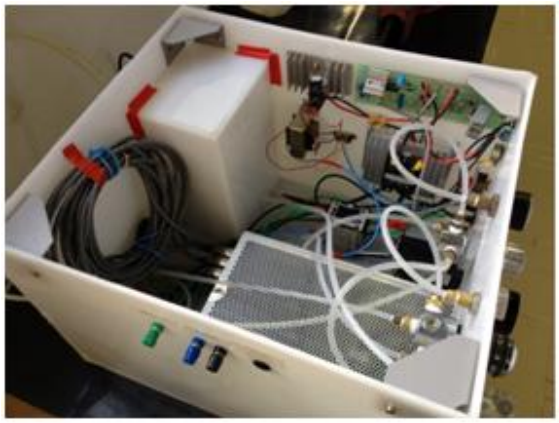
Thermal control and rate of deposition system of chemical vapor deposition at atmospheric pressure

production of thin tin-oxide films by an APCVD process



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Preface

When it was time for me to decide where I would want to do my internship, the decision was easily made to go abroad. After a really good experience in India in 2009, I wanted to discover another part of the world I hadn't been to yet: Brazil. Not knowing too much about Brazil I started reading about the country and when I arrived, this country exceeded my expectations on practically every area. I have had a most amazing experience staying here in Brazil.

The decision to go to CTI was an easy one for me since many people I know have done their internship here and the University of Twente has some good connections with the institute. Since my first day at work at CTI people have showed interest in me and made me feel welcome. The people I worked with have created an amazing working atmosphere which really made me enjoy working at CTI and enjoy my project.

The level of responsibility that I was given in doing my internship was extraordinary and quite differs from the level of responsibility we usually have when working in the Netherlands. I was free to use the resources at hand at CTI and got a lot of help from the people within the institute. Having this much responsibility and control over your own project is something of great value for me and after finishing the project, I truly learned a lot from this.

A great aspect of working at CTI is the freedom and flexibility you get in doing your internship. As long as you follow your planning and carry out your work as expected, you have the flexibility to enjoy Brazil and sometimes take a day off for special occasions or sightseeing. They really value it when you know their country a little. Something I really appreciated and enjoyed.

The project I worked on was an interesting project with many aspects. I have carried out practical work, research and learned a new programming language to write a control program for the designed reactor. This diversity in work is something interesting and has been a great learning experience for me personally; since I have not really experienced an assignment in its totality like this before.

Though I have worked alone several times before, it keeps on being a valuable aspect as it was too during my internship here. Being responsible for the end results yourself and not together, asks for different qualities from me than usually; which I really learned from.

I could not end this preface without sincerely thanking everyone who made my internship this wonderful and amazing; an experience I won't forget. I would like to thank everyone for their involvement, interests and hospitality; without you I wouldn't have enjoyed this internship this much. In special I would like to thank Thebano Santos for accepting my request to do an internship at CTI and guiding me throughout my research. Many thanks also to Luis Roberto Ribeiro without whose help I couldn't have carried out the project and Tania Lima for her support and help in arranging a lot of things here. For the rest I would like to thank all my colleagues at DMI who made my stay and work here really pleasant and truly fun.

Maarten Smit
August 2012

Summary

This report covers the design and manufacturing of a new CVD reactor for chemical vapor deposition (CVD) of tin-oxide to glass as well as several related issues such as written process control software, chemical process information and test run results. The CVD process is one step in the production of liquid crystal displays and many other applications. Currently CTI has a working discontinuous atmospheric pressure (APCVD) reactor that needs replacement to allow a process with more degrees of freedom as to conduct a more elaborate research.

The CVD process as carried out at CTI is a process that is chemically not fully understood yet, but results of deposition of tin-oxide are measured. The principles behind the general process as well as the specific process carried out at CTI are treated up to certain detail in this report as well as several important influences on the process.

The design of the new APCVD reactor concerns in fact not the entire reactor, but rather all the involved electrical equipment that is stored in a so called 'control box'. This control box houses all the mass flow control, electrical and temperature control equipment as well as equipment for the rotational motor of the heater that is necessary for the CVD process. The control box's design is optimized using 3D CAD software and both an electrical circuit as a gas flow circuit is drawn of the equipment inside.

To control the process digitally via a PC, it was necessary to communicate with the mass flow controllers (MFCs) and hence software has been written to do so. for the equipment came without any control software. The written program, named Deposition Control, is fully explained so anyone acquainted with the used software to write the program can understand and later on adapt the program if necessary.

Finally after finishing the assembly of the equipment and all the connections inside the reactor, several test runs have been carried out to assure proper functioning of the total reactor. During these tests, several improvements have been made to the equipment as well as to the Deposition Control program. This report is concluded with several recommendations for better control of the CVD process in future use.

Contents

1. Problem definition

1.1 Introduction

One of the key activities of the Brazilian research institute Center for Information Technology Renato Archer (CTI) is the research and small-volume production of Liquid Crystal Displays (LCD), Organic Light Emitting Diodes (OLED), Field Emission Displays (FED), and touch screens etcetera. Use of these applications are found in practically any digital device nowadays and hence further research is continuously ongoing.

Part of the production process of all of the above mentioned applications involve the deposition of a thin conducting layer of tin-dioxide (SnO_2) to a glass by a process called chemical vapor deposition, or shortly CVD. This process can generally be carried out in various ways in either a continuous or discontinuous process; depending on the equipment available and the production quantity needed. At CTI this process is carried out by means of a discontinuous reactor in which the substrate (the glass) is first heated to a temperature of around 300 to 400°C and subsequently deposited with SnO_2.

The deposition of SnO_2 is usually realized by the reaction of dioxide (O_2) or water (H_2O) with tin-tetrachloride ($SnCl_4$). However, at CTI the deposition of SnO_2 is realized using flows only containing $SnCl_4$ and methanol (CH_3OH); where CH_3OH and $SnCl_4$ are both carried by precisely controlled nitrogen (N_2) gas flows. This process is chemically highly interesting since it should not be possible to form SnO_2 solely with these components; as will be discussed in Chapter 2.2. Somehow CTI succeeds in depositing SnO_2 the way as described above, which yields further research for better understanding.

Two important characteristics of the deposited transparent conductive film (TCO), is that it must have a high transmissivity and a low resistivity. The resistivity of the TCO is proportional to the film thickness as where the transmissivity is inversely proportional to the film thickness; hence an optimum point has to be found between these two resulting in the most favorable layer thickness. More on the thickness of the film and characteristics of the CVD process related to this is found in Chapter 2.1.

The reactor used at CTI is a discontinuous one and hence only one substrate can be treated at a time, which is sufficient for the research done at CTI. The general principles behind the CVD process as well as the process used by CTI will be discussed in more detail in Chapter 2.

1.2 Current equipment

The current CVD reactor in use at CTI is depicted in Figure 1. The substrate is placed in the reaction chamber (within the furnace) above the heater. The substrate's support is slowly rotating to achieve an even temperature distribution on the substrate inside the reacting chamber. The temperature is of great influence on the deposition process, as explained in Chapter 2.1.1, and hence an even temperature distribution is crucial across the substrate in order to guarantee an evenly thick layer across the substrate. The temperature of the substrate is measured using a thermocouple and controlled by a temperature actuation device which is connected to the heater and can be manually set to a certain set point temperature.

The two gas flows that carry the reaction components (precursors) of the CVD process are controlled by two mass flow controllers (MKS, type 1179B) which can be digitally analyzed and controlled via a PC using specially developed software and a NI MKS-416 controller. All the equipment for controlling the rotational speed, heating actuation and measurement, mass flow controllers and pressure regulation is stored in a 'control box' for convenient reasons.

Since the current deposition process of SnO_2 from CH_3OH and $SnCl_4$ is not fully understood yet, numerous experiments have been carried out using the current equipment in order to figure out the working principles behind the process. However, important parameters involved could not be changed; such as additional gas flows for example. Therefore it is desired to have another reactor in which these parameters can be taken into account and a new series of experiments can be carried out to try to understand and determine the chemical process that is taking place.

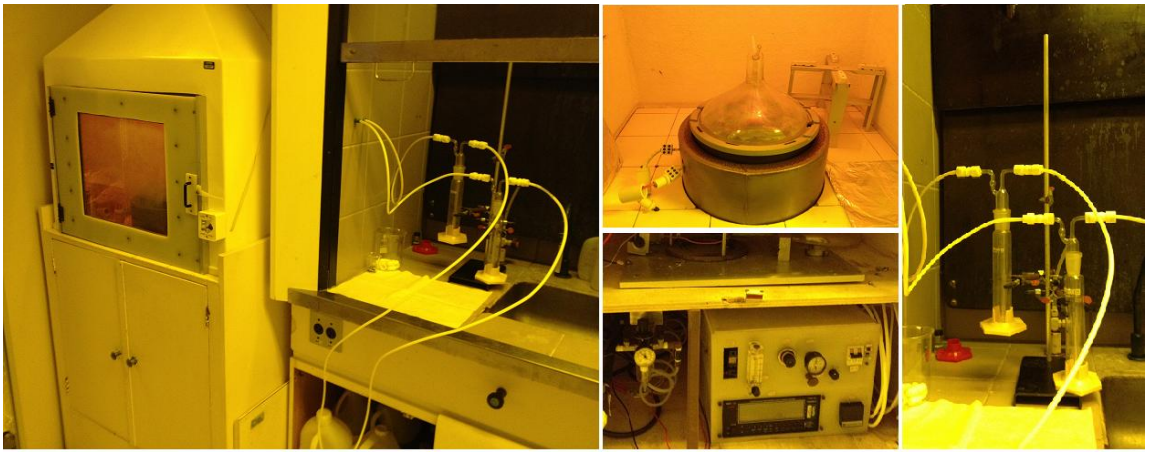


Figure 1: (clockwise) Furnace and bubblers; reaction chamber; bubblers; control box

1.3 Problem definition

Sufficient equipment is present at CTI to build another CVD reactor capable of depositing SnO_2 using a slightly different process than with the current reactor, which is desired. Also in the new setting not the substrate, but the heater will rotate; which is expected to lead to a better temperature distribution (see Chapter 2.1.1). Furthermore the chemical process of depositing the SnO_2 layer is different for an oxygen stream is also added. Together with some other small differences, this led to the following problem:

Design and build a CVD reactor that meets the following demands:

- Able of processing 200 x 200 mm substrates
- Able of controlling the temperature inside the reaction chamber
- Able of controlling the rotational speed of the heater (explained below)
- Able to measure and control the oxygen and nitrogen pressures
- The power of the entire system – i.e. all the separate equipment – should be controlled by one single switch
- Able to digitally control and measure two nitrogen and one oxygen stream
- Able to carry out the chemical process in four different phases (digitally controlled)

The last two points need some further explanation. The requirement to digitally control the deposition process yields that a program has to be written to read and control the mass flow data of the nitrogen and oxygen streams, since the equipment does not come with digital control software. The list of requirements that the program has to comply with is given in Chapter 4.3.

1.4 Problem approach

The heater of the process as well as the reaction chamber was already designed and built earlier; the rest of the equipment was only available in separate parts.

The following equipment is used in the process of designing and building the CVD reactor:

- Two 1000 sccm* nitrogen (N_2) mass flow controllers (MKS type 1159B)
- One 10 sccm oxygen (O_2) mass flow controller (MKS type 1159B)
- Vacuum gauge measurement and control system (MKS type 146C)
- Thermo couple and measurement device (Coel, type HW1440)
- Heater actuation equipment (connected to the thermo couple measurement device)
- Rotational motor control equipment
- Two pressure regulators
- Two pressure gauges (max 15 bar)
- One pressure meter (max 2 bar)

All of this equipment has to be correctly connected to one another and conveniently be stored in a 'control box'.

The project roughly consists of two parts: designing and building the CVD reactor – i.e. designing and building the control box – and writing the software used to control the mass flows and hence the deposition process. However, first of all the principles behind a CVD process in general as well as the specific process at CTI will be treated.

Finally, when the reactor is assembled and operative, several test runs will be done to assure that the equipment and software functions properly.

*sccm stands for 'standard cubic centimeter per minute'

2. The CVD process

2.1 General process

Chemical vapor deposition (CVD) is the process of chemically reacting a volatile compound of a material to be deposited, with other gases, to produce a nonvolatile solid that deposits atomistically on a suitably placed substrate. The process is often used in the semiconductor industry to produce thin films. In a CVD process the substrate is exposed to one or more volatile precursors, which react and/or decompose on the substrate surface to produce the desired deposit layer. Precursor gases, which are often diluted in carrier gases, are delivered into the reaction chamber at approximately ambient temperatures. As they pass over or come into contact with a heated substrate, they react and/or decompose. Figure 2 shows the basic principles of a CVD process.

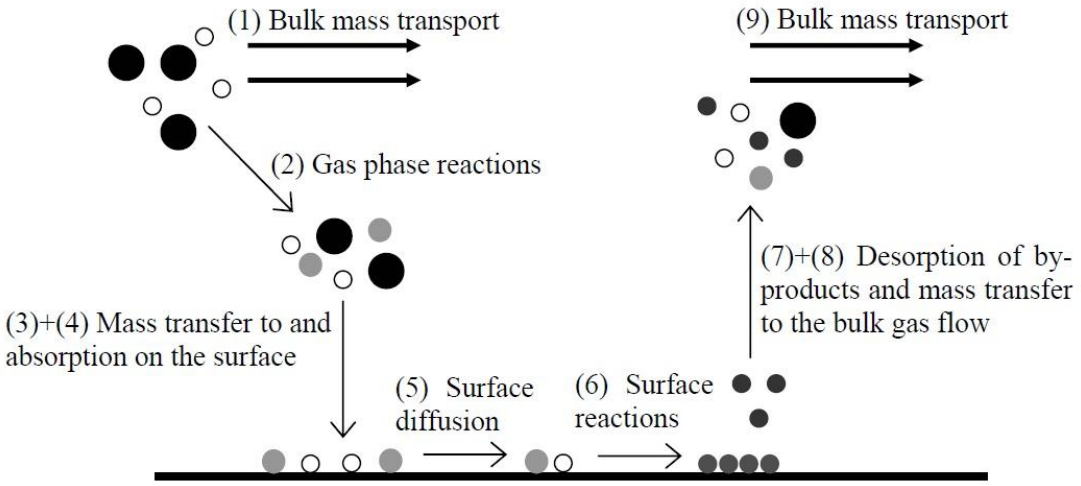


Figure 2: General principles of CVD process [1]

1. Mass transport of the gaseous reactants from the reactor inlet to the deposition zone.
2. Chemical reactions in the gas phase leading to new reactive species and byproducts.
3. Mass transport of the initial reactants and reaction products to the substrate surface.
4. Adsorption of these species to the substrate surface.
5. Surface diffusion of adsorbed species over the surface to the growth centre.
6. Surface reactions at the growth centre.
7. Desorption of by-products.
8. Diffusive mass transport of the by-products away from the surface.
9. Mass transport of the by-products to the outlet of the reactor.

The deposition process that takes place in the reaction chamber (or deposition chamber) is a precise process that requires the right proportions of reacting gases in order to guarantee the deposition results and hence a lot of equipment is needed to control the gas flows. The equipment used in the considered process will be discussed in Chapter 3.

The essential functions of CVD equipment are to create an appropriate vapor or gas mixture, and to make it flow over the substrate at an appropriate temperature. In most applications for coating large areas, CVD processes are carried out at normal atmospheric pressure. In some

specialized applications, particularly in the semiconductor industry, CVD is carried out at lower pressures. If the reactants are gases, then an appropriate gas mixture can be formed using standard mass flow controllers. If the reactant is a liquid or a solid, it must first be vaporized. Often the vaporization is done in a bubbler, by passing a carrier gas through the precursor. [1]

A CVD process can be carried out in a numerous amount of different ways; e.g. at different temperatures, different pressures and using different chemical reactions; all dependent on the desired end product and equipment at hand. The CVD reactor used at CTI works at atmospheric pressure and the process is therefore referred to as Atmospheric Pressure Chemical Vapor Deposition (APCVD).

2.1.1 Influence of substrate temperature

The substrate temperature (and reaction chamber temperature) is a critical factor the most important parameter to consider in the minimization of the electrical resistivity of tin oxide films deposited by CVD [7]. The temperature can vary from relatively low temperature of a several hundred degrees to up to 1500°C, all dependent on what the desired end result (e.g. layer thickness or atomic structure) is and what reactions are to take place. The process carried out at CTI is carried out at temperatures between 300 and 400 °C.

Not only the temperature but also the temperature distribution across the substrate is very important in a CVD process [5][6]. When a constant layer thickness is desired on the substrate, the temperature should be homogeneously distributed across the substrate's surface too, since the deposition and rate of deposition is temperature dependent. However, besides the sheer temperature and its distribution, the layer thickness and structure is also dependent on the gas mixture as will be discussed in Chapter 2.1.2.

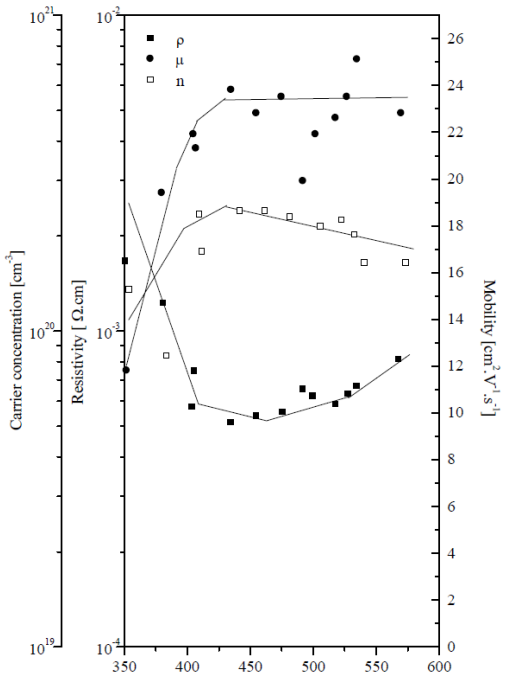


Figure 3: Influence of the substrate temperature on the electrical resistivity (ρ), mobility (μ) and carrier concentration (n) [1]

According to Antonius van Mol [1] and Vitor Baranauskas et al. [7], at low deposition temperatures the resistivity of the grown layers is high, but it decreases with increasing deposition temperature to a minimum at about 400-500°C and then increases again. Figure 3 shows the variation of the conductivity with deposition temperature for a tin oxide layer deposited in a cold wall reactor starting from dimethyltinchloride (DMTC) and O_2. Initially the resistivity decreases with increasing deposition temperature because the grain size and the crystalline of the layer increase. After a minimum value at about 450°C the resistivity of the layer increases as a result of a decreasing carrier concentration; which is caused by the elimination of oxygen vacancies [1]. Though this CVD reaction is not the same as the one used at CTI, the principles and the temperature dependence is very comparable.

In the current reactor present at CTI the substrate's support, and hence the substrate, is rotating in order to obtain an evenly distributed temperature profile as possible across the substrate's surface and inside the reaction chamber. In contrary to the current reactor, the new reactor will have the substrate fixed, but the heater rotating.

2.1.2 Influence of gas mixture homogeneity

The gas mixture homogeneity and distribution of the gas mixture across the reaction chamber is very important when it comes to the distribution of the deposited layer and hence its layer thickness and electrical resistance. For example, when the substrate is placed as shown in the left side of Figure 4 and the gas flow is from left to right, the reactants will have a relative higher density in the flow on the left side of the substrate than on the right side, where the flow is already partly depleted of reactants, and hence the layer thickness will vary in the same way; i.e. it will be thicker on the right side than on the left side. To overcome this effect in this particular case, the substrate should be tilted slightly to compensate for this effect so an evenly distributed layer thickness is obtained; as shown in the right side of Figure 4.

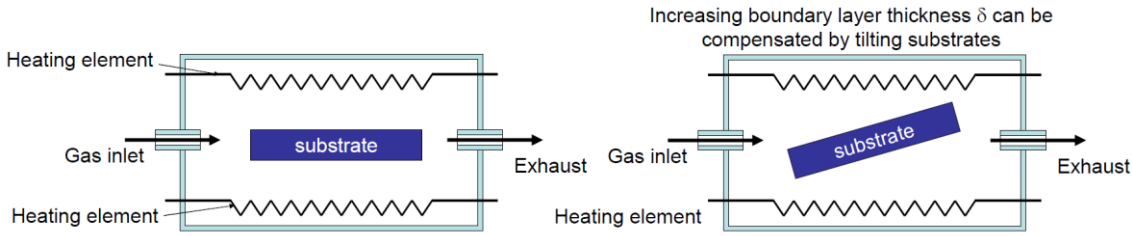


Figure 4: Effect of gas mixture and substrate placement on deposit layer thickness.

This example illustrates the importance of the gas mixture distribution on the end product and it is in fact a much more complicated fluid dynamical problem than shown in this relatively simple example. Though this topic will not be elaborated on in this report, it is presented here to bear in mind the importance of it and the influence it can have which is necessary for a better understanding of the entire process. Chapter 2.4 shows the reaction chamber as well as some gas mixture characteristics of the new reactor.

2.1.3 Influence of deposition time

When all other deposition parameters are kept constant, an increasing deposition time results in a higher thickness of the layer. In general, the electrical resistivity of the film decreases with increasing thickness to a more or less constant value. A longer deposition time results in larger grains in the crystalline structure of the film. The dependency of the electrical resistivity on the deposition time is not a single mechanism however and in some cases an increase of resistivity has been reported at thicker boundary layers [1]. When preparing and designing a CVD process, great care should be given to this to predict the expected end result of the film.

2.1.4 Other influences on the CVD process

There are many other factors that influence the CVD process and the characteristics of the $\boldsymbol{SnO_2}$ layer as is elaborately discussed in *'Chemical Vapour Deposition of Tin Oxide Thin Films'* [1] and *'The Materials Science of Thin Films'* [2]. Several other important factors influencing the CVD process are for example the flow rate of precursors, the type of substrate used and additional chemicals etc. For further reading see also reference [5] and [6].

2.2 Chemical process of SnO_2 deposition at CTI

As explained in the introduction (Chapter 1), the chemical process behind the current process is not fully understood yet and several chemical reactions are possible to occur that could realize the deposition of the SnO_2 layer on the substrate. There are many different ways of depositing SnO_2, but usually this reaction takes place either by hydrogenation or oxygenation of $SnCl_4$ by the following reactions respectively:

$$SnCl_4 + O_2 \rightarrow SnO_2 + 2Cl_2 \quad (1)$$

$$SnCl_4 + 2H_2O \rightarrow SnO_2 + 4Cl_2 \quad (2)$$

Both reactions require additional heat input (ΔT) to take place, which is realized by the temperature of the substrate. There are many other possibilities of forming SnO_2, but these will not be treated here for they are irrelevant for the case considered.

At CTI the SnO_2 deposition is realized using $SnCl_4$ and CH_3OH. When combining flows of $SnCl_4$ and CH_3OH the following possible reaction can take place where SnO_2 is formed (in two steps):

$$SnCl_4 + 4CH_3OH \rightarrow Sn(OCH_3)_4 + 4HCl \quad (3)$$

And using the formed HCl, $Sn(OCH_3)_4$ can be broken down as follows:

$$Sn(OCH_3)_4 \rightarrow SnO_2 + 2CH_3OCH_3 \quad (4)$$

Both reactions again require additional heat input in order to take place. The odd thing however about reaction (3) is that in order to break down the methanol (CH_3OH), the presence of a strong acid is needed to realize this. This strong acid is not present in the reactor and hence the formation of SnO_2 from $SnCl_4$ and CH_3OH remains quite a mystery which is not fully understood yet.

However, there are some theories about the formation of SnO_2 that may hold validity. The main theory is that since CTI is working with an APCVD reactor (non vacuum), there is normal air inside the reaction chamber which contains small amounts of H_2O depending on its humidity; which might result in reaction (2) taking place and hence the formation of SnO_2. It has been proven after several experiments that the humidity is of influence on the reaction taking place and at some levels of humidity the formation of SnO_2 does not occur at all. Also the $SnCl_4$ might react with the oxygen present inside the reaction chamber.

2.2.1 New reactor purpose

To investigate the principles behind the process taking place it is desired to let the process take place with an additional flow of oxygen, for example, to see how this affects the outcome of the deposition process. Since the current reactor does not allow an additional third flow into the reaction chamber, this yielded the design of a new reactor which does. The new reactor will be used to further investigate the process taking place and to carry out experiments allowing an additional gas flow inside the reaction chamber.

2.3 Process phases

The total cycle of the CVD process covers everything from the pressurized oxygen and nitrogen flows, to the reaction in the deposition chamber to the final exhaust of the gases. In this part the process steps will be discussed step by step. A schematic overview of the CVD process is shown in Figure 5.

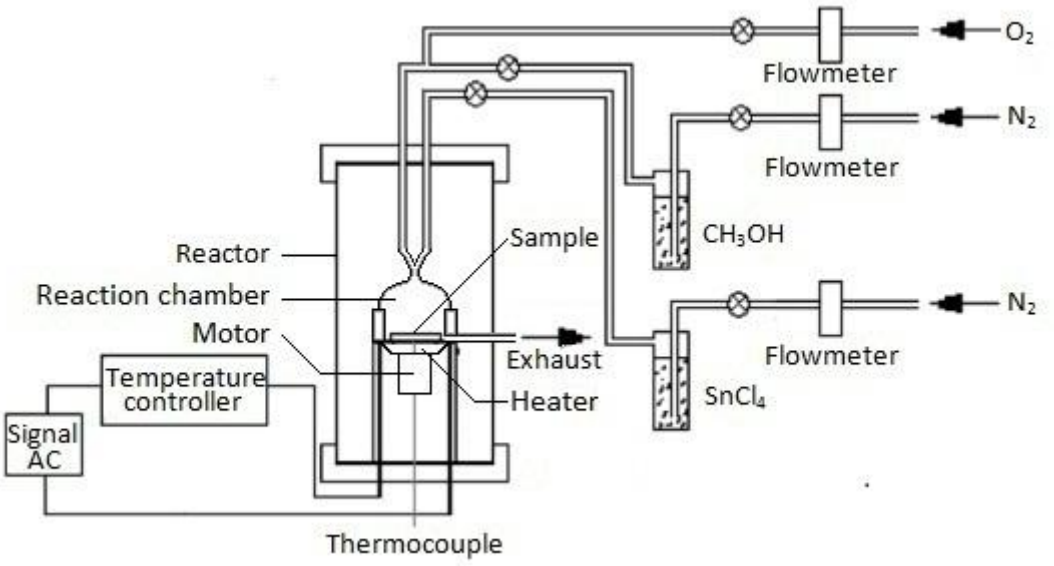


Figure 5: Schematic overview of the CVD process of the new reactor

2.3.1 Pressurized gas to the bubblers

The nitrogen and oxygen are maintained at a constant pressure of around 2 bars, which can manually be controlled by the pressure regulators on the control box. The gases are directly connected to the mass flow controllers where their flow rate is accurately controlled using the written Deposition Control software. From the mass flow controllers the two nitrogen flows go to the bubblers where they function as carrier gases for the $SnCl_4$ and CH_3OH, which together with the O_2 are the reaction components in the CVD process. The oxygen flow from the MFC is connected to the methanol stream before entering the reaction chamber; see Figure 5.

2.3.2 Bubblers to the reaction chamber

The process steps from the bubblers to the reaction chamber are the actual CVD process steps, or phases, where the reactions take place and hence the formation of the deposit layer. The formation of the layer is done in several different phases where the right proportions of precursors are realized up to the purge of the reaction components at the end of the process.

The process steps are as follows:

Phase 1: the first phase consists of a stream of mainly methanol and occasionally oxygen to fill up the entire reaction chamber. The $SnCl_4$ flow is still closed and hence no reaction is taking place.

Phase 2: In the second phase a small flow of $SnCl_4$ is added to the already ongoing flows of methanol. If not present yet, oxygen is occasionally also added. In this phase the first thin layer $SnO_2$is formed.

Phase 3: The third phase is the phase where the final SnO_2 deposit layer is formed. This phase is the grow phase where a strong flow of $SnCl_4$ is present as well as the flows of oxygen or methanol (or both).

Phase 4: The fourth phase is the final one and is the purge phase. In this phase only a strong flow of methanol is present to purge all the reactants out of the reaction chamber. There is no flow of $SnCl_4$ present in this phase and hence the growth of the deposit layer is stopped.

In the phases as described above, phase 2 and 3 can actually be combined if desired forming a 3-phase deposition process. After numerous runs with the old (current) reactor it was found that the total deposition process takes up 5 to 10 minutes to really start; hence phase 2 was introduced to assure the start of the process where phase 3 is the real deposit layer growth phase.

2.4 Reaction chamber

The reaction chamber is the most important facet of the CVD equipment since this is where the reactions take place. The reaction chamber is depicted in Figure 6.

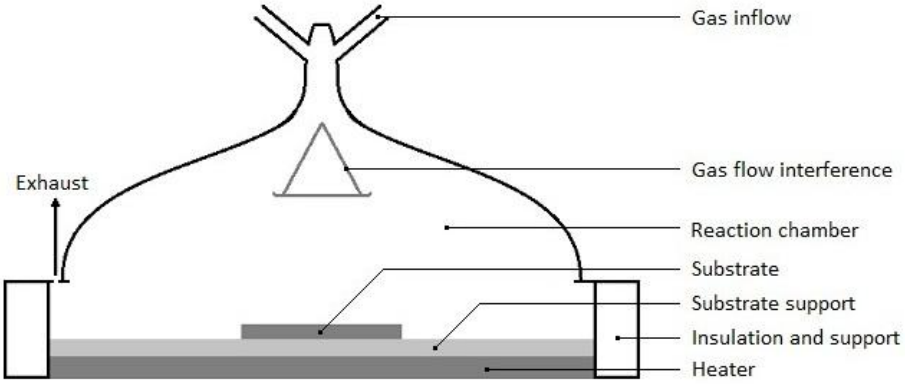


Figure 6: Reaction chamber of the new reactor

Since the two inflowing (reacting) precursor streams flow into the reaction chamber right above the substrate, a gas flow interference plateau is placed to prevent the flows from directly flowing towards the substrate and creating forced flow reactions. Since the plateau redirects the precursor flows, the flows become more or less random and cause natural and random reactions and layer growth on the substrate.

Also the gas flow interference plateau catches any solid substances resulting from reactions in the top of the reaction chamber so to protect the substrate right underneath it.

2.5 Heater, insulation, support and exhaust

The heater, insulation and exhaust holes are depicted in Figure 7. The heater consists of a resistance heating wire that is embedded in a low conducting ceramic plate to direct the heat towards the substrate's support. The substrate's support is a solid stainless steel plate with good heat conducting properties.

The thermocouple is put inside the center of this steel plate to measure its temperature. It is assumed that the substrate will approximately have the same temperature as the steel plate. This assumption is valid since the plate can store a sufficient amount of heat and has good heat conducting properties; hence the small (thin) substrate will approximately have the same temperature.

Insulation is placed both around the heater as well as around the substrate's support to keep the heat inside the deposition chamber. The insulation around the heater is to prevent unnecessary energy losses and ensuring an optimum heat transfer toward the substrates support instead of in other directions.

As can be seen in Figure 6, the glass dome (with the precursor inlets) is placed on top of the structure depicted in the right side of Figure 7. From the outside to the inside of the reaction chamber small hole are present all around the glass dome, which function as the exhaust holes (see Figure 7).

Figure 7: (left) heater and insulation; (right) Substrate support and insulation

3. Design and manufacture of the CVD control box

3.1 Problem definition

Controlling the mass flows, temperature, pressure and the rotational speed of the heater yields a lot of electrical and other types of equipment. It is convenient to have this equipment stored all together in a so called control box for practical reasons. The control box is meant to contain the following equipment:

- Two 1000 sccm nitrogen (N_2) mass flow controllers (MKS type 1159B)
- One 10 sccm oxygen (O_2) mass flow controller (MKS type 1159B)
- Vacuum gauge measurement and control system (MKS type 146C)
- Thermo couple and measurement device (Coel, type HW1440)
- Heater actuation equipment (connected to the thermo couple measurement device)
- Rotation motor control equipment
- Two pressure regulators
- Two pressure gauges (max 15 bars)
- One pressure meter (max 2 bars)
  (See Figure 5 and Figure 10 for visualization of the use of the equipment)

Besides containing the above listed equipment, the control box also has to comply with the following requirements:

- The control box should have one main power switch which powers all the equipment at once as well as a separate switch for the rotational motor of the heater.
- Since a certain space is reserved for the total CVD equipment at CTI, the maximum dimensions of the box should not exceed 500 x 500 x 400 mm (length x width x height).
- The control box should have the following electrical outputs:
  - Output for thermocouple
  - Output for earth wire of the heating equipment
  - Power output for the rotational motor
  - Power output for the heater
  - Output for RS-232 communication cable
- There should be a constant overpressure inside the control box; this will be explained in Chapter 3.1.2.

3.1.2 Hydrochloric acid backflow and leakage

Before and after every deposition process, or series of processes, the equipment used is thoroughly cleaned. It is possible that during the cleaning there is still water remaining inside the tubes that carry the gasses from the bubblers to the reaction chamber. In some cases it has been experienced that the $SnCl_4$, in the absence of additional heat, get hydrogenated and forms tin(IV)chloride-pentahydrate ($SnCl_4 \cdot 5H_2O$) crystals (solid) which block the tube. While the nitrogen is still running, a certain pressure is build up at the bubblers side due to the blockage and in time small amounts of hydrochloric acid gas (HCl) are formed. When the stream of nitrogen is turned off, the created overpressure on the bubblers side can cause the HCl, a very volatile and corrosive gas, to flow back towards the mass flow meters. Since the

mass flow controllers are not designed to cope with HCl, leakage may occur and cause corrosion to the equipment inside the control box. Therefore it is necessary to have a constant slight overpressure inside the box to purge HCl in case it leaks from the mass flow controllers. Though it is not very likely to occur, these necessary precautions need to be taken just in case.

3.2 Design of the control box

In the design phase of the control box care is given to obtain an as practical layout of the equipment as possible, taking into account the most practical positions of all the outputs and controls for practical matters. Also enough space must be available inside for maintenance purposes and special care is given to the HCl backflow problem.

3.2.1 Hydrochloric acid backflow and leakage

To cope with the problem as described in Chapter 3.1.2 several measures can be taken to realize the overpressure inside the control box. To realize this overpressure, a constant stream of nitrogen will be used which can be controlled with a pressure meter on the front panel of the control box. However, instead of creating an overpressure inside the entire control box, it is chosen to isolate the three mass flow controllers from the rest of the equipment with a small airtight box so that only an overpressure inside this smaller space has to be realized; which is much more efficient and safe. The airtight box is placed around the mass flow controllers and the gas can be purged out via ventilation holes in the sides of the control box. From the pressure meter on the front panel which controls the purge flow of nitrogen a tube is connected to the inside of the airtight box.

3.2.2 SolidWorks 3D model

The main dimensions are determined by the controls on the front panel (pressure gauges, MKS-416 controller, switches etc). To determine the final dimension most of the equipment has been modeled in SolidWorks 2011, a 3D CAD program, to visualize the design and ensure all the equipment will fit in properly. The design is shown below in Figure 8. The final dimensions of the design are 450 x 410 x 305 mm (L x W x H).

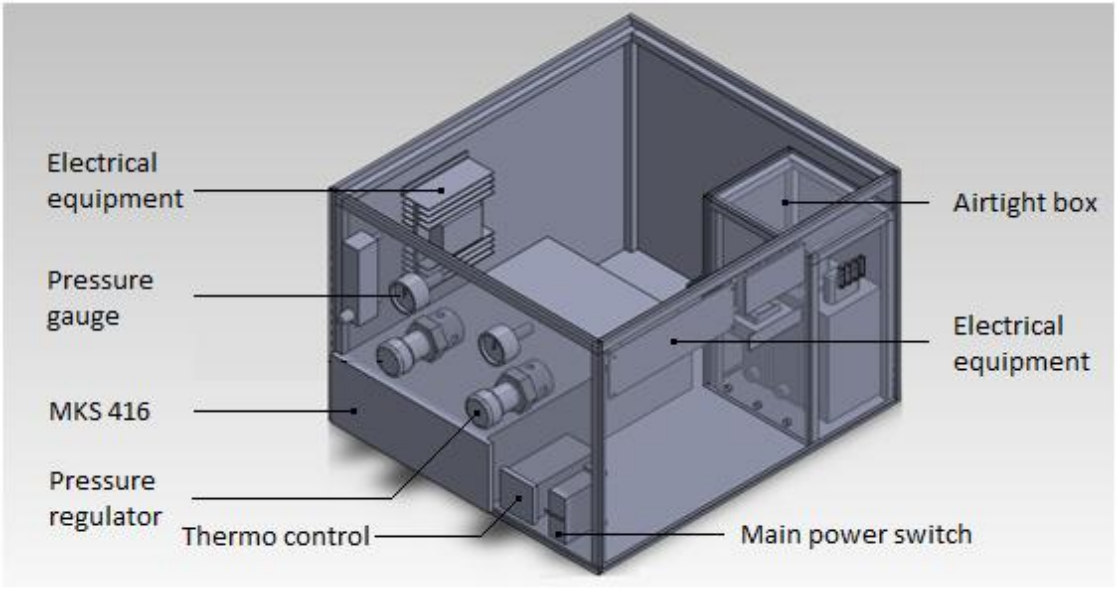


Figure 8: Design of the control box and placement of equipment

3.2.3 Electrical layout

All the equipment used for the CVD process runs on 110V, except for the MKS-416 console and the heater which both run on 220V. Therefore the system is powered by two plugs, one for each voltage. To realize the requirement that the system must have one main switch to turn on/off all the equipment, a relay switch is used. This particular switch is convenient since it works with a maximum amperage level and if that limit is exceeded the switch is turned off automatically; much like the effect of using a fuse.

Most of the equipment consists of multiple printed circuit boards (PCBs) which are interconnected and hence the full electrical circuit of the equipment inside the control box is quite complex. For this reason a simplified electrical circuit is drawn which clusters the PCBs of the thermocouple and the rotational motor. The scheme is shown in Figure 9.

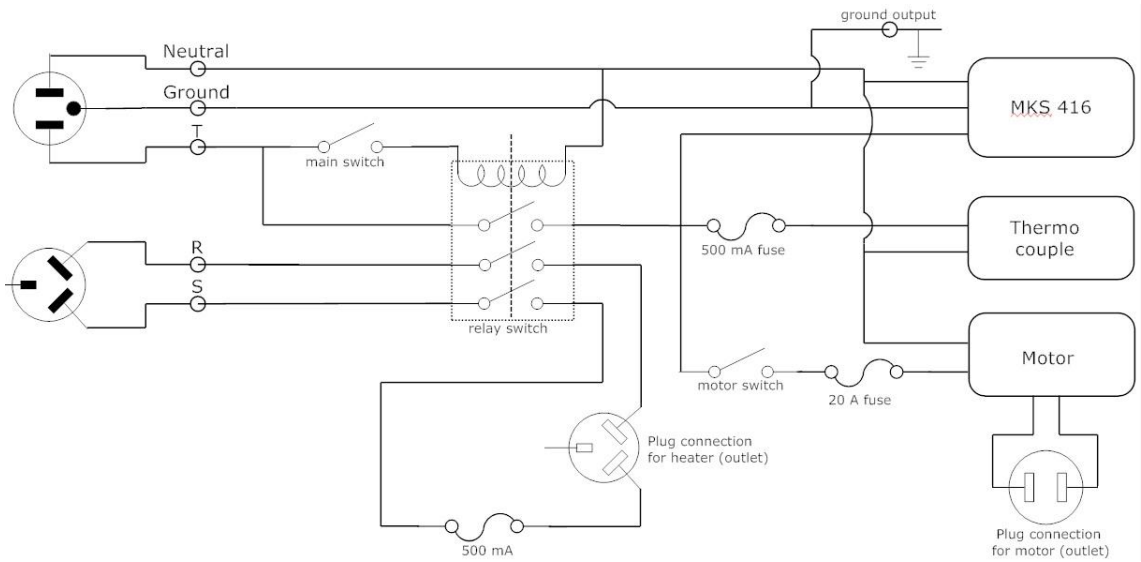


Figure 9: Electrical circuit of the components inside the control box

3.2.4 Gas flow circuit

The CVD process makes use of several streams of nitrogen and oxygen which are connected to pressure gauges, meters, regulators and the mass flow controllers. Much like the electrical circuit, a gas flow circuit has been made to show the gas flows and all the connections to the equipment from the pressurized oxygen and nitrogen tanks to the exhaust of the reaction chamber. The gas flow chart is shown in Figure 10.

Notice that the bigger dashed box indicates what equipment is placed inside the control box and the smaller dashed box (inside the control box) indicates the airtight space where the mass flow controllers are places. The stream from the 'pressure meter (purge)' is the N_2 gas flow that is used to create the overpressure inside the airtight box.

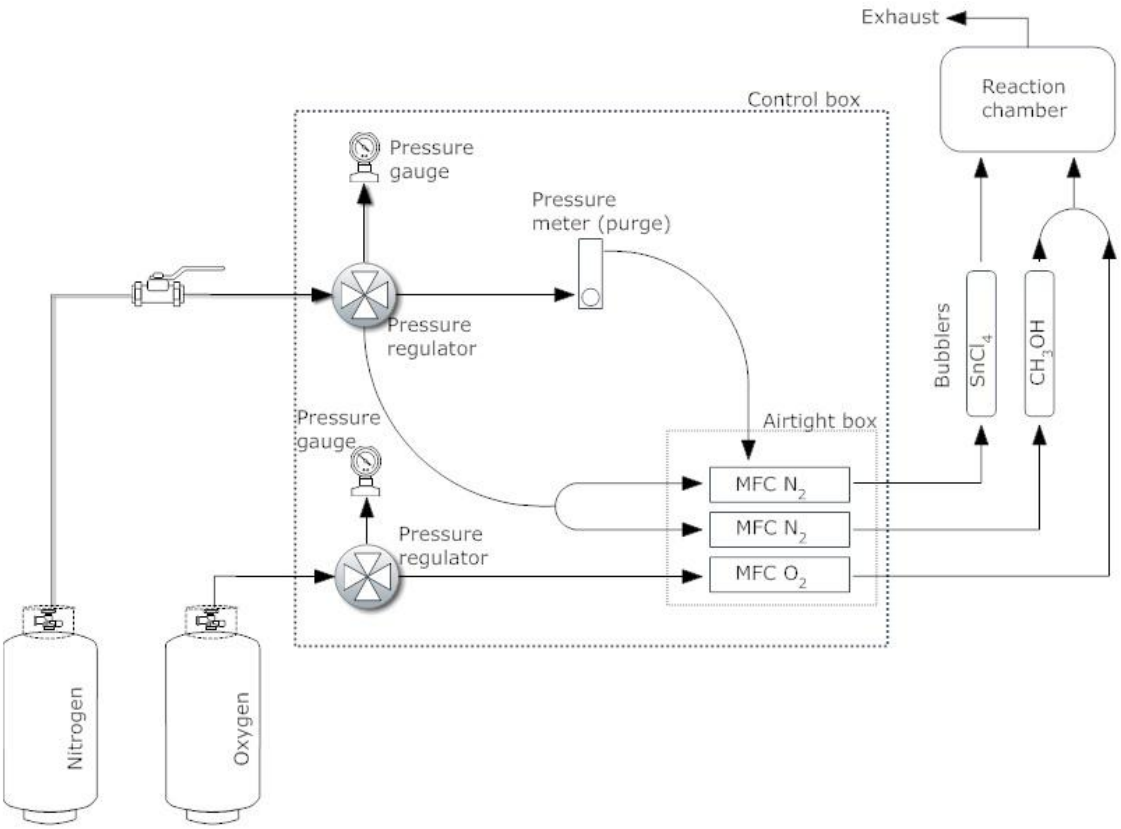


Figure 10: Gas flow layout of entire CVD process

3.3 Final result

For manufacturing the control box 5 mm thick plates of polypropylene (PP) are used. After cutting and preparing each plate for assembly, the plates are soldered to form the box. The final results are shown below in Figure 11 to Figure 14.

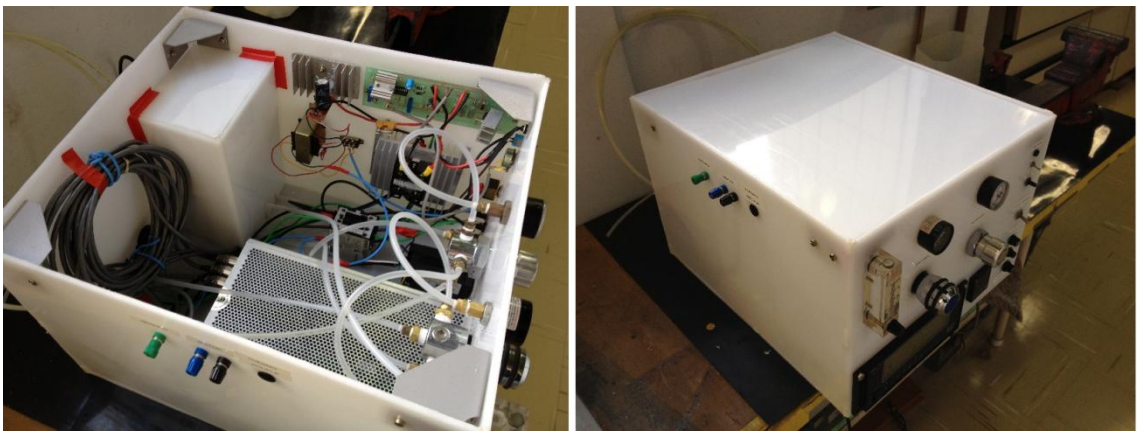


Figure 11: Isometric view of control box (open and closed)

Figure 12: Front panel of control box

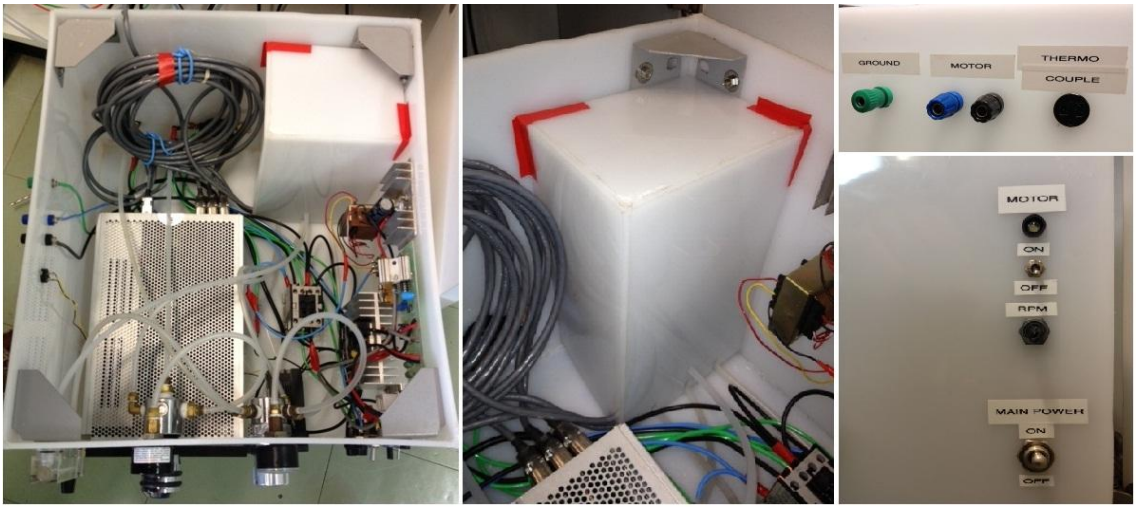


Figure 13: (clockwise) Top view; Airtight box; Electrical outlets (ground, motor and thermocouple); Main power switch and motor control

Figure 14: (left to right) Furnace and bubbler chamber; idem with computer; idem with control box

4. Deposition Control software

In order to digitally control the CVD process, software has been written to continuously acquire and control the mass flow data of the three mass flow controllers using a PC and the MKS-416 device. The written program is named Deposition Control and is designed using the software LabVIEW 8.5; see Chapter 4.2. In case the program might be adapted in the future, the general functioning principles of it will be discussed in this chapter; so anyone acquainted to LabVIEW can get a basic understanding of it. First off all the MKS-416 console and programming software will be discussed briefly; hereafter the written software will be treated.

4.1 MKS-416 Console

The MKS-416 console, shown in Figure 12, is used to control and acquire data from the mass flow controllers as well as to set all the important parameters before operation. This console is designed to be controlled manually and the data can be read from the console's screen directly. However, the console can also be connected to a PC for digital data analyzing and mass flow control. The connection between the console and a PC is realized using a RS-232 communication cable; detailed information about the connection of the device to a PC can be found in the MKS-416 manual.

4.2 Programming software: LabVIEW

In order to establish communication from the PC with the MSK-416 device, a program is written using the software LabVIEW (version 8.5). LabVIEW (short for Laboratory Virtual Instrumentation Engineering Workbench) is a system design platform and development environment for a visual programming language from National Instruments. The software allows the engineer to design a user interface (UI) for data communication as well as to design the data flow-sheet that realizes all the communication and the sequential order in which all the steps and commands are executed. The UI is easily understandable and user friendly, whereas the data flow-sheet – the actual programming behind it all – is rather complicated; hence the data flow sheet will be discussed and explained. The final results of the data flow-sheet and the UI can be found in Figure 15 and Figure 16 respectively.

4.3 Deposition Control software

The program that has been written for the CVD process at CTI, called Deposition Control, had to comply with the following requirements:

- Variable number of mass flow phases (maximum 4)
- Able to control the mass flows for the three separate flows in each phase
- Able to control the timing in each separate phase
- Able to read elapsed and remaining process time
- Read and show the mass flow data of all three flows continuously
- Write the process data to a file every time the process is started

The final result of the program, the data flow-sheet, is shown in Figure 15. Chapter 4.3.2 and further will discuss every part of the data flow-sheet in order to provide information about the working principles behind it for a better understanding of the software.

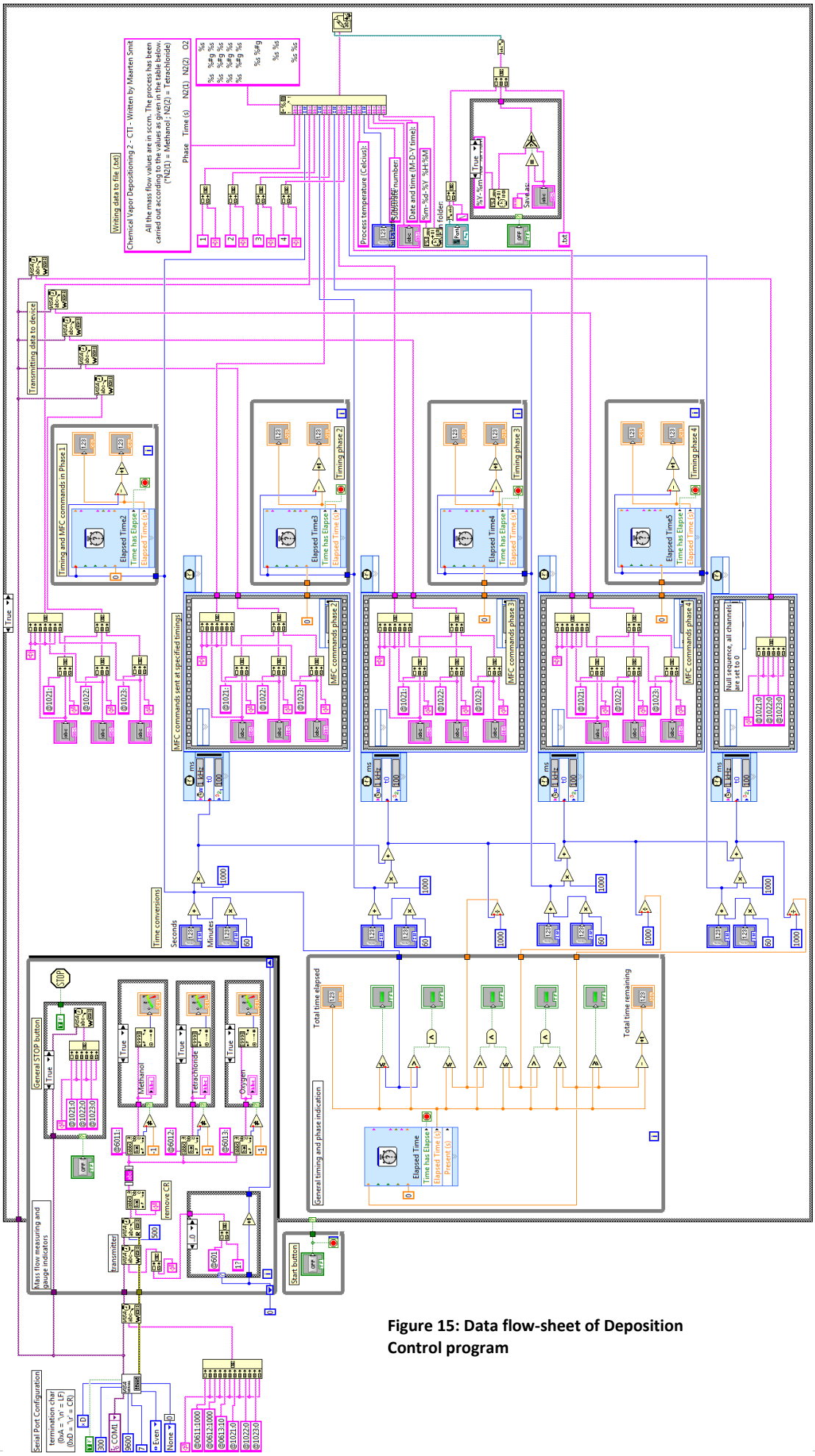


Figure 15: Data flow-sheet of Deposition Control program

4.3.1 The user interface

The UI of the Deposition Control program is the actual program as it appears to the user and is shown below in Figure 16.

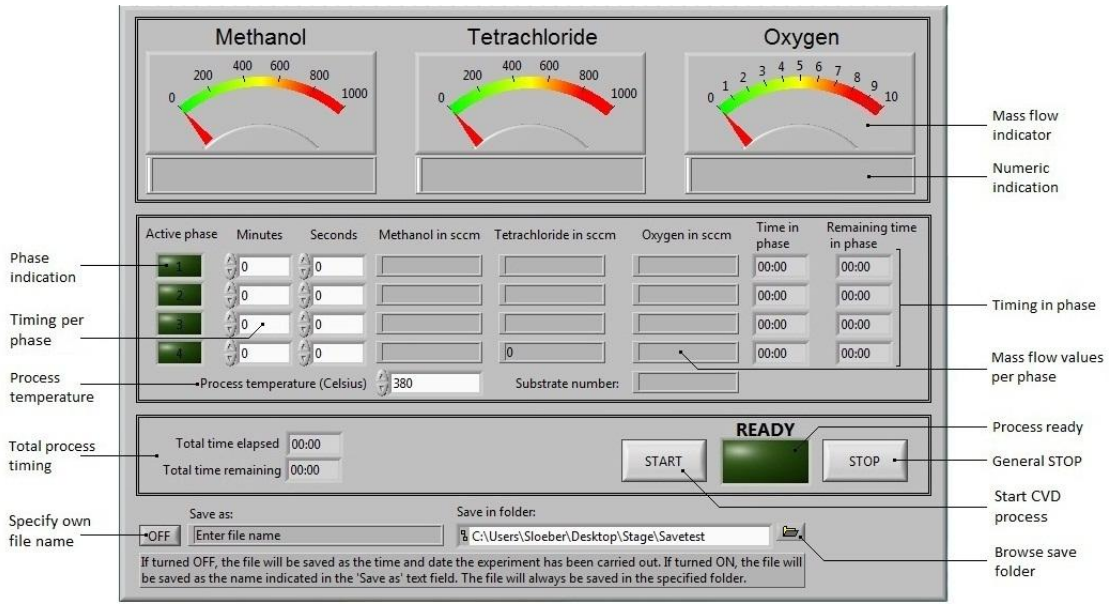


Figure 16: User interface (UI) of the Deposition Control program

The UI is rather simple and needs little explanation. The UI is where the process parameters can be set; i.e. the values of the different flows in each phase and the timing of each phase. On the upper part of the UI the values of the three different streams can be read. The total elapsed and remaining time of the entire process as well as of each phase are shown on the center of the UI. The lower part of the UI is used for specifying a name and path for the program to save the data of the process in after completion.

As can be seen the UI also has a few indicators which show in which phase the process is. In this case the light will change color from dark-green to light-green. Another indicator is the 'process ready' indicator which turns to light green when the process has finished.

4.3.2 Setting the initial values of the MKS-416

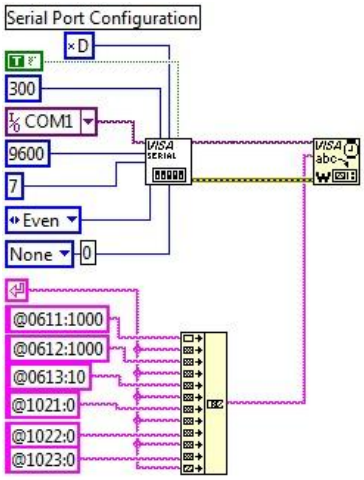


Figure 17: Code for setting the MFC ranges and initial values

The part of the code that sets the initial values of the MKS-416 console is shown in Figure 17. This concerns the initial values and the ranges of the mass flow meters. The upper part sets the serial port (RS-232) configurations (default MKS-416 values, see the manual [3] or Chapter 4.3.7). Notice that the values set here must match the values manually set on the MKS-416 console; else the communication will fail.

The lower part sends six commands to the MKS-416 console which sets the ranges of the three mass flow meters and the initial zero-values. Since the ranges of the mass flow meters are fixed, the ranges cannot be changed on the UI but can only be changed by changing the constants in the data flow-sheet.

4.3.3 Data acquisition and indication from mass flow meters

The part of the code that realizes the constant data acquisition from the mass flow controllers and shows the mass flow values on the UI is shown below in Figure 18.

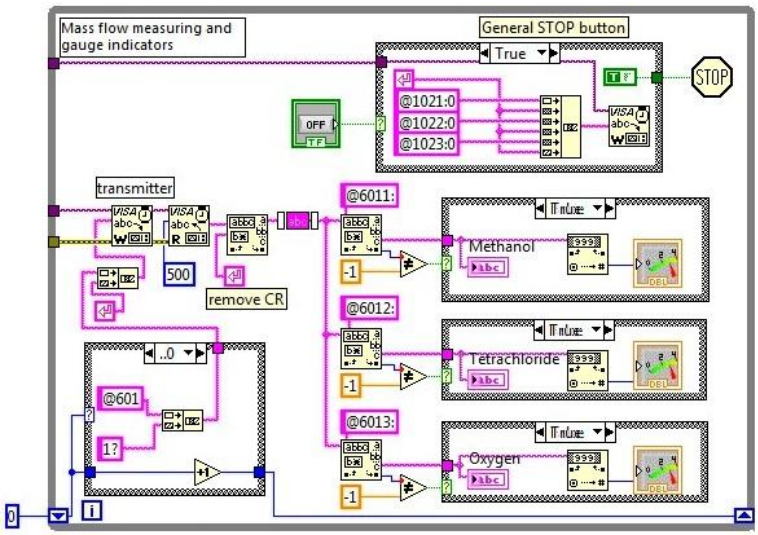


Figure 18: Code for continuous data acquisition and value indication from mass flow controllers

The total code is surrounded by a while loop which runs during the entire process. The left part of the while loop shows a small structure which is responsible for the data send to the MKS-416 console to request the value of the mass flow controllers. This is a so called case structure which changes its code inside with every iteration of the entire loop in order request the status of every mass flow controller in sequence, after which the total loop restarts; hence the status of every mass flow controller is sequentially checked, continuously.

The right side of Figure 18 reads the response from the MKS-416 console containing the values of the mass flow controllers. Since the response is a long string of characters, the string it cut down and manipulated so it only contains the numeric value of the mass flow controllers in the end and the result is linked to the correct mass flow meter and indicator on the UI.

Also the general STOP button is found in this piece of code, at the top of the main loop. If pressed this button makes sure the entire process is stopped at once. An important facet here is the case structure that makes sure that the MFCs are set to zero before the entire process is stopped. If this is not done, the MFCs will continue the process with the last received mass flow data (in the phase the process is stopped); hence the process will not stop, but only the program. Using this structure the program as well as the process is stopped.

4.3.4 Controlling the mass flows and time in each phase

In every phase of the process commands are sent to the MKS-416 console and the program keeps track of the time. This is done by the code structures as shown below in Figure 19.

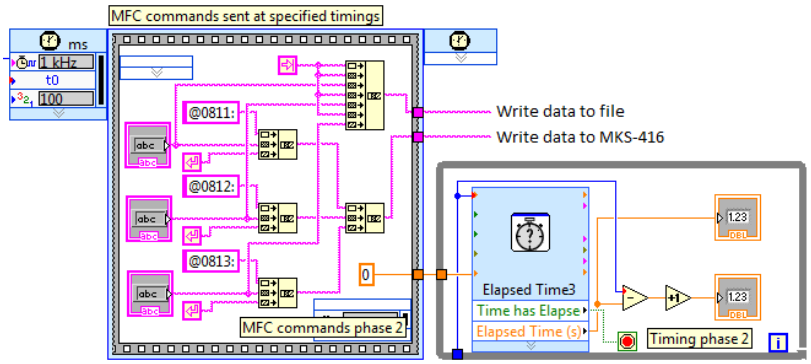
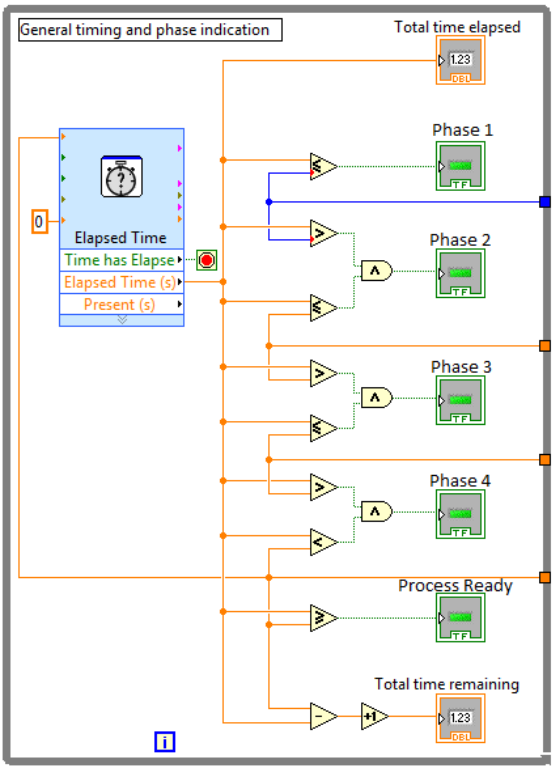


Figure 19: Code that is responsible for controlling the mass flows and timing in each phase

The left structure is a timed sequence which is a structure which only executes once at a given timing. This structure is used to send the mass flow details for each phase at a certain time as specified by the user in the UI. Inside the structure the codes are assembled which set the desired values of the mass flows and are then send to the MKS-416 device. The codes use the specified values for each mass flow meter as specified by the user in the UI.

Each sequence structure activates a while loop at the same time the commands are send to the MKS-416 console; thus at the beginning of each phase. This while loop is shown in the right side of the figure and upon activation of this loop a timer starts running until the next phase starts. The timer shows the total elapsed and remaining time in each phase, which is both shown on the UI, and is automatically stopped at the end of each phase.

4.3.5 Phase indication and general timing



The phase indication on the UI and the 'process ready' indicator, see Figure 16, is controlled by the code as shown in .

This while loop contains an 'elapsed time' block, which keeps track of the total timing and time remaining of the entire process. This block is used to compare the total timing with the times set per phase and these results are used to determine the active phase, using several comparison operators.

Figure 20: Code for the phase indication on the UI and the general timing of the process

4.3.6 Write data to file

All the by the user specified values – i.e. the mass flows and phase times – are stored in one text file using the code as shown in Figure 21.

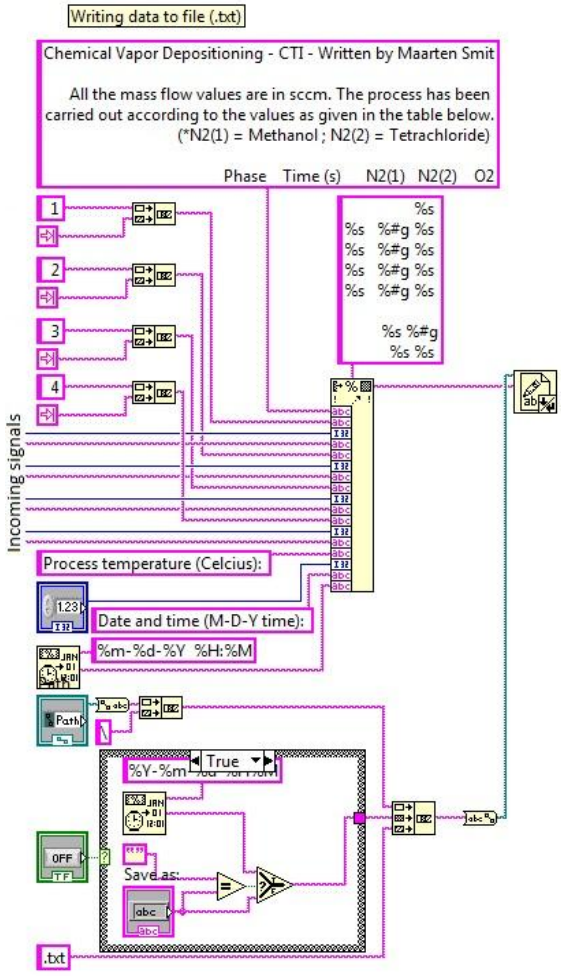


Figure 21: Code for writing the process data to a text file

The middle part of Figure 21 (the light-yellow rectangular box) is used to combine all the incoming strings into one piece of text which is subsequently written to a text file.

Just below the incoming signals the part of the code is found that saves the process temperature.

The lower part of the code, below the 'Date and time' string, specifies the path to the folder in which the results are saved and the name of the file. This part of the code included a case structure which determines the name of file. If the option 'Save as:' on the UI is turned *OFF*, the file will be saved as the date and time the process has been carried out. If the option is turned *ON* the file will be saved as the name specified in the field below '*Save as:*' on the UI. When no name is specified however and this option is turned *ON*, the file will be saved as the date and time the process has been carried out. This is to prevent data overwriting when the file is saved without a name multiple times.

The file path – i.e. the path containing the folder in which the file will be saved – can be selected by clicking on the 'browse folder' button next to the shown file path. For an example of the saved text file, see Figure 22.

4.3.7 More information on the program

After the process has been finished a null sequence follows, which looks like the structure used for sending the mass flow commands; see left structure in Figure 19. The null sequence sets all three flows to zero after the deposition process is finished.

The serial port communication settings of the Deposition Control program are set to the default settings of the MKS-416 device. In this way, when the device crashes and goes back to its initial settings, the communication with the PC will not be affected. The port settings are as follows: baud rate: 9600, data bits: 7 and parity: even.

More contextual help on the different operation blocks used in the code for the deposition control software can be found by using LabVIEW's '*context help*' which gives detailed information on the function blocks used when scrolling over the code.

The Deposition Control program code does not contain any subVI's and hence the program's code is all stored in just this one file as depicted in Figure 15.

4.3.8 How to use the program for a CVD process

The user guide of how to use the Deposition Control program for a CVD process can be found in *Appendix 2: Deposition Control user guide*.

4.4 Deposition Control process data file

The text (.txt) file in which the process data is stored after each run is shown in Figure 22. To change the text that appears in the file, the text boxes inside the LabVIEW code, see Figure 21, has to be changed.

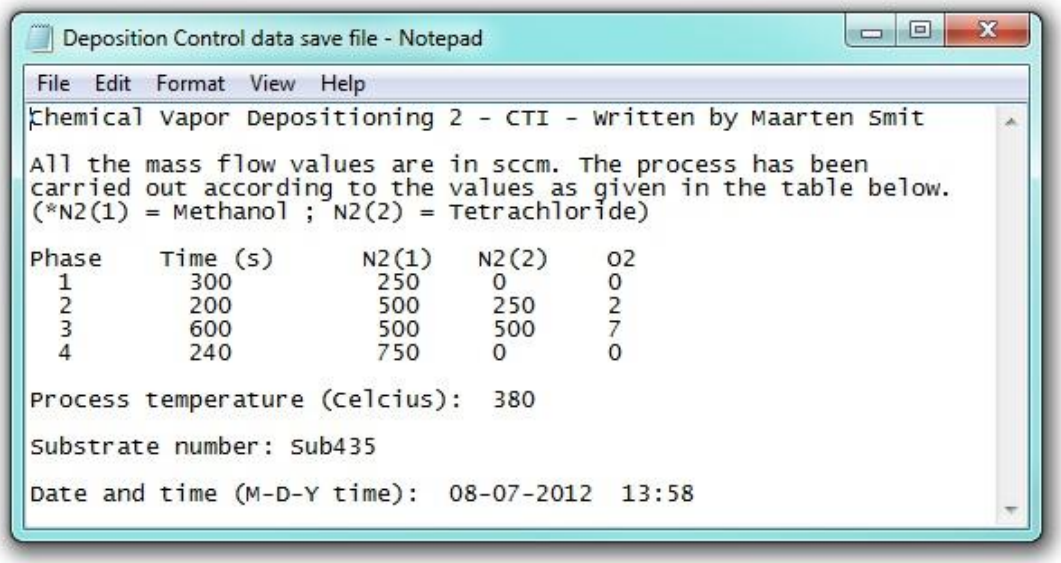


Figure 22: Deposition Control process data file

5. APCVD Reactor test runs

After assembly of the control box, connection to the rest of the equipment and installation of the Deposition Control software, several test runs have been carried out. These test runs provide some important information on the process and equipment used. Several important aspects will be discussed here.

5.1 Deposition process parameters

The parameters of the deposition process that has been carried out for testing purposes are shown in Table 1.

Table 1: Deposition test run process parameters

Phase	Time (min)	Methanol (sccm)	Tetrachloride (sccm)	Oxygen (sccm)
1	5:00	500	0	0
2	3:00	250	750	0
3	1:00	500	0	0
4	-	-	-	-
Process temperature: 380°C				

As can be seen this trial run is a 3-phase deposition process and no oxygen is used; this is done to mimic the deposition process with the old reactor from which the results are known and can hence be compared.

5.2 MFCs zero value and calibration

5.2.1 MFCs zero value

The MFCs show a (small) inaccuracy when their value is set to zero. In operation mode the MFCs report their values as set in the Deposition Control program with a slight inaccuracy of around ±0.5 sccm. However when the MFCs are set to zero all three of them show different inaccuracies:

- Methanol zero value: +20 to +70 sccm (temperature dependent)
- Tetrachloride zero value: 0±1.0 sccm
- Oxide zero value: -1±0.02 sccm

In the current reactor settings, the zero values of the mass flow meters are good. The fact that the methanol stream is always positive, even when set to zero, is no problem since the methanol is actually used in every phase with a flow value higher than the zero value. Also the methanol is used to purge the reactants from the reaction chamber; hence a small constant stream of methanol is not harmful.

The tetrachloride stream is completely shut down when set to zero, which is very important since the flow of tetrachloride determines the reaction rate. The oxygen stream of -1±0.02 sccm, when set to zero, is harmless. However, when checked the stream turned out to be slightly positive instead of negative.

5.2.2 Calibration of the MFCs

Since the zero values of the oxygen and methanol stream are not exactly zero, this yields that their mass flows when set to a certain other value, e.g. during the process, might not be accurate too. It is therefore highly recommended that the sensors are calibrated for better control of the deposition process.

5.3 Temperature control

5.3.1 Heating time

The total heating time of the substrate's support takes about 90 minutes to reach a temperature of 380°C from a start temperature of 20°C. The heating time is quite high and it is observed that the heating goes much faster when the door of the furnace is left open instead of closed. For comparison, the heating time of the old reactor is about 60 min. The long heating time is of course related to the power of the heating element but it is also a result of the temperature control as will be discussed in Chapter 5.3.3.

5.3.2 Temperature drop

During the process the temperature dropped from 380 to 350°C on average, which is about 10% of the process temperature. The temperature should increase again as soon as it drops below 380°C and the heat dissipation inside the furnace (reactor) is small enough for the heater to be able to counteract the temperature drop. In other words, there is no reason for the temperature to drop as much as it does if the system was correctly controlled.

5.3.3 PID temperature control

The temperature is controlled by the COEL HW1440 temperature controller which uses a PID controller to determine the corrective action if the temperature deviates from the set-point. The PID controller's parameters are calibrated according to a certain temperature inertia of the system, which is different for every system and dependent on the equipment used; hence this determines how appropriate a controller is for the system considered.

Since the HW1440 temperature controller has been used for another system before, the PID is most likely not calibrated to suit the current system. Even though eventually the controller will correct any deviations from the set-point temperature, the corrective actions will be much faster and accurate when the PID parameters are calibrated to suit the current system.

5.4 Substrate thermoshock

After the substrate's support is heated to about 380°C, the substrate is placed inside the reaction chamber directly on the stainless steel support. As was quite expected however, after a few seconds the glass substrate just breaks due to a thermoshock. The temperature difference between the substrate's support and the substrate itself is too big and the sudden increase of temperature results in a rapid thermal expansion the material cannot cope with.

5.5 Results

5.5.1 General

The test runs showed that the reactor and all the equipment works as planned. The following can be concluded:

- The system is airtight (there is no, or very little, gas leakage)
- The temperature control works properly (aside from the PID settings)
- The rotational motor control works properly
- Mass flow control works properly
- The Deposition Control program works properly
- Power ON/OFF works properly
- The APCVD process can be carried out using this reactor

Aside from the fact that everything works properly and deposition processes can be carried out using this reactor, there are a few issues that need to be improved as will be discussed below and in Chapter 6.

5.5.2 SnO_2 deposition layer distribution

Since the deposited SnO_2 film is semi-conducting, its thickness can be determined measuring the conductivity of the material. After several measurements it is concluded that the thickness lies within the right range, as was expected with the used process parameters, but it is not uniform across the surface. This non-uniform layer thickness implies a non-uniform temperature distribution across the substrate, an inhomogeneous vapor mixture inside the reaction chamber, forces flow layer growth chamber or all of the above.

5.5.3 Vapor mixture

It is observed that at the top of the reaction chamber where the two streams (methanol and $SnCl_4$) enter, a white crystalline powder is formed which might imply the reaction of $SnCl_4$ with water [9]. The crystals are caught by the gas flow interference plateau, see Figure 6, and do hence not contaminate the substrate. However, the formation of the crystals is not desired.

It is also observed that the flow is not homogeneously distributed across the reaction chamber; this can actually be seen with the naked eye.

5.6 System improvements

After the first few test runs with the new reactor, several changes have been made to the process and/or equipment for improved functioning.

5.6.1 Substrate thermoshock

The effect of the thermoshock has been counteracted by using a small carbon support for the substrate as to decrease the rate of heat transfer when placed in the reaction chamber. With the lower rate of heat transfer it will take a little while longer to get the substrate to the required process temperature, but the substrate will not suffer from a thermoshock anymore which is essential. The estimated time it takes for the substrate to reach the process temperature is about 5 minutes.

5.6.2 Thermocontrol PID parameters

Since the temperature control of the system was not optimal during the first settings, the PID controls of the HW1440 have been tuned to suit the system and so to improve the temperature control of the CVD process. After calibration the P, I and D values of the PID controller are respectively 0.2, 127 and 19. Using these values the temperature characteristics of the process are as follows:

- Heating time (from 20°C to 360°C): 50 min
- Temperature drop during process: 0 °C

5.7 APCVD process characteristics

Important characteristics about the reactor and the CVD process are given below in Table 2.

Table 2: Process and equipment characteristics

General	
Process type	Atmospheric Pressure Chemical Vapor Deposition (APCVD)
Substrate material	Glass
Max substrate dimensions	200 x 200 mm
Deposition film	SnO_2
Precursors	$SnCl_4$ and CH_3OH and O_2
Carrier gas	N_2
Process pressure	2 bars
Process temperature	About 400°
Temperature Control	
Total heating time	Circa 50 min
Temperature drop during process	No temperature drop
P, I and D parameters of PID control	0.2, 127 and 19 respectively
Equipment used	
Mass flow control	MKS-416 device 2 MKS 1179B MFCs (1000 sccm) for Nitrogen 1 MKS 1179B MFC (10 sccm) for Oxygen
Temperature control	HW 1440 thermo couple and controller
Gas flow equipment	2 pressure regulators 2 pressure gauges (max 15 bar) 1 pressure meter (max 2 bar)
Power switch	15 Ampere relay switch
System power supply	1 220 volt power plug 1 110 volt power plug
System power outputs	1 ground output 1 output (110 V max) for rotational motor 1 output (220 V max) for heater 1 output/input for thermocouple 1 output for RS-232 data communication
Rotational motor	Several PCBs (and a transformer) 1 power switch and indication light 1 RPM control switch
MKS-416 serial port settings	Baud rate: 9600 | Data bits: 7 | Parity: even

6. Recommendations and further research

After carrying out the project as described in this report, several recommendations will be made based upon encountered problems or difficulties as to improve the performance of the CVD process and research for future use.

Temperature distribution

The temperature distribution across the substrate is not optimal in the current situation – though better than in the old reactor. It is recommended that the temperature distribution is measured and the substrate's support is slightly altered so to make the temperature distribution more even; i.e. changes in the stainless steel support such as thickness variation determines the rate of heat flow and hence the temperature distribution across the substrate.

Homogeneity of the gas mixture

The gas mixture inside the reaction chamber is not yet optimal, as has been noticed during the test runs. Research should be done to determine how to optimize the gas mixture homogeneity. This can either be done using computational fluid dynamics numerical models (recommended) or by trying to change the flow in practice. A possible solution, for example, is using a showerhead like unit in the top of the reaction chamber to disperse the gas flows and improve their mixture and homogeneity.

Reactor tools and equipment

Currently the placement or removal of the substrate inside/from the reaction chamber is hard since the correct tools are not present. Also, when opening the reaction chamber, there is no support to leave the glass dome (top of the reaction chamber). The correct tools and support for the glass dome should be realized to prevent unnecessary accidents from happening.

Chemical process and influences

Research should be carried out to determine what factors influence the CVD process so a better control on the rate of deposition and hence the process can be obtained. Currently the chemical process occurring is not fully understood yet, but it is known that several factors influence the rate of deposition. Experiments should be carried out to determine the factors the process depends on.

Calibration of MFCs

The MFCs should be calibrated to assure their proper functioning. In the current situation the MFCs are functioning with certain inaccuracies, affecting the CVD process; see Chapter 5.2. Calibration of the MFCs would yield a better control of the process.

Mechanical friction and control of rotational motor

The mechanical friction in the rotational motor and heater equipment sometimes blocks the rotational movement and thus affecting the CVD process. Whether the rotational control or the mechanical friction is the main cause, or both, it is recommended tot sort this out and solve this.

Appendices

Appendix 1: CVD Process user guide

To start a CVD process, the following steps have to be carefully carried out. Figure 23 shows the front panel of the control box, which will be referred to several times. Before starting any process, make sure all the electrical equipment is connected correctly; see *Appendix 3: Control box connections*.

Figure 23: Front panel of the CVD control box

The following steps have to be carried out to prepare a CVD process:

1. ***Turn 'Main power switch' on the control box ON (see Figure 23).***
   - The equipment needs time to warm up and this takes at least 5 minutes.
   - Check whether all the equipment is turned on (the motor and MKS-416 can be turned OFF/ON manually with their own power switch).
   - Make sure the heater is set to the right temperature, using the '*Thermo control*' (see Figure 23), and warming up
   - If the MKS-416 produces a loud beep when turned on, the sensors are in overrange. This beep can be turned off by pressing the QUIET/CANCEL key on the MKS-416 front panel.

2. ***Make sure the heater is rotating at the right speed***
   - The rotational speed of the motor can be adjusted using the '*RPM motor*' control button on the front panel of the control box (see Figure 23).

3. ***Carefully prepare the bubblers and tube connections to the reaction chamber***
   - The equipment is disconnected and cleaned after the (series of) processes and hence needs to be connected before another series of processes is started.
   - Fill the bubblers with their chemicals and make sure all the tubes are rightly connected.

4. ***Turn the Oxygen and Nitrogen flows on***
    - Turn on the oxygen and nitrogen flows and check their pressures on the pressure gauges (see Figure 23)
    - If necessary adjust the pressures using the pressure regulators (see Figure 23)

5. ***Check the nitrogen purge flow to the airtight box***
    - Make sure a small purge stream is flowing to the airtight box, the stream can be controlled using the '*Pressure meter (nitrogen)*' (see Figure 23).

6. ***<u>Make sure the temperature is stabilized at the specified value</u>***

7. ***<u>Make sure the MFCs are stabilized before starting the CVD process!</u>***
    - If the MFCs are not warmed up enough yet, their value has not stabilized. If not stabilized, wait a few more minutes before starting the process. (O_2 is stabilized around -1 sccm; $SnCl_4$ around 0 sccm and methanol around 20 to 50 sccm)

8. ***Carefully place the substrate inside the reaction chamber***
    - Make sure the substrate's surface is clean and placed on the carbonate support to prevent it from breaking due to a thermoshock. If the surface is not clean, this will affect the final result of the deposited film.

9. ***<u>Start the process by going through the 'Deposition Control user guide'</u>***

Appendix 2: Deposition Control user guide

Here the steps of how to use the Deposition Control program to control the mass flows of the CVD process will be discussed so anyone is able to use the program. The UI is depicted in Figure 24; which will be referred to multiple times in this guide. ***Before using the program, make sure all the equipment of the CVD process is ready to use and the MFCs and the heater are warmed up (see CVD Process user guide).***

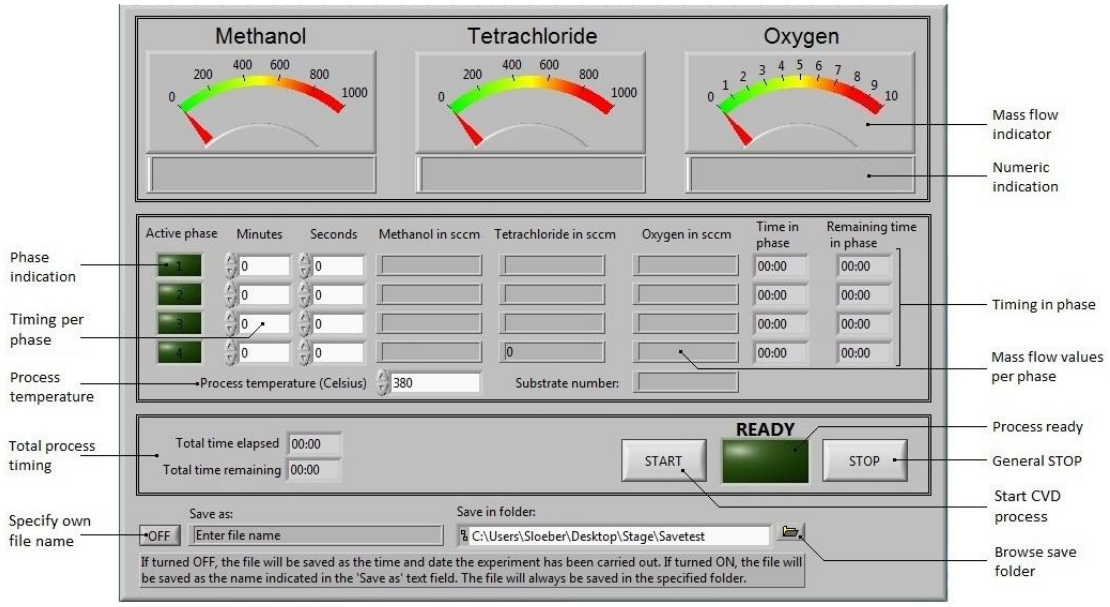


Figure 24: User Interface (UI) of Deposition Control program

The following steps have to be carried out to start a CVD process

1. ***Turn on the computer and log in***

2. ***Double click (open) the shortcut to the Deposition Control program***
   - LabVIEW 8.5 is opened and the UI as shown in Figure 24 pops up.

3. ***Press the RUN button in LabVIEW or press CNTR+R.***
   - The program starts measuring the MFC mass flows. If the mass flow values are not shown in the indicators (see Figure 24, the program is not running!)

4. ***Fill in the values for the process time in every phase***
   - The time can be entered manually or set by using the up and down buttons next to the time fields. The left field is for the minutes, the right field for the seconds.

5. ***Fill in the mass flow values of the different streams in every phase***
   - The mass flows are manually set for every phase, in sccm. Fill in 0 if you don't use a stream or if the stream needs to be zero.

6. ***Fill in the process temperature and substrate number***

7. ***Indicate if you want to save the process data in a file with a specified file name***
    - The process data is ALWAYS saved. If the button '*Specify own file name*' (see Figure 24) is turned *OFF*, the file is saved as the time and date the experiment is carried out.
    - If you want to save the process data in a file with an own specified name, click the '*Specify own file name*' so it says *ON* and turns green. Now enter you file name in the box under '*Save as:'*.

8. ***Specify the folder in which you want to save the file***
    - The box underneath '*Save in folder:*' (see Figure 24) shows the path to the folder in which the file will be saved. By clicking the 'browse folder' button next to the shown path, a destination folder can be selected in a popup menu.

9. ***To start the process click the START button***
    - To start the CVD process, press the START button on the UI. The UI will show that the process is running as long as the '*Process Ready*' indicator (see Figure 24) is still dark green. Also the time indicators are running.

10. ***Process ready***
    - The CVD process is ready when the '*Process Ready*' indicator (see Figure 24) turns from dark green to light green and the text '*READY*' is clearly visible.

11. ***Stop process***
    - If the process is ready ALWAYS click the '*General STOP*' button (see Figure 24), even if you want to carry out another CVD process.

12. ***Start new process***
    - To start a new process, repeat steps 3 to 11 if the Deposition Control program is still opened.

IMPORTANT NOTES:

1. ***After a process is finished and the substrate removed from the reaction chamber, allow the substrate to cool down by waiting a few minutes. After the substrate is removed and another substrate is placed, the process can be started again.***
2. ***Always check whether the temperature is stabilized or not, do not start the process if the temperature is not yet stabilized at the specified value.***
3. ***If the process is to be stopped during operation, press the stop button. The mass flows are automatically set to zero and the process is stopped.***
4. ***If 3 or less phases are desired, just fill in the data for the first few phases. The values of the phases left out can be set to zero or be left empty.***
5. ***The file path – the path containing the folder in which the file will be saved – can only be changed in the LabVIEW code, not on the UI.***

Appendix 3: Control box connections

The control box has several connections to other devices which will be discussed here. In order to start a CVD process, the following connections have to be checked and be right.

1. RS-232 between computer and MKS-416 device
   - To realize data communication and process control via the PC, the RS-232 cable has to be connected.
2. Power plugs of the control box
   - The control box uses two power plugs (one of 110V and one of 220V).
3. Heater power
   - The power of the heater is controlled by the control box. The heater is connected to the power socket at the back of the control box.
4. Motor power
   - The power of the rotational motor is controlled by the control box. The motor is connected to the power socket at the left side of the control box (black and red output).
5. Electrical ground of the external equipment
   - The external equipment (heater and motor) is connected to the electrical ground using the ground output at the left side of the control box (green output).
6. Thermocouple
   - The thermocouple is connected to the box at the left side of the control box (black input).
7. Oxygen and nitrogen input
   - The oxygen and nitrogen inputs are connected to the control box at the back of the control box.
8. Gas flow outputs
   - The two nitrogen streams and the oxygen stream connections are found at the back of the box.

Appendix 4: Troubleshooting

Several possible errors or problems that can be encountered while using the equipment and Deposition Control program will be treated here and are as follows; see Table 3.

Table 3: Troubleshooting equipment errors

Error	Possible cause	Corrective action
Deposition Control software does not read the mass flows	Mass flow meters are not correctly connected to MKS 416 device (see MKS 416 error E0)	Check the connections and make sure the plugs are connected correctly
	RS 232 cable is not connected correctly	Check the connection of the RS 232 cable to the PC and the MKS 416
	Wrong communication port selected in Deposition Control software	Try to change the communication port and try to run the program again
	Communication port settings of the MKS 416 and DC program do not match	Check the port setting of the MKS 416 (see page 221 of MKS 416 manual) and the settings of the DC program (see Chapter 4.3.2)
MKS 416 error E0 (shown on front panel of MKS 416 and/or on Deposition Control program)	Disconnected sensor (MFC controller)	Check the connections and make sure the plugs are connected correctly.
MKS 416 error E12 (shown on front panel of MKS 416 and/or on Deposition Control program)*	System recovery failure.	Turn the MKS 416 ON and OFF and run the Deposition program again. If this does not work, check the MKS communication port settings and make sure these values match the DC program values.
Thermocontrol *OPEN* message on HW1440 display	One or both of the thermocouple wires is not connected	Check the connection of the thermocouple to the control box
Heater does not start (temperature does not increase)	Temperature set point is not set correctly	Check the temperature set point and set again
	Heater plug is not connected to the control box	Check the connection (back of the control box)
Rotational motor does not work	Motor plugs are not connection correctly	Check the connection of the power plugs (left side of control box)
	Too much mechanical friction in the equipment	Manually try to overcome the friction and the movement will start.
Equipment does not work	Fuses are blown	Check the fuses (front panel of control box)
Power switch does not work	Power plugs are not connected	Check the connection of the 2 power plugs.

**For more error/status messages of the MKS 416, see page 307 and further of the manual [3].*

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