm)

2.1.3 Recrystallized alumina tube and boat properties

In the set-up, an alumina tube was used for heating graphite samples. Alumina can occur in various crystalline phases, in which hexagonal alpha phase is the most stable at elevated temperatures. Compared to all ceramic oxides, alpha phase alumina is the stiffest and strongest. Its high hardness (23000 MPa), good thermal properties, refractoriness and elevated dielectric properties make it the appropriate material for a wide range of applications. It provides the combination of low thermal expansion coefficient ($\sim 8.5 \times 10^{-6}$ /K), superior thermal conductivity and high compressive strength (3500 MPa),

making it thermal shock resistant. High purity alumina is used in the both reducing and oxidizing atmosphere up to 1925 °C temperature. It is also resistant to attack by all gases except fluorine and all common reagents except phosphoric acid and hydrofluoric acid. Low purity alumina is attacked by only alkali metal vapors at high temperature. In our experimental set-up, the maximum furnace temperature can reach a value of 1600 °C. All this survey presented it as the ideal material for this application. The tube is carefully needed to be balanced horizontally inside the split furnace using the leveler to prevent unnecessary stress, whenever reassembled after cleaning. The heat was supplied to the alumina tube by a PID controlled furnace. To prevent breakage of the alumina tube which has poor thermal conductivity, a suitable heating rate was chosen. The temperature of the furnace was raised up to 600 °C at a heating rate of 4 °C/min followed by a constant temperature of 600 °C for 30 minutes to thermally stabilize the system followed by a lower heating rate of 2 °C/min beyond 600 °C. The heating rate was lowered because it has been reported that the effect of thermal shock becomes more severe at higher temperatures (Aksel, 2003). Graphite sample was kept in an alumina boat. The properties of alumina tube are shown in Table 2.1.

Table 2.1: Properties of alumina

Properties	**Value**
Principal constituent Al_2O_3 purity	> 99.9% (Al_2O_3)
Sintering additives	Nil
Appearance	Pearl White
Apparent density	> 3.89 g/cc
Grain size	~10 μm
Open porosity	Nil
Hardness	23000 MPa
Compressive strength	3500 MPa
Melting point	2030 °C
Maxima working temperature	1800 °C
Thermal conductivity	5-30 W/mK
Coefficient of linear expansion 0 - 1000 °C	~ 8.5×10^{-6} /K
Emissivity at 1000 °C	~ 21%

2.1.4 Quartz tube

In the experimental set-up, alumina and borosilicate tubes were replaced by a single quartz tube when graphite was heated at heating rates above 4 °C/min. This change was necessitated by the fact that an alumina tube can safely withstand a maximum heating rate of 5 °C/min. The most significant properties of quartz are its low expansion coefficient and excellent thermal shock resistance. For example, when thin sections of a quartz tube are quickly heated to 1100 °C and then suddenly cool down to 20 °C, the quartz tube does not break. Quartz is inert to most of the chemical reagent. The only compound that can etch the quartz tube even at low temperatures is hydrofluoric and phosphoric acid. A few significant properties of quartz are shown in Table 2.2.

Table 2.2: Properties of quartz

Properties	**Value**
Colour	Transparent
Density	2.2 g/cm^3
Silica content	99.9%
Coefficient of Expansion	5.5 x 10^{-7} /°C
Refractive index	1.45
Deformation	1120 °C
Melting point	1730

2.2 Clean air supply unit

Air coming out from the air compressor is led to a Y joint by a PVC tube of 8 mm internal diameter which divides the flow into two parts. One part goes to the atmosphere after passing through a flow regulator to reduce the load on the compressor. The other part passes through the main line via another flow regulator to a stagnation container. The air compressor used to supply air is of a reciprocating piston type. Air coming out from the compressor outlet has a pulsatile flow. To dampen the pulsation in the flow, the air is supplied to the stagnation container which is a large air reservoir. Stabilized air coming

out from the container is passed through a rotameter (Model No. CVG-LFR, Make-CVG Technocrats, India) and then through a HEPA filter before entering into the recrystallized alumina tube. The schematic diagram of clean air supply unit shown in Figure 2.7 and picture of components used in supply unit are shown in Figure 2.8.

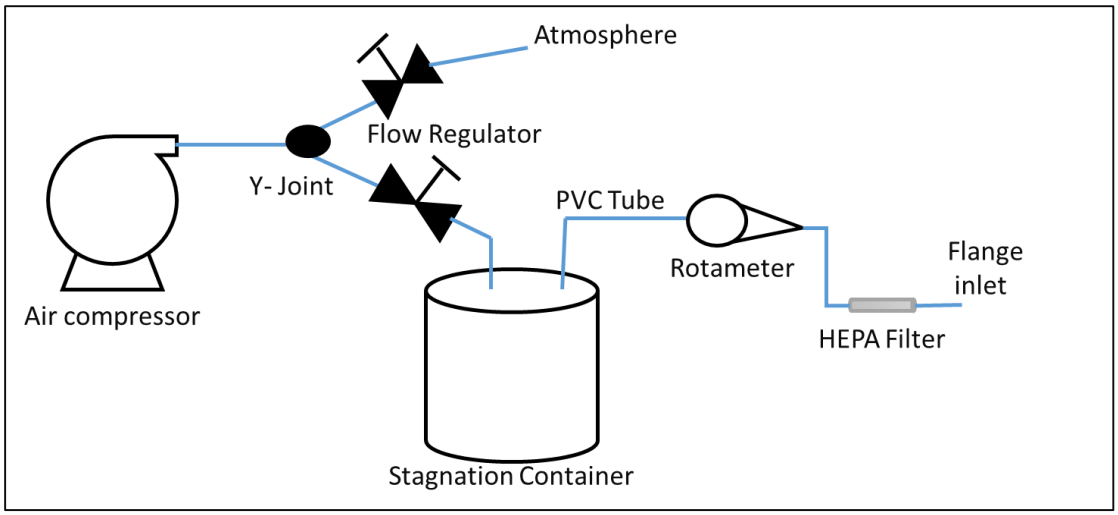

Figure 2.7: Systematic diagram of clean air supply unit

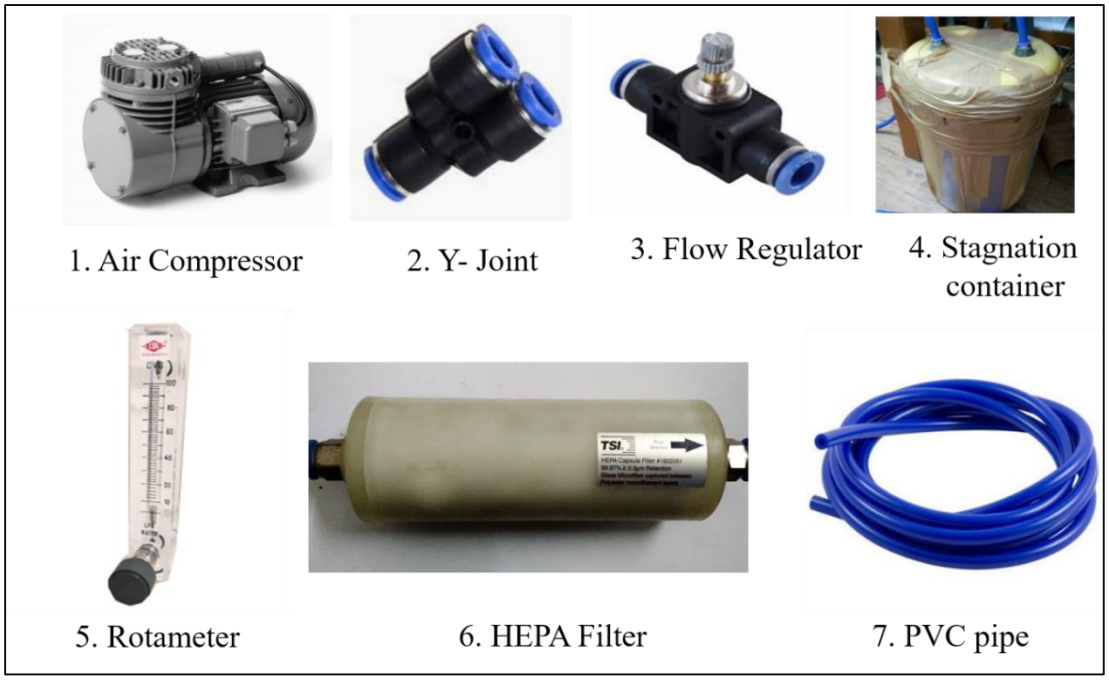

Figure 2.8: Component of clean air supply unit

2.3 Sampling-aerosol measurement system

This unit consists of a borosilicate tube that receives the hot exhaust from the exit of alumina tube and transports it to the isokinetic probe. The probe collects the aerosol samples and directs them through a counter-flow heat exchanger which cools the hot sample and after passing through diluter, the flow of aerosol is divided into two streams using the flow splitter. One stream is directed to the Nanoscan and other is sent to the OPS.

2.3.1 Borosilicate glass tube

Borosilicate glass is a highly chemical resistant, mechanically and thermally stable material that can withstand temperature up to 600 °C with a very low coefficient of thermal expansion (3.3×10^{-6} /K). It is optically transparent and is an ideal material for a test duct. Its smooth surface allows easy cleaning and maintenance with no interference with the particle flow. For measuring aerosol concentration, four sampling ports are fabricated in the borosilicate tube as shown in Figure 2.9. The properties of borosilicate tube are shown in Table 2.3.

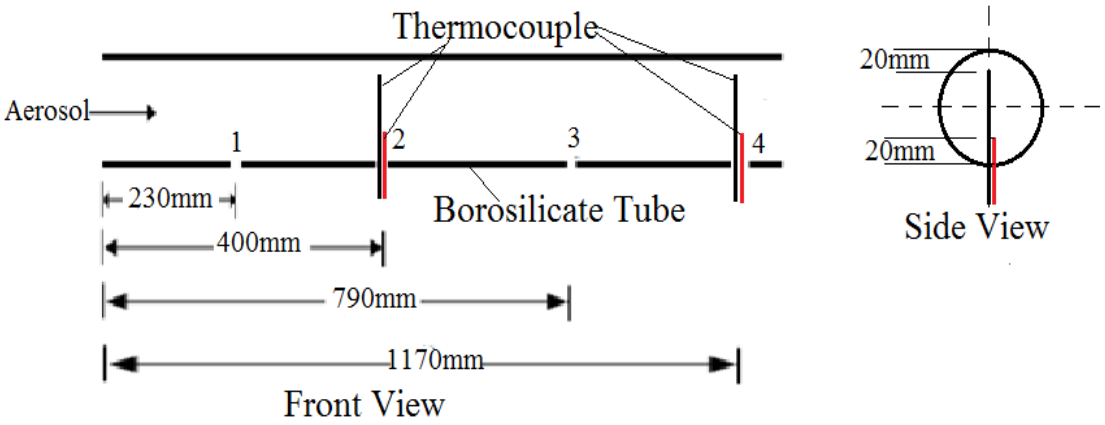

Figure 2.9: Sampling ports in borosilicate tube

Table 2.3: Properties of borosilicate

Properties	Value
Coefficient of thermal expansion	3.3×10^{-6} /K
Transformation temperature	600 °C
Density	2.23 g/cm^3
Poisson's ratio	0.20
Thermal conductivity	1.2 W/mK
Refractive index	1.473

2.3.2 Isokinetic probe

For aerosol measurement, especially those concerned with aerosol particle size distribution, it is essential to sample under isokinetic conditions (Arouca et al., 2010). The isokinetic sampling procedure has been espoused to avoid error in critical parameters (particle concentration, collection efficiency, etc.) (Ichitsubo and Otani, 2012; Krämer and Afchine, 2004; Tsuji et al., 2008). An isokinetic sampling requires that the gas velocity in a probe equals the velocity outside the probe, else its sizing distortion takes place. The distortion favors large particles if the velocity of flow inside the probe is less than the velocity in tube and favors small particles, otherwise. These methodology standards are determined by the US Environmental Protection Agency (Xie et al., 2018). Equal velocities can be achieved by equalizing the ratios of the flow rates to the cross-sectional areas of the probe and the tube (Ichitsubo and Otani, 2012). In the present work, the diameter of the isokinetic sampling probe was calculated to be 29.7 mm. Detail of the isokinetic probe calculation was given in next chapter. The fabricated probe and its design are depicted in Figure 2.10.

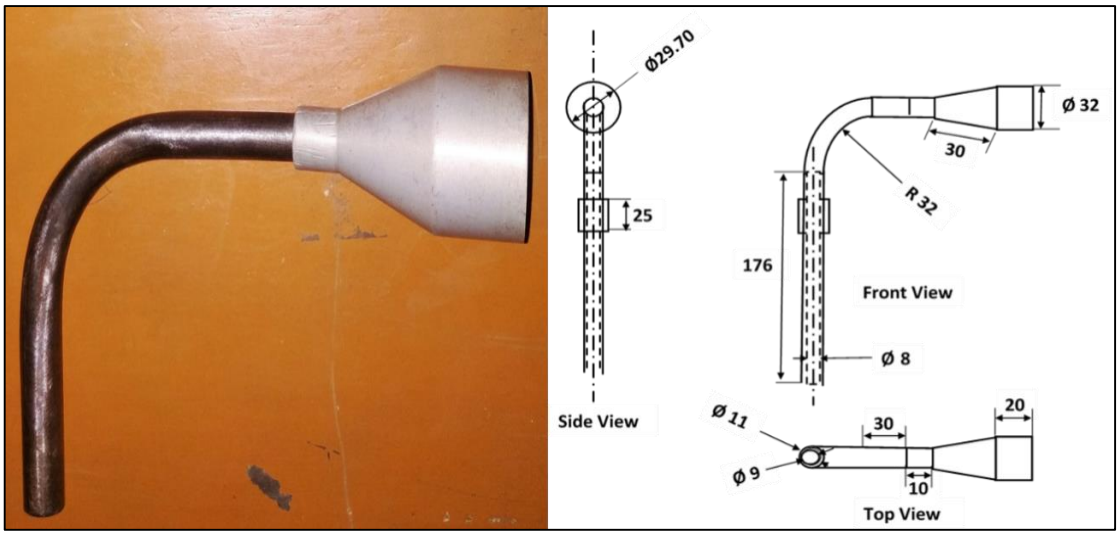

Figure 2.10: Isokinetic probe (All dimensions are in mm)

The probe collects the aerosol samples which were then passed through a heat exchanger to cool them. The cooled aerosol was then passed through a diluter followed by a flow splitter. The flow splitter divides the flow into two streams. One stream is directed to Nanoscan and other is sent to OPS.

2.3.3 Heat exchanger

The Nanoscan and Optical Particle Sizer have a maximum operating temperature of 40 °C. The excess heat from the aerosol laden hot air has to be removed before it can be supplied to these instruments. The device used for efficiently transferring heat between two fluids is called a heat exchanger. In the set-up, a counter-flow heat exchanger is used for cooling the air. The heat exchanger contains two concentric tubes in which the two fluids (cold water and hot aerosol) flow in opposite direction. The outer tube is made of PVC for insulation while the inner one is made of copper for enhanced hear transfer. Hot aerosol flows in the inner copper tube while the cold water flows in the annulus. The cold water is supplied by a chiller in the range of 1 - 4 °C. The design and specifications of the heat exchanger are shown in Figure 2.11.

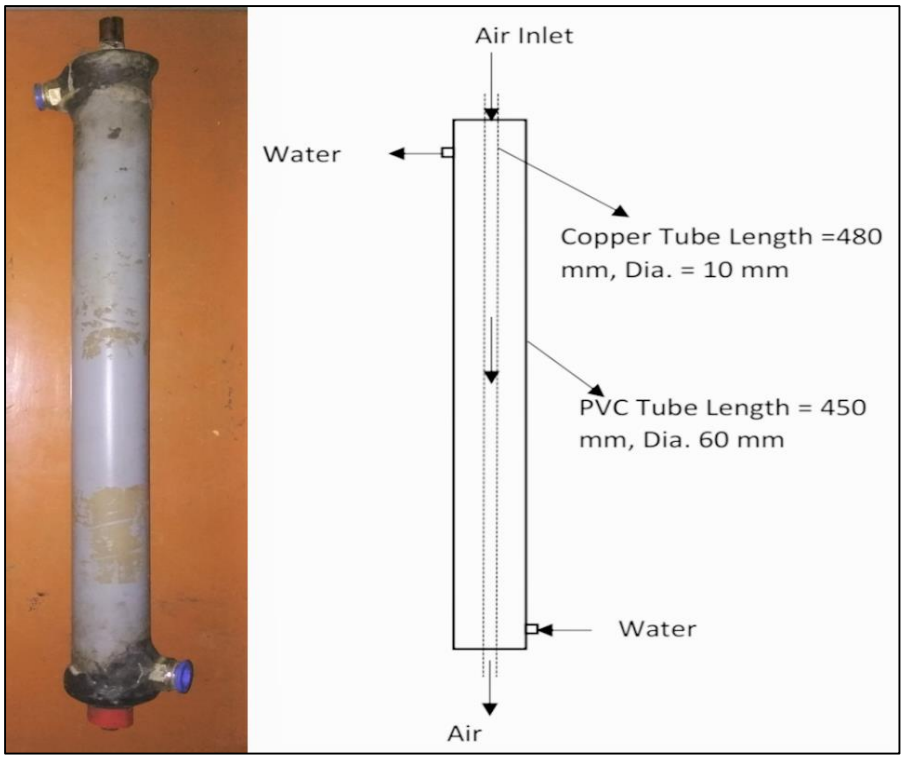

Figure 2.11: Heat exchanger

2.3.4 Aerosol diluter

The maximum aerosol number concentration that can be measured using the Nanoscan and OPS is 10^6 #/cm^3 and 3×10^3 #/cm^3 respectively. During the burning of graphite, it was found that the concentration of aerosol produced exceeded the maximum limit of the spectrometers. Hence a diluter was used to dilute the aerosol sample so that the spectrometers could measure it. The diluter used in the set-up is of Model No. 3332 of TSI having a dilution ratio of 1:100 (Figure 2.12). The main stream of aerosol enters the inlet of the dilution system where it splits into two: one goes to a capillary tube (i.e. capillary volume flow) and the other passes through the HEPA filter (i.e. by-pass volume flow). The concentration of particles is constant in the capillary volume flow, while in the by-pass flow, the particles are removed by a HEPA filter. Both the sub-streams combine together at the outlet of the dilution system. The dilution factor is the ratio of by-pass flow to the capillary flow.

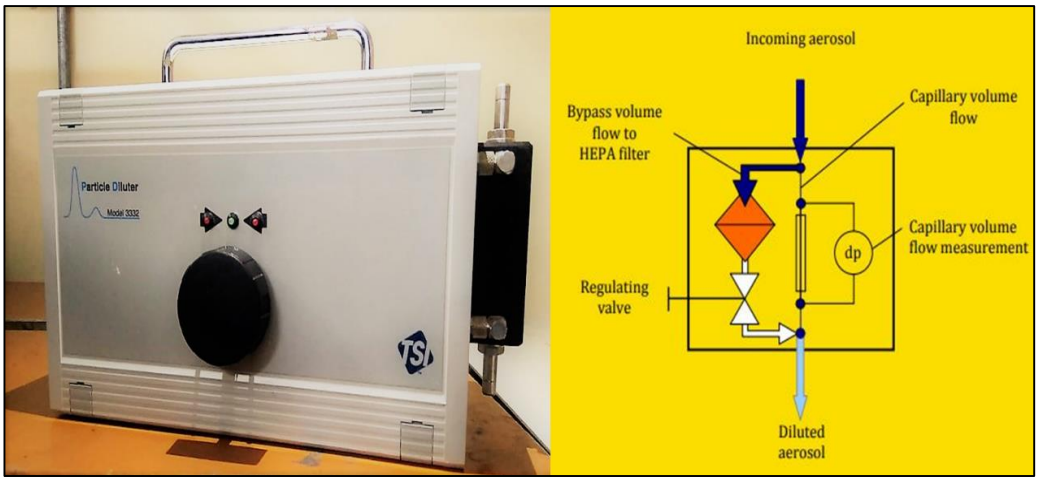

Figure 2.12: Picture of diluter and schematic of its internal circuit

2.3.5 Flow splitter

The flow splitter (Figure 2.13) was used to split the flow into the Nanoscan and the OPS. The flow splitter can direct the aerosol sample to multiple destinations. This accessory is useful when comparing or calibrating instruments and when connecting two or more instruments to a single sample port. Its smooth flow transitions offer a distribution of equal flow. Its electro-polished stainless-steel structure prevents contamination of the aerosol. The flow splitter used has a straight tube inlet of diameter 3/8 inch and four 1/4 inch diameter outlets. Conducting tubes join the inlet of Nanoscan and OPS with the two outlets of the flow splitter and the remaining two stay closed.

Figure 2.13: Flow splitter

2.3.6 Aerosol measurement system

Various real-time aerosol sizers are available commercially to measure aerosol number concentration and size distribution. In the present thesis, two instruments, i.e. portable scanning mobility particle sizer (Nanoscan 3910) and optical particle sizer (OPS 3330) were used (Figure 2.14). The Nanoscan works on the principle of particle mobility and OPS works on the principle of light scattering (Tritscher et al., 2013). The Nanoscan was used to measure the particle size distribution between the size range of 10 - 420 nm, while OPS measured particle size distribution between 300 - 10,000 nm. In combination, these instruments cover the full range from 10 - 10,000 nm. The specifications of OPS and Nanoscan are shown in Table 2.4.

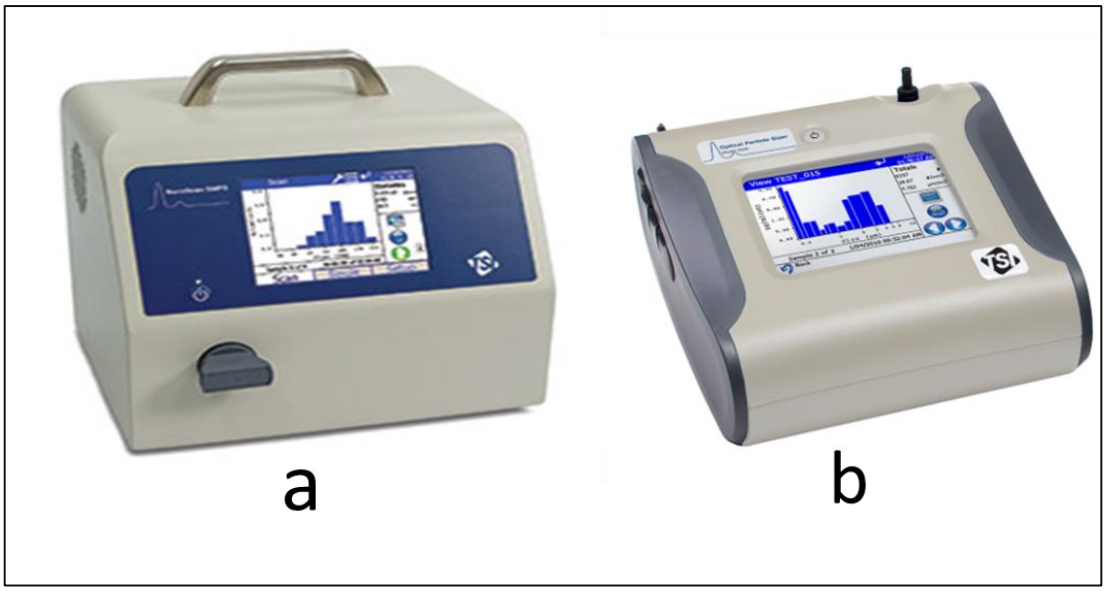

Figure 2.14: Picture of (a) Nanoscan (b) Optical Particle Sizer

Table 2.4: Specifications of the Nanoscan and OPS

Instrument	**Nanoscan**	**OPS**
Model	3910, TSI Inc.	3330, TSI Inc.
Measuring Size Range (nm)	10 - 420	300 - 10,000
Sample Flow Rate ($Lmin^{-1}$)	0.8	1.0
Number Concentration (cm^{-3})	10^2 - 10^6	< 3000
Size Channels	13	16

2.3.6.1 Nanoscan working principle

Schematic diagram of the internal parts of Nanoscan is shown in Figure 2.15. An inlet cyclone, unipolar diffuser charger, radial differential mobility analyser (rDMA) and Condensation Particle Counter (CPC) are the main components of the instrument. The inlet cyclone having a cut off diameter of 0.5 μm, is used for removing coarse particles (particle above the Nanoscan measuring range).

Poly–dispersed aerosol pass through the orifice into a corona jet charger. In the corona jet charger, a charge is provided on the aerosol on the basis of the particle size

with more charge provided to the larger size particles and less to the smaller size particles. In the corona jet charger, a reverse flow is provided to ensure proper mixing and charge repeatability. After passing the corona jet, the flow of aerosol is split into two streams: one stream passes through a HEPA filter and is used as a sheath flow in the instrument and the other stream enters tangentially in radial DMA. The radial DMA comprises of two plates in which one is grounded while the other possesses a high negative voltage. A large electric field is generated between plates due to the potential difference. The charged particles separate in the DMA according to their electrical mobility in the electric field. Larger particles with high electrical mobility travel long distance in the electric field while smaller particle with low electrical mobility travel a small distance (Figure 2.16). After separation, the aerosol finally exists from the bottom center port of the DMA and goes to the CPC. The CPC counts the number of particles and measures their size using a condensation technique.

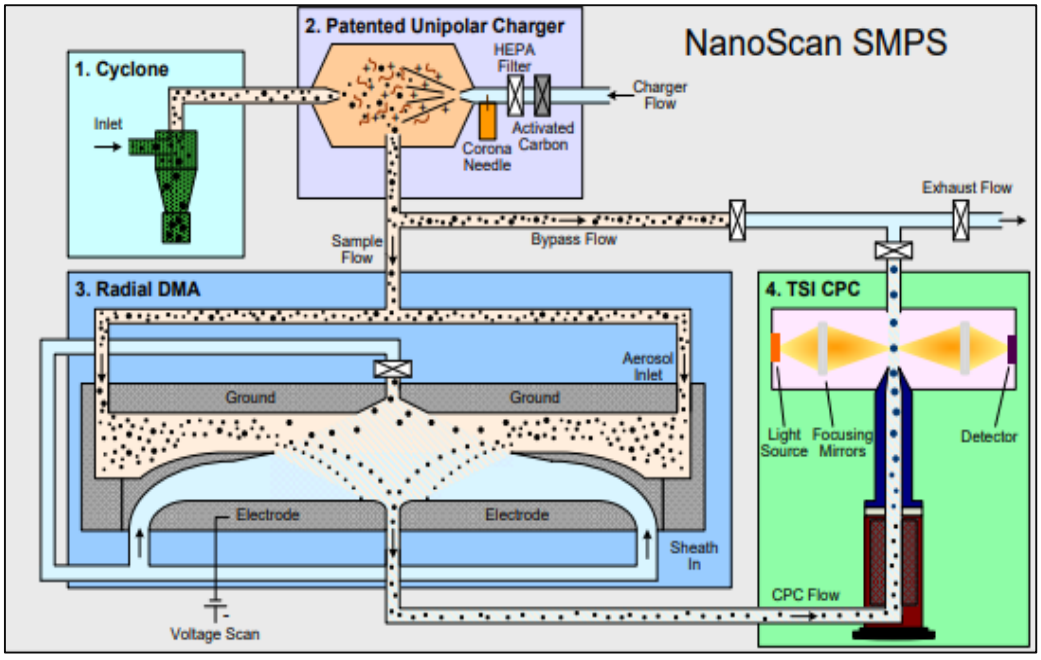

Figure 2.15: Schematic diagram of Nanoscan working principle (TSI, 2013)

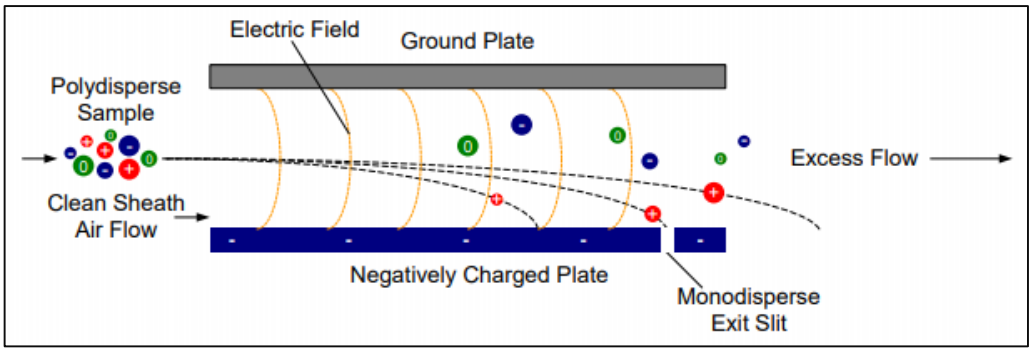

Figure 2.16: Schematic of differential mobility analyzer (TSI, 2013)

2.3.6.2 Working of OPS

The Optical Particle Sizer (OPS) Model 3330 is a light, portable spectrometer that provides fast and accurate measurement of particle concentration and particle size distribution using single particle counting technology. The OPS measures particles from 0.3 to 10 µm in 16 user adjustable size channels.

When aerosol passes through a laser beam/focused light as a thin stream surrounded by sheath air, so that only one particle at a time is illuminated and scatters light to the detector, which is converted to an electrical signal. Electronic pulse height (or area) analysis is used to interpret the pulse and direct a count to the proper size channel, where the total count in each size range is accumulated. The particle size distribution by count is obtained from the accumulated counts in each size channel. This instrument is works on the principle that the scattered light intensity is a monotonic function of a particle size. After the counting and size measurement of particles, the aerosol sample moves from the optic chamber to the filter cartridge where particles are collected on a 37 mm filter for gravimetric, chemical or microscopic analysis.

OPS have proven to be a valuable tool in air pollution studies, aerosol research and clean room monitoring because of their ability to rapidly provide information on aerosol size distributions. Commercial instrument covers the size range of 0.05 - 20 µm, although

0.1 - 5 µm is most common and provide measurement of number concentration in 5 - 16 size channels.

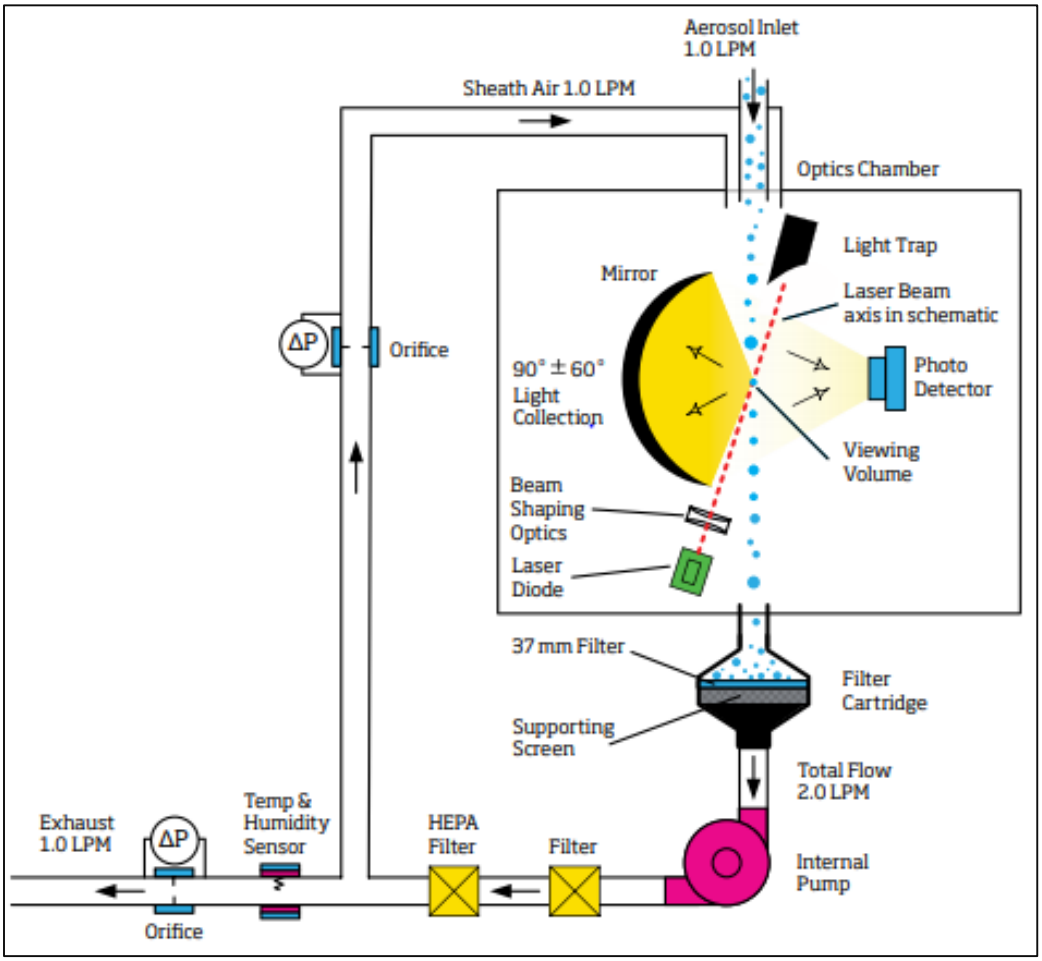

Figure 2.17: Schematic diagram of OPS working principle (TSI, 2015)

Flexible, conductive silicon tube was used to prevent deposition of charge particles during transportation on the sampling line. The total length of tube between the outlet of test tube and the inlet of each instrument (OPS and Nanoscan) were approximately 0.3 m. Both instruments were put in parallel (OPS, Nanoscan). The particle concentration and size distribution were evaluated continuously. After leaving the borosilicate tube through an aluminum flange resting on a support, the exhaust is led by a bent glass tube to the filtration system. It consists of an inverted truncated cone made up of glass that holds a mesh over which lies an appropriate filter paper. As the exhaust passes through the filter

paper, the aerosol particles of the desired size are deposited on the filter paper. The gases after passing through the filter paper are directed out into the atmosphere using a polyvinyl chloride tube.

2.3.7 Gas analyser

Burning of graphite results in generation of CO and CO_2 gases. Measurement of these gases gives vital information about the state of oxidation reaction taking place in the sample. These gases were measured using a gas analyser (Model No. AVL Digas 444, Make-AVL, India) which uses nondestructive sensors to measure the concentration of the gas species in the flow stream. A probe connected to this device was inserted inside the borosilicate tube through a hole drilled in the tube wall. The digital readings on the display panel of the gas analyser were noted as a function of time. The least count of the instrument for the two gases was: CO- (0.01 v/v%) and CO_2 -(0.1 v/v%). The picture of gas analyser was shown in Figure 2.18.

Figure 2.18: Gas analyser

Summary

An experimental set-up was developed to study the particle emission behavior of combustible materials. This facility can attain a maximum furnace temperature of 1350 °C with a maximum air flow rate of 25 $Lmin^{-1}$. Air supply by a compressor was passed through a HEPA filter and measured by a rotameter. Isokinetic sampling of graphite aerosol was achieved using an isokinetic probe designed for this purpose. A cooling system employing a heat exchanger was used to cool the aerosol to the working limit of the spectrometers. A diluter was used to lower the concentration of the aerosol up to the measuring range of the aerosol spectrometers. Special provisions like water-cooling of flanges helped maintain the integrity of the PVC and sealant components. Special care was taken to hermetically seal the set-up against external air ingress which would contaminate the aerosol generated.

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