The present invention concerns methods for supplying gas to a very high sensitivity analyser of residual traces of gas in a pure gas (hereinafter referred to as "traces"), of the type comprising the stages of sequential supply to a section of feed line to the analyser at a given flow rate:

a) of a gas to be analysed, via a first line,

b) of a pure gas produced by purifying the gas to be analysed, via a second line,

c) of at least one calibration gas, diluted by the pure gas, initially containing a higher level of residual gas traces than the gas to be analysed, via a third line,

the downstream parts of the two first lines being connected to the section of feed line, the downstream part of the third line being connected to the section or to the second line.

A method of this type is described in the document EP-A-370.871.

The use of very high sensitivity analysers of traces or impurities, notably of the ultrasensitive hygrometer or Fourier-transform infrared (FTIR) spectroscope type, capable of detecting and analysing traces of gases at levels below parts per billion and used, for example, for gases of very high purity (nitrogen, argon) for the very large-scale integration (VLSI) semiconductor industry, requires the ability to supply sequentially to the analyser, at a given flow rate, the gas to be analysed, a so-called "zero" gas, obtained by purifying the gas to be analysed and defining the zero of the analyser, and a calibration gas, obtained by heavy dilution of a gas which initially contains a given large quantity of traces, so as to calibrate the reading range of the analyser.

The known methods and devices use three lines, each provided with a flow regulator and a stop valve to isolate the lines which are not involved in one of the phases of supply of gas to the analyser. These processes have two major drawbacks, in that the flow regulators are contaminating components liable to add uncontrolled traces or impurities to the different gas lines and in that, in the periods when the lines are isolated, the gas in these lines is liable to be rapidly contaminated by simple adsorption or desorption on the walls of the tubes.

To reduce the above-mentioned drawbacks substantially, there was proposed, in the above-mentioned document, a method and a device, using two main lines, the first supplied with the gas to be analysed, the second with a so-called "zero" gas (defining the zero of the analyser). The third type of line (calibration) is connected, in its downstream part, to the second line, upstream of the point at which this second line is connected to the section feeding the analyser.

Each of these lines is provided with a static pressure-drop device, which is less contaminating than a flow regulator, and, in its downstream part, with an isolation means enabling the line to be isolated, with scavenging of the upstream part of the line.

This approach enables the lengths of isolated, non-scavenged tubes to be reduced but not totally eliminated. Furthermore, the supply pressure in the first and second lines may vary over a wide range, so that the method and device proposed involve risks of considerable drift over time which do not enable the supply of a given flow to the analyser to be guaranteed, and which therefore may impair the accuracy of the measurements.

The object of the present invention is to propose a method for supplying gas to an analyser making it possible to eliminate any contaminating component along the route of the different

gases, to keep all the lines in the system continuously scavenged, to provide regulation of the flow rate which is precise and unlikely to drift over time, and therefore to guarantee a reliable and markedly improved quality of analysis.

To achieve this, according to one characteristic of the invention, the upstream parts of the three lines have continuously passing through them the gas corresponding to a constant flow rate, in that there is provided, in an intermediate part of each line, a discharge branch equipped with a controllable flow regulator, the constant flow rate in the first and second lines being greater than the given flow rate for the analyser, and in that, for each above-mentioned phase a) and b), the flow regulators are controlled so that a first part of the excess flow in the line corresponding to one phase is discharged by the associated discharge branch and so that the second and third parts of the excess flow of the said gas are recirculated in each of the downstream parts of the other two lines, the whole of whose excess flow proper and the second or third part of whose recirculated flow are discharged by the corresponding discharge branch.

According to a more particular characteristic of the invention, the constant flow rate in each line is ensured by a calibrated restriction functioning sonically, the upstream pressure of the calibrated restriction being measured in order to control at least the flow regulator of the associated discharge branch.

Another object of the present invention is to propose a device for supplying gas to an analyser for the implementation of the above method, of the type comprising:

- a source of a gas to be analysed;

- a source of a calibration gas;

- a first line, including a first flow-regulation means, connecting the source of the gas to be analysed directly to a section of feed line of the analyser;

- a second line connected, upstream, to the source of gas to be analysed and including, in series, a gas purifier and a second means for regulating the flow, and, downstream, to the section of feed line to the analyser;

- at least one third line, including a third flow-regulation means, connected to a source of calibration gas initially containing a larger quantity of traces than the gas to be analysed and, downstream, to a downstream part of the second line or to the section of feed line to the analyser;

and characterised in that each line includes, in an upstream part, a calibrated restriction forming a sonic neck and, in an intermediate part, a discharge branch provided with a discharge flow regulator.

According to another characteristic of the invention, each line comprises, immediately upstream of the restriction in this line, a pressure sensor which is typically mounted on a pressure measuring branch opening out to atmosphere via a calibrated orifice.

With the process and device according to the invention, each line is continuously scavenged throughout by a flow of gas, upstream by the gas circulating in the line and, downstream and in non-operative phase, by a part of the recirculated gas from another line. In the active part of each line, only an orifice or calibrated restriction is found, which naturally causes little contamination and is in this case most unlikely to add impurities because of the continuous scavenging of the line. The contaminating components, such as the pressure sensors and flow regulators, are disposed in the branches of the line proper, also continuously scavenged by a flow of

gas. Since the calibrated restrictions in the lines function sonically, the flow rates of gas in the lines downstream of the restrictions depend only on the upstream pressure and therefore can easily be calculated from the latter, which makes it possible to ensure the precise operation, which varies little over time and can be corrected, of the system according to the invention.

Other characteristics and advantages of the present invention will become clear from the following description of embodiments, given by way of illustration but in no way limiting, with respect to the accompanying drawings in which:

- Figure 1 is a diagrammatic view of a device for the implementation of the method for supplying gas according to the invention, the gas flows shown corresponding to an analysis phase of the gas to be analysed, and

- Figure 2 is a diagrammatic view of another embodiment of a device according to the invention.

In the following description and in the drawings, identical or similar components are indicated by the same reference numbers.

The device shown in Figure 1 has three lines I, II, III, typically in the form of tubes made of killed electropolished stainless steel, for supplying gas selectively to a section of feed line 1 to a very high sensitivity analyser 2, designed to analyse traces of residual gas in a principal gas from a source of ultra-pure gas 3, typically for continuously measuring any traces in a gas produced in situ by a gas supply installation 3, for example a generator of high-purity nitrogen (HPN).

The gas from the source 3 is supplied, at a pressure generally between 5 and 15 x 10^5 Pa, to a first line I, or analysis

line, and to a line II, both being connected downstream at A to the feed section 1 to the analyser 2.

The line I includes a restriction 4, typically a calibrated orifice, functioning sonically and, upstream of the restriction 4, a pressure-measuring branch 5 provided with a pressure sensor 6 upstream of a calibrated pressure-drop device 7 allowing a very low leakage into the branch 5. The line I includes, downstream, a discharge branch 8 provided with a controllable flow regulator 9.

The line II, designed to supply the "zero" gas and the dilute calibration gas to the analyser 2 includes, upstream, a pressure regulator 10 and, in series, a purifier 11, for example of the adsorbent type or of the "GETTER" chemical purification type. Downstream of the purifier 11, the line II includes, like the line I, a pressure-measuring branch 12, with a pressure sensor 13 and a calibrated discharge orifice 14, a calibrated restriction 15 and a discharge branch 16 provided with a controllable flow regulator 17.

The line III establishes communication between a calibration-gas cylinder 18, provided with a pressure reducer 19 and containing a gas of the same type as the gas to be analysed, with the same traces but with a content around 1000 to 10,000 times greater. The line III communicates with the line II at a point B situated downstream of the branch 16 and upstream of the point A. The line III, like the other lines, includes, successively, a pressure-measuring branch 20 with a pressure sensor 21 and a calibrated discharge orifice 22, a calibrated restriction 23 and a discharge branch 24 provided with a controllable flow regulator 25.

The implementation of the method according to the invention will now be described with respect to the gas analysis phase. The restrictions 4, 15 and 23 determine, in the lines I, II and III, constant flow rates of gas Q_1, Q_O and Q_C respectively.

In the analysis phase, the lines II and III are not used, the gas from the source 3 being directly sent, via the line I, to the analyser 2. The analyser 2 functions at a given gas supply rate Q_a of around 1 litre per minute. The nominal flow rate Q_1, defined by the restriction 4, is determined so as to be considerably in excess of the given flow rate Q_a, in this case $Q_a + 2q$, q being a scavenging rate chosen arbitrarily at around 0.2 litres per minute. In this analysis phase, the regulator 9 in the discharge branch 8 of the line I is regulated so as to allow a leakage of $Q_1 - Q_a - 2q$ to escape. At point A, the flow $(Q_a + 2q)$ downstream of the line I is divided into a flow Q_a in the feed section 1 and a flow 2q going back into the downstream section AB of the line II. At point B, the recirculation flow 2q is divided into a flow q going up the line II and a flow q going up the line III. According to the method of the invention, the flow regulator 17 in the discharge branch 16 of the line II is adjusted so as to discharge, from the line II, the nominal flow Q_0 delivered by the restriction 15 and the above-mentioned recirculation flow q. Similarly, the flow regulator 25 in the line III is adjusted so as to discharge, from this line III, the whole of the nominal flow Q_c delivered by the restriction 23 and the recirculation flow q. As can be seen, all the active circuitry portions of the pipework of the device according to the invention are scavenged at the same time by a flow of gas in accordance with the object of the invention.

The functioning is similar during the other phases of gas supply to the analyser. Thus, during the supply of the "zero" gas via the line II, the regulator 17 for the line II diverts a flow $Q_0 - Q_a - 2q$, the regulator 25 for the line III diverting the same flow $Q_c + q$ as previously and the regulator 9 for the line I diverting a flow $Q_1 + q$ from the latter.

In the embodiment in Figure 2, the components of the circuitry in Figure 1 can be seen again with in this case several calibration lines, including a line III' similar to the line

III previously described, with a calibration gas source different from the cylinder 18, and a line IV for calibrating traces of gas in the liquid phase, for example H_2O starting from a point D in the line II between the purifier 11 and the calibration 15 and joining the feed section 1 to the analyser 2 at a point C downstream of the junction point A with the line I. The line IV includes, like the lines I to III, an upstream calibration 26 followed by a first discharge branch 27 equipped with a flow regulator 28, enabling a humidifier 29 to be provided with a precise, constant scavenging flow, and then a second discharge branch 30 equipped with a flow regulator 31. The humidifier 29, which may for example be of the permeator type, is supplied, at 36, by a source of pressurised liquid. The pressure sensor 13 is common to the lines II and IV. Compared with the embodiment in Figure 1, the feed section 1 to the analyser 2 is extended downstream by a first discharge branch 32 equipped with a stop valve 33 and by a second discharge branch 34 equipped with a flow regulator 35.

To ensure a particularly reliable quality of analysis and one that is independent of any pressure and temperature variations in the gas to be analysed, the device according to the invention may advantageously, as shown in Figure 2, be automated, the pressure sensors 6, 13 and 21 providing continuously, to a central control unit 38, typically of the type with a microprocessor, pressure signals which, compared with reference values stored in memory in the central unit 38, enable the latter to send control signals to the different flow regulators 9, 17, 25, 28, 31, 35 in order to adjust the opening of the latter to the actual flow circulating in the lines and derived from the pressures measured by the pressure sensors.

CLAIMS

1. Method for supplying gas to a very high sensitivity trace analyser (2), comprising the stages of sequential supply to a section of feed line (1) to the analyser (2) at a given flow rate Q_a:

a) of a gas to be analysed, via a first line (I) connected at its downstream end to the section, at a first point (A);

b) of a pure gas produced by purifying the gas to be analysed, via a second line (II), connected at its downstream end to the section, at the said first point (A);

c) of at least one calibration gas, diluted by the pure gas, initially containing a larger quantity of traces than the gas to be analysed, via at least one third line (III, III', IV), connected at its downstream end to the second line (II) at a second point (B, B') situated upstream of the first point (A), or directly to the section at a third point (C) situated downstream of the first point (A);

characterised in that the upstream parts of the three lines (I; II; III; III'; IV) each have the corresponding gas passing through them continuously at a constant flow rate, in that there is provided, in an intermediate part of each line, a discharge branch (8, 16, 24) equipped with a controllable flow regulator (9, 17, 25), the constant flow rate (Q_1, Q_0) in the first (I) and second (II) lines being greater than the given flow rate (Q_a), and in that the flow regulators are controlled, for each phase a) and b), so that a first part of the excess flow (Q_1, Q_0) of the gas in the line (I, II) corresponding to one of these phases is discharged by the associated discharge branch (8; 16) and so that the second and third parts (q) of the excess flow of the said gas are recirculated in each of the downstream parts of the other two lines, the whole of whose total excess flow proper and the

second or third part of whose recirculated flow are discharged by the corresponding discharge branch.

2. Method according to Claim 1, characterised in that the constant flow rate (Q_1, Q_0, Q_c) in each line (I; II; III; III'; IV) is ensured by a calibrated restriction functioning sonically (4, 15, 23) and in that the pressure upstream of the calibrated restriction is measured (6, 13, 21) in order to control at least the flow regulator (9, 17, 25) of the associated discharge branch (8, 16, 24).

3. Method according to Claim 2, characterised in that the pressure is measured in a pressure-measuring branch (5, 12, 20) through which a small quantity of gas escapes continuously.

4. Device for supplying gas to a very high sensitivity trace analyser (2) for the implementation of the method according to one of Claims 1 to 3, comprising;

- a source of gas to be analysed (3);

- a first line (I), including a first flow-regulation means, connecting the source of the gas to be analysed (3) directly to a section of feed line (1) to the analyser (2);

- a second line (II) connected, upstream, to the source of gas to be analysed (3) and including, in series, a gas purifier (11) and a second means for regulating the flow, and, downstream, to the section of feed line (1) to the analyser (2);

- at least one third line (III; III'; IV), including a third flow-regulation means, connected to a source of calibration gas (18, 19) containing a larger quantity of traces than the gas to be analysed and, downstream, to a downstream part

(B; B') of the second line (II) or to the section of feed line to the analyser (C);

characterised in that each line (I, II, III) includes, in an upstream part, a calibrated restriction forming a sonic neck (4; 15; 23; 26) and, in an intermediate part, a discharge branch (18; 16; 24; 24'; 30) provided with a discharge flow regulator (9; 17; 25; 31).

5. Device according to Claim 4, characterised in that each discharge flow regulator (9; 17; 25, 31) can be actuated between at least one first state in which it discharges part of the nominal flow (Q_1, Q_O, Q_c, Q_u), delivered by the restriction (4, 15, 23, 26) of the corresponding line, and a second state in which it discharges the whole of the nominal flow increased by a recirculated flow (q) from another line.

6. Device according to Claim 4 or Claim 5, characterised in that each line (I; II; III; III'; IV) comprises, immediately upstream of the restriction (4, 15, 23), a pressure sensor (6, 13, 21).

7. Device according to Claim 6, characterised in that the pressure sensor (6, 13, 21) is mounted on a pressure-measuring branch (5, 12, 20) opening out to atmosphere via a calibrated orifice (7, 14, 22).

8. Device according to one of Claims 4 to 7, characterised in that the section of feed line (1) communicates, downstream, with a discharge pipe (34) provided with a discharge flow rate regulator (35).

9. Device according to one of Claims 4 to 8, characterised in that it comprises at least two third lines (III; III'; IV).

10. Device according to Claim 9, characterised in that at least one third line (IV) is connected, upstream, to the

second line (II), between the purifier (11) and the calibrated restriction (15) of this second line, and, downstream, to the section of feed line (1), and comprises, upstream of the discharge pipe (30), a trace generator scavenged at a constant flow rate (29).

11. Device according to Claim 10, characterised in that the trace generator (29) introduces traces from a source of liquid (36) into the third line (IV).