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# Method for Production of Uniform Thin Films from the Vapour Phase*

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*Abstract*

A general method is described for production of uniform thin films from the vapour phase. A brief review of the prior art reveals that various approaches to this problem, and in particular substrate rotation, fail to achieve uniformity of thickness and composition in such technologies as molecular beam epitaxy (MBE) or chemical vapour deposition (CVD). The new technique, for which patent protection has been obtained, involves establishment of uniform growth conditions in one direction, and translation of the substrate at a constant rate in a direction perpendicular to the first. The effect is to integrate and average any property in the direction of translation, so that every point on the substrate experiences an identical history, ensuring complete uniformity of the deposited film.

## 1. Introduction

Thin films of many materials such as metals, oxides, chalcogenides, pnictides, superconductors, etc. are of immense technological importance in the realisation of countless practical applications. A key requirement for mass production, in order to maximise yield and maintain quality standards, is the attainment of uniformity as close to ideal as possible. In addition, high-throughput, continuous (as distinct from batch) processes are economically desirable.

The invention revealed here is applicable to batch or continuous processes and to production of thin films by any technique which provides the constituents of the thin film in gaseous or aerosol form. Thus chemical vapour deposition, physical vapour deposition, sputtering and spray pyrolysis are examples of deposition techniques which will be improved by application of the invention.

## 2. Review of Prior Art

In order to describe the benefits of the invention, earlier work in the field of chemical vapour deposition will be reviewed. However, as described above, the invention has the potential for wider application.

Scientific papers and patents cited in the references describe state of the art development of reactor chambers for production of thin films. (See for example Bulsari *et al.* 1988; Chern and Maserjian 1977; Giling 1985; Jensen *et al.* 1987; Ouazzan *et al.* 1990; Park and Chun 1987; Peev and Zambar 1987; Sherman

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1988; Schumaker *et al.* 1988, 1989; Thrush *et al.* 1987; Yamawaki *et al.* 1979. Other relevant references are cited elsewhere in the text.) Improvements in thin film uniformity have been made both through empirical iterative modifications as a result of measuring properties of the films (Tanaka *et al.* 1990; Takenaka *et al.* 1988; Johnson *et al.* 1987*b*), and by mathematical modelling (Ouazzani and Rosenberger 1990; Fotiadis *et al.* 1990; Holstein *et al.* 1989; Makarov and Zhmakin 1989; Holstein and Fitzjohn 1989; Lee and Liubinas 1988). Modelling in two or three dimensions is extremely complex and is currently limited by the memory and speed of supercomputers (Ouzzani and Rosenberger 1990; Fotiadis *et al.* 1990; Jensen *et al.* 1989).

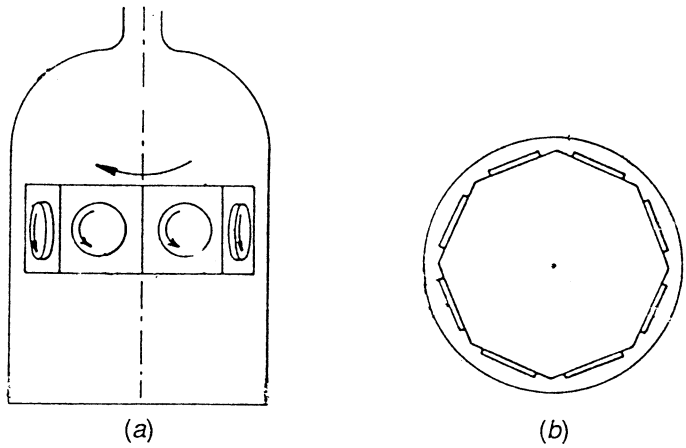


**Fig. 1.** (*a*) Side view of the vertical chemical vapour deposition reactor cell with the directions of susceptor and wafer rotation indicated. (*b*) Section through the cell showing disposition of substrate surfaces with respect to cell wall.

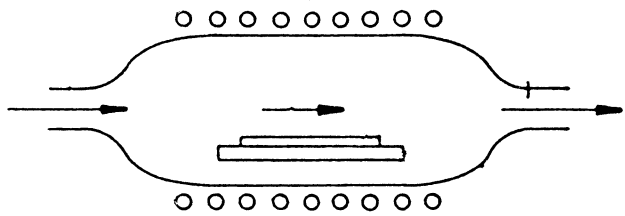

**Fig. 2.** Side view of a horizontal reactor cell with the substrate mounted on the bottom susceptor and the gas flow direction indicated.

Reactor chambers fall into two broad categories, namely vertical (also known as barrel) and horizontal cells. These basic cells are illustrated in Figs 1 and 2. In both designs a reaction gas is passed at atmospheric or reduced pressure through the cell. Both designs are used primarily as batch reactors, although the horizontal design is adaptable to continuous throughput (Akai *et al.* 1975) in production of such materials as aluminium-coated plastic sheet and architectural glass.

In the vertical configuration substrates which are to be coated are mounted on a support which is rotated about the vertical axis of symmetry to improve distribution of the flowing gas over the substrates. Tanaka *et al.* (1990) described the uniformity of wafers produced in a vertical reactor where each individual wafer was rotated additionally about an axis inclined to the vertical symmetry axis of the cell. This design was disclosed in a patent (Goto and Sekiya 1986). A fundamental limitation of this and related designs is the impossibility of providing a uniform flow of gas over the flat wafer and support with a curved bell jar, which is required if the wafer support is to be rotated. Thus, the distance between the substrate and the wall will always decrease from the centre to the edge of the wafer (Fig. 1*b*). This limitation has been discussed elsewhere (Shih *et al.* 1988; Lord 1987).

It has also been demonstrated recently by means of flow visualisation and modelling that the gas flow distribution in the vertical configuration can be asymmetric (Keijser *et al.* 1988), rather than symmetric as would be predicted on intuitive grounds. This finding reinforces the need to demonstrate that a particular design variation yields the desired results through measurements of thickness profiles of thin films.

In the horizontal reactor the substrate is disposed essentially along the lines of flow of the reaction gas and there are differences in flow velocity, temperature and reactant concentration in the direction of gas flow. Recirculation cells and longitudinal rolls can develop due to buoyancy, cell geometry and gravity so that lateral uniformity (perpendicular to the direction of gas flow) has not been achieved over the entire length of a horizontal cell.

An extension of the horizontal cell to enable continuous production of coated substrates has been disclosed (Akai *et al.* 1975). However, the method is useful only for production of films with composition profiles graded with depth due to depletion and temperature variation in the direction of the gas flow.

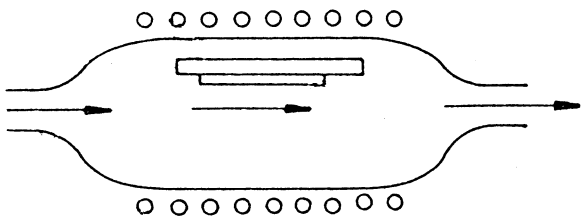

**Fig. 3.** Side view of an inverted horizontal cell with the substrate mounted on the top susceptor.

A recent development of the horizontal reactor (the 'inverted' reactor) entails mounting the substrate on the top of the cell and making this the hottest part of the cell (Fig. 3) (Puetz *et al.* 1988). This method necessitates the use of complex mounting designs, but has a number of advantages over the bottom-heated configuration. It has been shown by means of modelling (Fotiadis *et al.* 1988; Lee and Liubinas 1988) that such a cell yields stable laminar flow under a wide range of flow rates and that isotherms are essentially parallel to the direction of gas flow after stabilisation over an entrance length. Very good lateral uniformity has

been reported for this design (Puetz *et al.* 1988) and modelling results suggest that cell dimensions perpendicular to the gas flow can be increased to allow deposition over a larger area. However, this design with the heated substrate at the top still suffers from variation of growth rate in the direction of gas flow, limiting the tolerable area that could be grown in one batch.

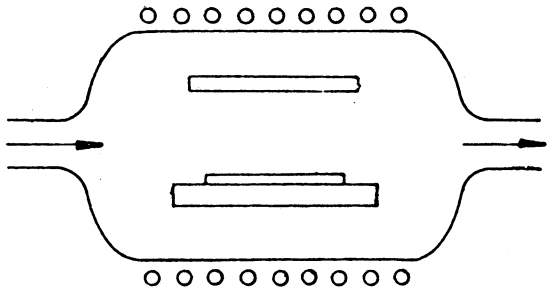

**Fig. 4.** Horizontal cell with heated susceptor at top and the substrate mounted on an independently heated support at bottom.

A novel variation (Czerniak and Robinson 1986) of the inverted horizontal reactor, designed primarily for growth of the compound semiconductor mercury cadmium telluride, has the highest temperature zone at the top, but in this case growth takes place both on the top susceptor and on the cooler parts of the cell (Fig. 4). Material in the gas phase is driven by thermal and concentration gradients to the cooler parts. Recent experiments (Pain *et al.* 1990) have shown that although this cell too suffers from growth rate variation in the direction of gas flow, lateral growth rates are very uniform on the bottom of the cell. This is to be expected due to the good lateral uniformity observed for substrates at the top of the inverted cell referred to above and also to hydrodynamic calculations.

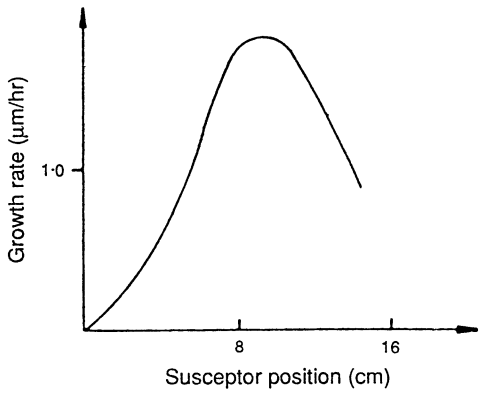


**Fig. 5.** Typical variation of growth rate in the direction of gas flow for a horizontal cell with any of the designs discussed in the text.

## 3. A Method of Obtaining Uniform Films and Why Rotation Fails

The present invention provides a means of obtaining an unlimited number of uniformly coated substrates, provided that two essential conditions can be met.

First, the lateral uniformity over the dimensions of interest must be established. As discussed above, technology exists to satisfy this criterion. The novelty of the method lies in overcoming the lack of uniformity which exists in all previously disclosed reactor cells.

Fig. 5 shows an arbitrary distribution of growth rate in the direction of gas flow. Consideration of this curve reveals that it would not be possible to average the thickness of the film by rotation of the substrate, unless a zone could be found which had a rigorously linear variation in the direction of gas flow and lateral uniformity existed. However, if the substrate is moved at a constant rate in the direction of gas flow (or counter to the flow) the amount of material deposited at each and every point of the substrate is the integral of the curve in Fig. 5 with respect to time.

The method can be extended to the case where lateral uniformity does not exist if the special condition that lateral uniformity varies linearly is met. In this case, the substrate must be rotated and translated at constant rates while traversing the deposition zone. Experiments have shown that the deposition profile is independent of the substrate position in the cell.

## 4. Annealing and Diffusion

The method described above is particularly useful for deposition of multilayer films, where each layer is required to be uniform with depth. This can be achieved either by passing the substrate in the manner described through an unlimited number of deposition zones, each depositing a unique layer, or by changing the composition of gas in a particular zone. If the gas composition is uniform throughout the zone, the film will be uniform with depth. Graded composition with depth can be achieved either by changing the gas composition uniformly with time, or by exploiting the technique of Akai *et al.* (1975) where the gas composition at a particular point in the direction of gas flow is determined by temperature and depletion.

The method is well suited for production of superlattices and films derived by solid state interdiffusion of multilayers, as described by Irvine and Mullin (1981) and Irvine *et al.* (1984). In this case it is necessary to establish not only growth rate uniformity in the lateral direction but, additionally, the temperature must be laterally uniform in order to ensure complete interdiffusion. Calculations indicate that this condition is met in the inverted horizontal reactor with the substrate either at the top or bottom of the cell.

In the method described here each point on the substrate experiences identical thermal histories provided that lateral temperature uniformity is established. Thus, the technique of translating the substrates at a constant rate will be useful for heat-treating by annealing, even in the absence of deposition. This is similar to the heating methods described by Shibamata *et al.* (1984) and Hampikian *et al.* (1970). However, those methods could not achieve lateral uniformity due to the hydrodynamics and radiative inequivalence of the reactor designs.

## 5. Conclusions

The method of deposition of uniform thin films revealed in this paper and in national and international patent applications (Pain 1990) should lead to materials with variations in composition and thickness below the detection limits

of available analytical techniques. This should have dramatic effects on device yield and reproducibility.

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