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# Thin film silicon modules: Contributions to low cost industrial production

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## Summary

The present report describes the 2 year period 2003-04 of research work at the Thin-Film Solar Cell Laboratory of the Institute of Microtechnology (IMT), University of Neuchâtel. These two years marked a transition period, from more fundamentally-oriented research work to concrete industrialization issues, and also a transition within the research team: arrival of a new professor and many changes within the research staff.

During these two years, the following main results were obtained:

- All the basic techniques for fabricating state-of-the art p-i-n type solar cells on glass were taken over by a fully new team (the whole former IMT research team working in this specific field was transferred – together with the technology – to the industrial laboratory of UNAXIS SOLAR). Hereby, continuity in fabrication know-how has been assured for amorphous, microcrystalline and micromorph tandem solar cells. Specific progress has been achieved in quantifying current and efficiency gains through light trapping by front TCO and ZnO intermediate reflector layers; also, very encouraging results were obtained by a combination of the high-pressure depletion regime with VHF plasma excitation frequencies resulting in device-quality microcrystalline layers deposited at 2.5 nm/s.
- New techniques have been introduced in the field of n-i-p solar cells on low-cost flexible substrates; specially successful were cells deposited on polyethylene tetraphtalate (PET) substrates on which periodic diffraction gratings had been embossed. Here, record efficiencies were obtained: 7% for single-junction microcrystalline cells and 8.3% stable for micromorph tandem cells.
- IMT's Zinc oxide (ZnO) layers, that are produced by low-pressure chemical vapour deposition, have been systematically studied and optimized. The relationship between crystal growth, doping and layer thickness, on one side, and layer surface roughness and light scattering capacity, on the other side, has been elucidated. The stability problem of various types of LP-CVD ZnO layers in hot and humid atmosphere has been investigated; various novel avenues with a potential for improving TCO properties have been explored.
- The key role of substrate chemistry and roughness in the nucleation and growth of microcrystalline silicon layers has been established; these factors, thus, also decisively influence microcrystalline silicon solar cell performance. New characterization/diagnostic tools for analyzing microcrystalline silicon solar cells have been set-up and tested on full series of solar cells. The light-induced degradation effect (also called Staebler-Wronski Effect, SWE) has been shown to be present in a mild form in microcrystalline silicon solar cells, contrary to earlier expectations. However, with present deposition technology, SWE is not a matter of concern for microcrystalline single-junction solar cells and is certainly not important at all for the microcrystalline bottom cell within the micromorph tandem.

## Zusammenfassung

Der vorliegende Bericht beschreibt die zweijährige Forschungsperiode 2003-04 des Laboratoriums für Dünnfilmsilizium und Solarzellentechnologie am Institut für Mikrotechnik (IMT) der Universität Neuchâtel. Diese 2 Jahre stellten eine Uebergangsperiode dar von einer mehr grundsätzlichen Forschungsarbeit hin zu konkreten Fragen der Industrialisierung, sowie auch eine markante Uebergangsperiode in der Forschungsgruppe: Ankunft eines neuen Professors und viele Aenderungen im Forschungspersonal.

Während diesen 2 Jahren wurden die folgenden Hauptresultate erzielt:

- Alle Grundtechniken für die Herstellung von hochentwickelten Solarzellen in der p-i-n Konfiguration auf Glasssubstraten wurden von einem völlig neuen Forschungsteam übernommen (die ganze frühere Forschungsgruppe des IMT, welche in diesem Sektor arbeitete, ist zum Industrielabor der UNAXIS SOLAR transferiert worden, samt dem ganzen zugehörigen technologischen Know How). Dabei musste am IMT eine Kontinuität aufrecht erhalten werden für die Herstellung von amorphen und mikrokristallinen Einzelzellen und von mikromorphen Tandemzellen. Spezifische Fortschritte wurden erzielt bei der Quantifizierung der Gewinne in Kurzschlusstrom und Wirkungsgrad durch „light trapping“: sowohl durch die Lichtstreuung am ZnO Frontkontakt, wie auch durch einen ZnO Zwischenreflektor in der Tandemzelle. Ebenso wurden sehr ermutigende Resultate erreicht durch eine Kombination vom „High Pressure Depletion“ und VHF-Plasma: intrinsische mikrokristalline Schichten mit Solarzellenqualität wurden bei 2.5 nm/s abgeschieden.
- Neue Techniken wurden eingeführt im Sektor der n-i-p Solarzellen auf billigen, flexiblen Substraten. Besonders erfolgreich war das für Solarzellen, welche auf Polyethylenteraphtalat (PET) Substraten mit einem periodischen Diffraktionsgitter abgeschieden wurden. Hier wurden Rekordwirkungsgrade erzielt. 7% für mikrokristalline Einfachzellen und 8.3% stabil für mikromorphe Tandemzellen.
- Die transparenten Kontaktschichten aus Zinkoxid (ZnO), welche am IMT durch Low Pressure Chemical Vapor Deposition (LP-CVD) abgeschieden werden, wurden auf systematische Weise untersucht und optmiert. Die Zusammenhänge zwischen Kristallwachstum, Dotierung und Schichtdicke einerseits, sowie Flächenrauheit und Fähigkeit zur Lichtstreuung anderseits, wurden abgeklärt. Die Stabilitätsprobleme verschiedener Arten von LP-CVD ZnO-Schichten wurden gegenüber Feuchtigkeit und Hitze untersucht. Verschiedene neue Wege, um bessere transparente Kontaktschichten zu erhalten, wurden sondiert.
- Die Schlüsselrolle des Substrats bei der Nukleation und Wachstum von Schichten aus mikrokristallinem Silizium wurde aufgezeigt; es geht dabei um die chemische Natur und die Oberflächenrauhigkeit des Substrats. Diese Faktoren beeinflussen wesentlich die Eigenschaften der darauf deponierten mikrokristallinen Solarzellen. Neue Methoden zur Charakterisierung und für die Diagnostik von mikrokristallinen Siliziumzellen wurden aufgebaut, und an mehreren Serien von Solarzellen getestet. Im Gegensatz zu früheren Erwartungen konnte das IMT aufzeigen, dass die lichtinduzierte Degradation (auch „Staebler-Wronski Effekt, SWE“ genannt) ebenfalls in mikrokristallinen Solarzellen vorhanden sein kann, allerdings in einer sehr „milden“ Form. Immerhin ist dieser Effekt, bei der heutigen Abscheidetechnik, kein wesentlicher Faktor für die mikrokristalline Einzelzellen, und ganz vernachlässigbar bei der mikrokristallinen „Bottomzelle“ innerhalb der mikromorphen Tandemzelle.

## Résumé

Le présent rapport décrit les résultats obtenus pendant la période de travail 2003-2004 par le Laboratoire de recherche « silicium en couches minces et technologie photovoltaique » de l’Institut de Microtechnique (IMT) de l’Université de Neuchâtel. Les résultats présentés ont été obtenu grâce au fiancement de l’OFEN (Office Fédéral de l’Energie). La période décrite a été, pour le laboratoire, une période de transition entre une phase (antérieure) de recherche plus fondamentale et la phase actuelle de transfert technologique vers l’industrie, avec tous les problèmes concrets qu’elle implique. Cette période a également représenté une phase de transition au niveau du personnel de l’équipe de recherche (arrivée d’un nouveau professeur, départ de certains chercheurs expérimentés).
Ainsi, un transfert important de la technologie de fabrication des cellules p-i-n sur verre a dû être assuré. L’ancien groupe de collaborateurs qui s’occupait de ce secteur à l’IMT a été transféré en bloc, avec tout son savoir-faire, chez Unaxis Solar. Il s’est agit pour l’IMT d’assurer la continuité du savoir-faire pour les cellules pin (amorphes, microcristallines et micromorphes) à une nouvelle équipe de jeunes chercheurs.

Des progrès scientifiques spécifiques à ce secteur ont été :

- La quantification des gains en courant et en rendement que l'on peut obtenir par piègeage de la lumière grâce aux couches de ZnO utilisées comme contact électrique transparent ainsi que comme réflecteur intermédiaire dans les tandems micromorphes.
- Le dépôt rapide de couches microcristallines : des résultats très encourageants ont été obtenus pour le dépôt rapide de couches de silicium microcristallin par la combinaison du régime « High Pressure Depletion, HPD » avec celui du plasma VHF : des couches de qualité suffisante pour être incorporées dans de futures cellules solaires ont été déposées avec un taux de 2.5 nm/s.

Les résultats scientifiques principaux qui ont été atteints dans les autres secteurs de recherche sont résumés ci-dessous :

- Introduction de substrat plastique pour la fabrication de cellules solaires de silicium en couches minces. Avec l'utilisation de substrat en polyethylènetetraphtalate (PET) pour des cellules de type n-i-p, l'IMT a emporté un succès remarquable en fabricant de bonnes cellules sur ce substrat flexible, disponible à un prix très raisonnable.
- Afin d'améliorer le rendement de conversion des cellules sur PET, le susbstrat a été nanostructuré avec des réseaux périodiques. Ceci permet de pièger la lumière par diffraction à l'intérieur des couches de silicium. Ainsi, l'IMT a obtenu les rendements de conversion records de 7% pour une cellule simple microcristalline, et de 8.3 % (stable) pour une cellule tandem micromorphe.
- Les couches transparentes conductrices en oxyde de zinc (ZnO) dévelopées à l'IMT ont été optimisées. Ces couches sont déposées par la méthode de dépôt physique en phase vapeur à basse pression (LP-CVD). Elles servent de contact électrique transparent sur les deux faces des cellules. Ces couches ont été systématiquement étudiées, et le lien entre la nano-rugosité de la surface et la croissance cristalline, le dopage et l'épaisseur de la couche a été établi. Par ailleurs, le lien entre la nano-rugosité de la surface et les propriétés diffusantes pour la lumière a aussi été établi. Ce dernier permet d'optimiser le choix de la couche de ZnO en fonction du type de contact électrique recherché.
- Les problèmes de stabilité des propriétés optiques et conductrices des couches de ZnO plongées dans une atmosphère chaude et humide ont été évalués.
- Différentes voies nouvelles pour améliorer les caractéristiques et la stabilité des couches de ZnO ont été explorées et certaines ont montré un potentiel prometteur.
- Le rôle majeur joué par le susbtrat sur la nucléation et la croissance des couches en silicium microcristallin a été dévoilé. En particulier, nous avons pu montrer que la nature chimique du susbtrat est un paramètre majeur de contrôle de la nucléation. La rugosité du susbtrat, quant à elle, joue un rôle sur la croissance des couches. Ces facteurs ont donc également une influence décisive sur les performances des cellules solaires.
- De nouvelles techniques de caractérisation et de diagnostique de la qualité du matériau et des performances des cellules solaires ont été mise en place. Leur application à des séries de cellules solaires choisies a permis d'interpréter les améliorations observées des performances des cellules solaires.
- L'effet de la dégradation induite par la lumière (appelé aussi effet Staebler-Wronski) a été observé dans certaines cellules solaires microcristallines. Une étude systématique a permis de montrer que les observations antérieures de la stabilité du silicium microcristallin étaient liées à la grande cristallinité des couches étudiées. Dans les meilleures couches actuelles, qui sont moins cristallines qu'initialement, on retrouve une forme très réduite de dégradation, qui ne devrait guère poser de problèmes pour l'application de ces couches dans des cellules simples microcristallines. Quant aux cellules tandem micromorphes, on peut estimer que la stabilité de la cellule microcristalline est certainement assurée avec les couches actuelles.

# 1. Introduction

## 1.2 Background

Thanks to funding by the Swiss Federal Government, and especially by OFEN (Office fédéral de l'énergie) in previous years, the Institute of Microtechnology (IMT) at the University of Neuchâtel was able to make significant innovations in the field of thin-film silicon solar cells. These relate to the following aspects:

- Very High-Frequency (VHF) Plasma Deposition of amorphous silicon, which enables high-rate deposition and, thus, cost reduction
- Introduction of Microcrystalline silicon, deposited by VHF, as novel photovoltaic material with reasonable cell performances (efficiencies over 5%)
- Introduction of microcrystalline/amorphous, or so called "micromorph" tandem solar cells
- Development of ZnO layers for use as transparent contact layers with light scattering capabilities, leading to record efficiencies in amorphous silicon solar cells.

These have been significant advances in the field of thin-film silicon photovoltaic solar cells. They have indeed been copied by many other laboratories – both University and Industry laboratories – in Japan, Germany, USA and several other countries.

On the other hand, IMT has now been able to start to transfer its know-how in this field to 2 industries:

- VHF-Technologies S.A., Le Locle/Yverdon (a start-up firm founded in 2000, and presently setting up a 100 kWp/year pilot production line in Yverdon for production of flexible amorphous silicon PV modules on low-cost plastic substrates)
- UNAXIS A.G. Balzers/Truebbach (a strategical business unit of Unaxis, founded in 2003, with laboratories in Truebbach and Neuchâtel. The goal of this unit is to adapt UNAXIS production equipment to production processes and solar cell structures developed at IMT).

In the course of these two technology transfer operations, each time some four key scientists/technicians left IMT to join Industry. This meant that IMT had to pass through a difficult transition period. We had indeed to give full priority to maintaining our know-how inspite of a very substantial change in the composition of our team.

A further difficulty was given by the fact that Prof. Arvind Shah, who founded the Thin-Film Silicon/Photovoltaic Research Laboratory in 1985 will be retiring in September 2005. Fortunately, in mid 2004, Prof. Christophe Ballif was nominated as his successor. Thus, the continuity of work in the field of Photovoltaics, at IMT Neuchâtel, is now finally assured. It is, thus, also possible again for IMT to engage Ph.D. students to work in this field.

IMT's photovoltaic research group can, thus, from now on, resume actively pursuing long-term innovative research issues in solar cells. This is now possible, because the continuity of our know-how has been preserved, inspite of the difficult transition period.

## 1.2 General Project Goals 03-04

The project goals and achievements (detailed in section 7 of this report) relate to four different sectors:

1. p-i-n solar cells: microcrystalline (µc-Si:H) tandem solar cells: in this sector it was possible to make partial progress and to identify and solve specific technical problems. An overall progress has not yet been achieved, but the obstacles have been clearly identified.

2. n-i-p solar cells on plastic substrates: this sector was brought up to a high level of competence and record efficiencies were obtained on PET substrates.

3. ZnO layers by Low-Pressure Chemical Vapor Deposition (LP-CVD): significant advances were made in layer characterization and in process description; many new avenues were

tried out; for reasons of confidentiality, they will not all be described hereunder. Further work is required and will be carried out, if budget and working time permits.

4. Diagnostic and characterization tools for µc-Si:H layers and cells, µc-Si:H cell stability issues: significant advances made.

# 2. p-i-n solar cells on glass substrates

## 2.1 Joint optimisation of the front contact layer (TCO) and of the microcrystalline silicon solar cell

In the past research work of our group, we had been individually optimising ZnO layers and µc-Si:H solar cells, and were looking at each one of these sub-components separately. In this phase now, we have started to look at joint optimisation of ZnO layers and µc-Si:H cells; this is important because the growth of microcrystalline silicon is influenced by the type of underlying TCO layer (e.g. surface roughness, chemical nature).
In a first study, we have determined the contributions of the long wavelength spectral region (550nm < λ < 1100nm) of the incoming AM1.5 light to the short-circuit current density $J_{SC}$ by integration over the External Quantum Efficiency curve (EQE). All these µc-Si:H test cells were deposited onto ZnO layers of varying thickness, which also means in practice varying surface roughness. The results are shown in Fig. 1. As the thickness of the ZnO films increases (at constant solar cell absorber thickness), the roughness also increases and the light-scattering capacity also increases. The light-scattering capacity here is determined by integrating the diffuse transmittance of the ZnO layer itself, in the spectral range 550-1000nm. The increase of light-scattering capacity has a direct influence on the $J_{SC}$ values integrated over the spectral range 550-1000nm (Fig. 1b). Thus, this first study confirms the excellent light-scattering properties of our LP-CVD ZnO layers, especially in the specific long-wavelength spectral region that is of interest for light-trapping in µc-Si:H solar cells.
A second study of the interaction between ZnO layers and µc-Si:H solar cells is still under way. We have in fact noticed that rougher ZnO layers lead, on one hand, to higher $J_{SC}$ values, but on the other hand to a reduction of the fill-factor FF of devices. The microscopic nature of the interface ZnO – microcrystalline silicon cell must now be investigated in order to identify this drop in FF and to possibly find remedies for this situation.

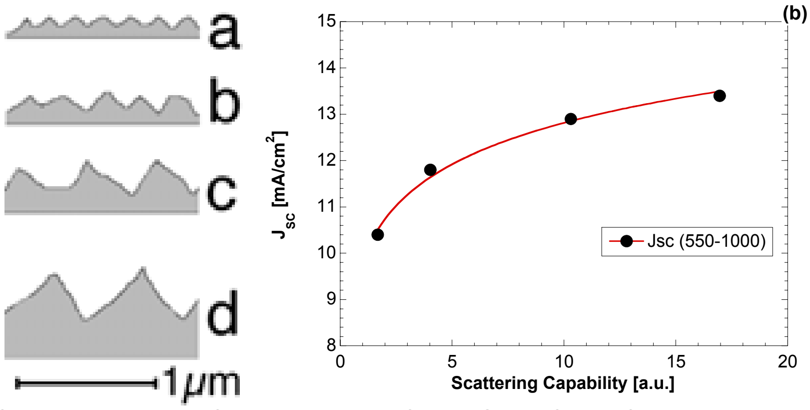



Fig. 1a: Sketch of the increasing surface roughness of Zinc Oxide films of increasing thickness. In the study presented here, the thickness of the ZnO films deposited by LP-CVD was varied from 0.8µm up to 2.5µm.
Fig. 1b: Correlation between optical scattering capability of the front contact ZnO layers and the $J_{SC}$ value (within the spectral range of 550-1000nm) of identical µc–Si:H solar cells which were deposited onto four different types of front contacts.

## 2.2 State of the art amorphous solar cells

We have changed the doping gas for p-type doping from diborane (which is chemically unstable over long time periods of storage) to Trimethyl Boron. Thereby, the critical p-layer had to be re-optimised. We are now again able to produce amorphous silicon p-i-n solar cells with over 10% initial efficiency. However, in the stabilised state after light-induced degradation our solar cells are not as good as before. We can explain this by not fully optimised intrinsic layers w.r.t. light-soaking and we exclude a relation of this situation to the newly introduced doping gas. We have also noticed a part of non-reversible degradation that is certainly due to a slight degradation of the electrical contacts (from the TCO to the measurement probe). Thus, the conversion efficiency obtained for our single-junction amorphous silicon solar cell is not yet as high as promised (7.4% instead of 8.2%) and the reasons for this will have too be seriously investigated in the coming year.

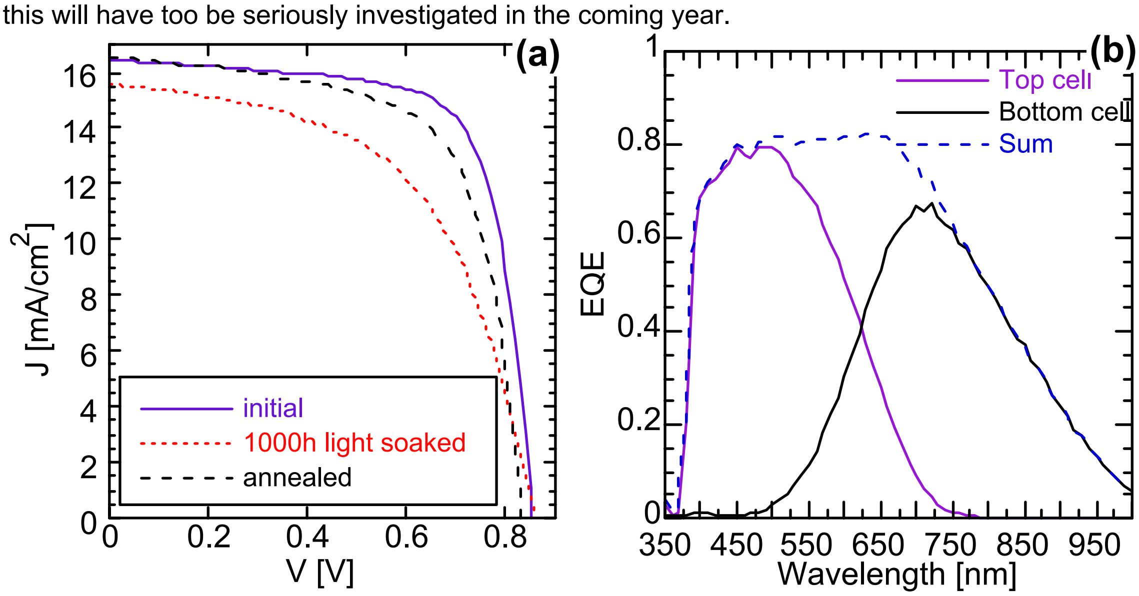



Fig. 2a: The I-V curve of recent amorphous solar cell: Initial state $V_{OC}$=854mV, FF=72%, $J_{SC}$=16.4mA/cm$^2$ $\eta$=10.1% / light soaked state: $V_{OC}$ 885mV, FF=56%, $J_{SC}$=15.5mA/cm$^2$, $\eta$=7.4%
Fig. 2b: The Quantum efficiency of the micromorph solar cell with an intermediate reflector and the well-balanced $J_{SC}$=11mA/cm$^2$ value of the two individual cells ($V_{OC}$=1.2V, FF=61%, $\eta$=8%)

## 2.3 Micromorph solar cells

During the project period our team has studied further the use of a ZnO intermediate reflector (ZiR) between the amorphous top-cell and the microcrystalline bottom-cell in order to further increase stability towards light-induced degradation within micromorph tandem cells. Current matching of the two contributing solar cells is thereby an important topic: Fig. 2b shows the well-balanced EQE curves of a micromorph tandem cell containing a ZiR. Optical modeling of devices with/without ZiR was carried with a numerical Monte-Carlo simulation programme[1]. However, so far there is not sufficient correspondence between our experimentally encouraging results and the simulation results. This inaccuracy is understandable, because the simulation model is based on flat interface layers. In our case of highly surface-textured interfaces (ZnO / a-Si:H / ZiR / µc-Si:H) the experimental device does not correspond at all to the simulation assumptions. Precise simulation becomes a difficult task because the geometrical feature-size of the surface-roughness is of the same order of magnitude as the wavelength of light involved. This means, however, that for the moment the only viable way of optimising the micromorph tandem cell with a ZiR is to experimentally vary the involved parameters, within the complex device structure.

## 2.3 High-rate deposition of microcrystalline silicon absorber layers

In order to deposit the µc-Si:H absorber layer of solar cells at even higher rates, we have been studying the high-pressure depletion (HPD) regime in combination with Very High Frequency (VHF) plasma excitation. Latest results show remarkably high deposition rates for µc-Si:H films on glass: around 2.5 nm/sec. The good news are the moderately low total gas flows of only 100 sccm (SiH4 + H2) to achieve such high deposition rates; this fact is of economical interest. These films basically possess device quality properties, such as midgap character, low sub band gap absorption and a

[1] The source code of this simulation programme is edited and maintained by the Institute of physics in Prague.

highly crystalline structure (based on FTPS and Raman Spectroscopy measurements, respectively). First promising µc-Si:H solar cells under these high pressure conditions could be fabricated.
But, no significant increase of the device efficiencies well above 5% could be obtained. That is why we reverted back to our "standard" deposition conditions (moderate working pressure below 1 mbar and deposition rates below 0.5 nm/sec) and systematically looked for the reason for the limitations in cell efficiencies.
This took up a lot of working time and we also needed some hardware changes on the deposition reactor to arrive at our first reasonable cell efficiencies in a dedicated high-rate deposition system. The process conditions now found under low-pressure conditions yield very promising solar cell characteristics: $\eta$=6.5%, $V_{OC}$=516mV, FF=69%, $J_{SC}$=18.5mA/cm$^2$. More time than expected was necessary to obtain these results; unfortunately this time is now missing on the side of high-rate device optimization, since the PhD student involved is about to finish his thesis.

# 3. nip-type microcrystalline (µc-Si:H) silicon solar cells

Thin film silicon solar cells with the n-i-p configuration allow one to use cheap flexible substrates such as Poly Ethylene Terephtalate (PET). At IMT, amorphous silicon (a-Si:H) solar cells on plastic substrates were investigated directly in collaboration with industry, in a project funded by the Federal Commission for Technology and Innovation (CTI). In the present project, funded by OFEN, we concentrated on two issues:

(1) Optimisation of light-trapping into µc–Si:H and micromorph tandem cells using also glass and stainless steel substrates, in order to simplify the laboratory fabrication of an efficient randomly textured reflector (Ag layer evaporated at different temperatures).

(2) Realisation of µc-Si:H and micromorph solar cells on low-cost substrates like PolyEthylene Terephtalate (PET).

Significant progress was achieved in this sector during the reporting period.

### 3.1. Upgrade of the deposition reactor

During the reporting period, the former **single**-chamber deposition system for µc-Si:H n-i-p solar cell deposition received an upgrade to become a completely new **double**-chamber system. This bears several advantages, such as separate deposition of doped and un-doped (intrinsic) silicon films, a higher throughput and a better reproducibility of the devices.

As a consequence of this major hardware upgrade, all deposition parameters had to be investigated again. The well-known tendencies were confirmed again: $V_{oc}$ increases steadily as a function of the Silane Concentration (SC), the Fill-Factor (FF) values remain stable up to the transition (at 6% SC) and the short-circuit current density ($J_{SC}$) decreases steadily.

### 3.2. Surface-textured and reflecting substrates for n-i-p solar cell

During the reporting period of two years, many different categories of surface-textured and reflecting substrates have been tested in order to enhance the light trapping into microcrystalline-based solar cells.
On one hand, **randomly-textured** reflectors were fabricated in the configuration stainless steel/Cr/as-grown textured Ag/sputtered ZnO or Glass/Cr/as-grown textured Ag /sputtered ZnO. In both cases, the surface textures result from the hot silver deposition.
On the other hand, a periodic back-reflector design was used: it consisted of **optical diffraction gratings** realized by a photolithographic process in our laboratory as well as by embossing in an industrial facility. For the fabrication of diffracting and reflecting substrates, the photolithographic or embossing process is followed by the deposition at low temperature of thin sputtered Ag and ZnO layers. Periodic textures, with different dimensions and shapes, were fabricated on glass as well as on PET and incorporated into cells.

## 3.3. Single junction µc-Si:H solar cells

n-i-p µc-Si:H solar cells have been developed on different textured substrates including periodically textured PET substrates. For all cells, as-grown textured LPCVD-ZnO was used as top contact.

### Randomly textured substrates

Here, an Ag layer deposited at different temperatures was used to provide a randomly textured back reflector deposited on a stainless steel substrate. The short-circuit current density (Jsc) of nip µc-Si:H solar cells was studied as a function of the typical dimensions of the random texture of the Ag layer surface. The texture was varied by increasing the temperature of the substrate for the deposition of the Ag layer. The best Ag back reflector was obtained for a substrate temperature of 380°C. For the cells deposited on this back reflector a gain in current density of approximately 20% was observed, as compared with cells implemented on an Ag back reflector deposited at room temperature.

µc-Si:H cells have been fabricated on this kind of substrate (i.e. glass/as-grown textured Ag/sputtered ZnO): up to 9.2% initial efficiency was achieved ($V_{oc}$ = 0.52 V, FF = 73 %, $J_{sc}$ = 24.2 mA/cm$^2$).

### Optical diffraction gratings

**For the first time worldwide, µc-Si:H solar cells were deposited on periodically textured PET substrates yielding an initial conversion efficiency of $\eta$=7%**, with $V_{oc}$ = 0.51 V, FF = 68 %, $J_{sc}$ = 20.4 mA/cm$^2$ (Fig. 3).

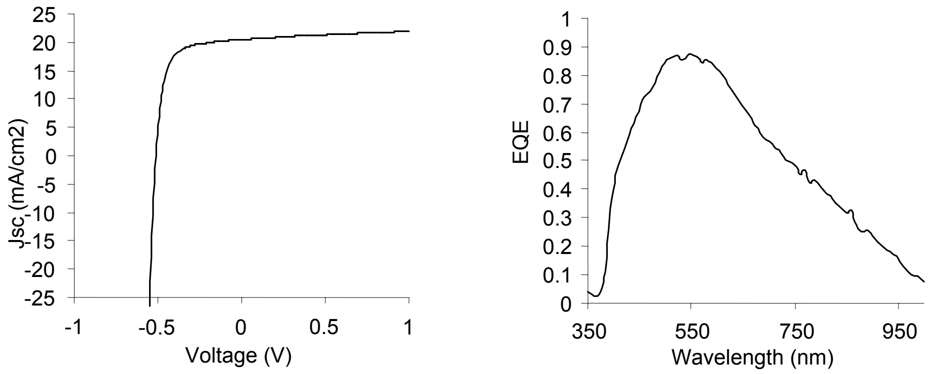



Fig. 3: IV and external quantum efficiency (EQE) curves of the best microcrystalline n-i-p silicon solar cell deposited on low-cost textured PET substrates.

## 3.4. µc-Si:H/a-Si:H tandem cells

n-i-p tandem solar cells have been extensively studied on glass. The best performances have been obtained using as-grown textured hot silver as scattering back reflector. The best initial conversion efficiency obtained so far is $\eta$=10.3%, with $V_{OC}$=1.4V, FF=66%, $J_{SC}$=11.1mA/cm$^2$ (Fig. 4).

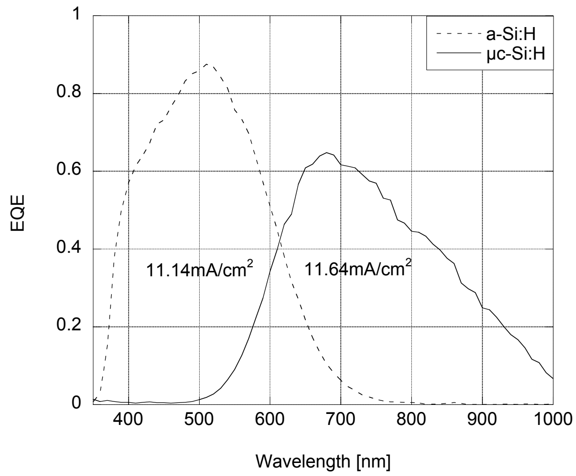



Fig. 4: External Quantum Efficiency (EQE) of micromorph solar cell on glass/Cr/as grown textured Ag.

### Optical diffraction gratings

**The worldwide first flexible micromorph solar cell was deposited on PET with a stable efficiency as high as 8.3%** with $V_{OC}$=1.33, FF=66.2%, $J_{SC}$=9.4mA/cm$^2$ (Fig.5). Thereby, a periodically textured back reflector was used for light trapping.

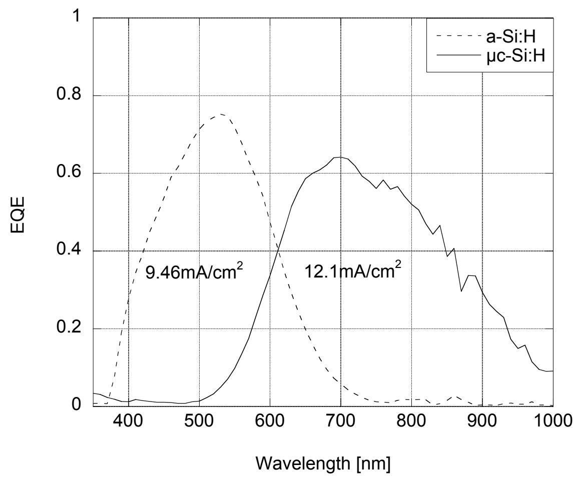



Fig. 5: External quantum efficiency (EQE) of the best micromorph solar cell on PET with Cr/Ag/sputtered ZnO back reflector.

# 4. Transparent conducting oxides

Transparent Conductive Oxides (TCO) are an essential part of thin-film solar cells, both cost-wise and performance-wise. Amongst these TCO's, doped Zinc Oxide (ZnO) is a very promising candidate for future thin-film silicon solar cell technology, especially because of its low cost and because of the wide availability of its constituent raw materials.
Doped ZnO layers deposited by the Low Pressure Chemical Vapour Deposition (LP-CVD) technique have been studied at IMT for their use as transparent contact layers for thin-film silicon solar cells. The LP-CVD technique allows us to obtain ZnO films that are not only transparent and electrically

conductive, but also possess a pronounced light-scattering capability. The light-scattering capability increases the path of the light within the solar cell, and, hence, enhances also its probability to be absorbed in the cell. This aspect is especially important in the case of amorphous and microcrystalline silicon (a-Si:H and µc-Si:H) thin-film solar cells, because of their relatively low optical absorption coefficient in the red and near infra-red (NIR) spectral range. As a valuable proof of concept, such a rough ZnO layer as front TCO has been integrated in an amorphous p-i-n solar cell yielding a stabilized conversion efficiency of η=9.5%, confirmed by NREL [10].
The light-scattering property, which is quantified by the Haze factor, i.e. the ratio of diffuse optical transmittance to total optical transmittance, is linked to the surface roughness of the TCO layer used in the cell. Indeed, a rough surface allows one to scatter efficiently the light that enters into the solar cells through the TCO layer.

### 4.1 The growth of LP-CVD ZnO

Research at IMT in the ZnO field has been mainly dedicated to a comprehensive analysis of our LP-CVD ZnO layers. Deposition parameters (i.e. gas ratios, doping level, substrate temperature, process pressure) have been varied over a large range of values. ZnO layers have been characterized in terms of optical properties (transmission and light scattering capacity), electrical properties (resistivity, free carrier concentration and mobility), and structural properties (grain size, crystallographic orientation). Correlations have been established between the optical and electrical properties, on one hand, and the growth and structural properties on the other hand.
The TEM micrograph of a side view of a LP-CVD ZnO layer is shown in Fig.6. This micrograph shows that the ZnO layers are constituted of large monocrystals that grow from almost the bottom of the ZnO layer and appear at the surface as pyramidal grains, which can be seen on the SEM micrograph of Fig.7(a). These monocrystals are all oriented along the same crystallographic axis.

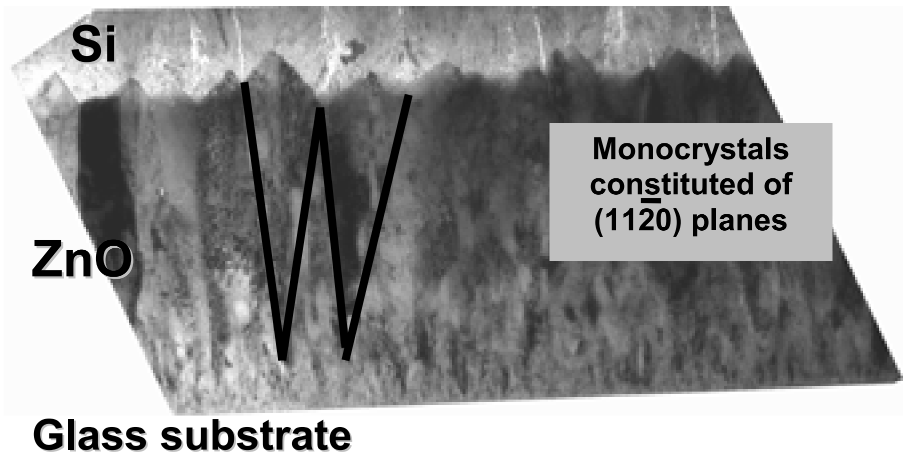
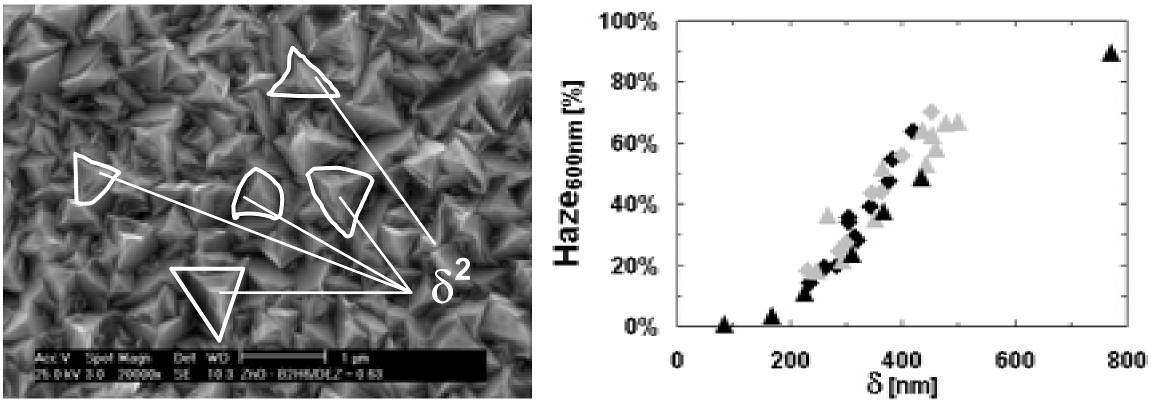



Fig.6: TEM micrograph of a ZnO layer deposited by LP-CVD on a glass substrate. The LP-CVD ZnO layer is constituted of large monocristals growing almost from the glass substrate, and appearing at the layer surface as pyramidal grains, which give to the ZnO layer its roughness.

The mean projected area of the pyramidal grains present at the ZnO surface has been evaluated from SEM micrographs, and the square root of this value ($\delta$) has been taken as dimensional parameter to characterize the microstructure of LP-CVD ZnO layers. The haze factor of several ZnO samples deposited under various conditions is plotted in function of this dimensional parameter, in Fig.7(b).

This graph clearly establishes that the larger the pyramidal grains at the surface of the LP-CVD ZnO layers, the stronger the light scattering capacity of these layers.




**(a)** **(b)**

Fig. 7: **(a)** SEM micrograph of the surface of LP-CVD ZnO layer. The mean projected area of the pyramidal grains observed at this surface has been evaluated from SEM micrographs, and the square root of this value (δ) has been taken as dimensional parameter for the microstructure of LP-CVD ZnO layers. **(b)** Haze factor measured at 600nm (i.e. light scattering capacity of the ZnO layers) in function of the dimensional parameter (δ) of the microstructure of these layers.

## 4.2 The LP-CVD ZnO layer integrated into microcrystalline p-i-n solar cells

A part of the research at IMT in the ZnO field has also been dedicated to the specific optimization of these ZnO layers with rough surfaces when they act as efficient light scatterers within p-i-n microcrystalline solar cells. Surface roughness has been tuned over a large range of values, by varying thickness and/or doping concentration of the ZnO layers (see Fig.8 which illustrates the roughness variation of ZnO layers due to a variation in the doping level). A series has also been implemented, for which the doping level has been varied while keeping the square resistance of all the ZnO layers equal to 10 $\Omega_{sq}$ (thereby, the thickness of the ZnO layer has been adjusted, in order to compensate the high resistivity of poorly doped ZnO layers). The constant resistivity allows one to maintain the series resistance of the whole solar cell device, deposited on this front TCO, in order to observe only the "optical" influence of the front ZnO roughness on the solar cell performance.

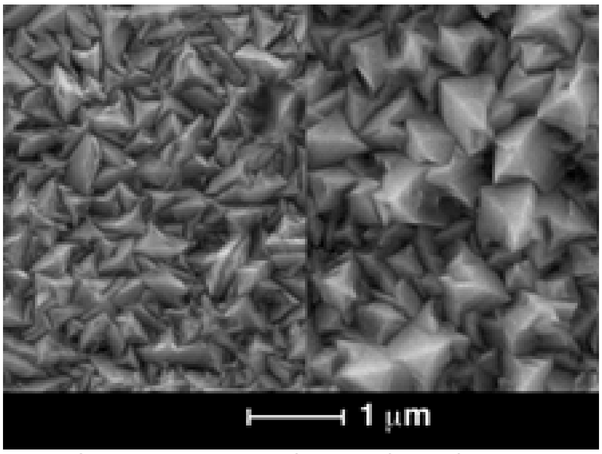



Fig.8: SEM micrographs of the surface of a (a) highly doped ZnO layer, and a (b) non-doped ZnO layer. Both layers have the same thickness. The variation of the surface roughness (and therefore of the light scattering capability) of the ZnO layer is due to the doping level: The more the layer is doped, the less it scatters the light. But a minimum doping level is still necessary to assure satisfactory electrical conductivity of the TCO.

Identical p-i-n microcrystalline solar cells have then been deposited on all these front ZnO substrates developed at IMT. In all cases, a clear increase of the photo-generated current has been observed in the red and NIR spectral range for rougher ZnO substrates. Increasing three times the grain size of the front ZnO leads to an increase of ~ 20% of the photo-generated current in the microcrystalline p-i-n solar cell, and this without any back reflector as yet. However, if the ZnO layer becomes too rough, this has so far resulted in a negative impact on the $V_{oc}$ and FF values of the solar cell. To avoid this drawback, the solar cell deposition on these very rough ZnO substrates has to be newly optimized.

All these results highlight the major advantage of the LPCVD ZnO layers developed at IMT, where the surface roughness of the substrate can be “tuned” over a wide range of values: this will allow us to optimize the whole solar cell structure for the highest photo-generated current without negative impact on the electrical behaviour of the solar cell.

### 4.3 Stability of LP-CVD ZnO

In parallel to the development of transparent, conductive and rough LP-CVD ZnO layers for thin-film silicon solar cells, several tests have been done in order to study the behaviour of LP-CVD ZnO in various atmospheres. These experiments give essential information about the limits of stability and the precautions needed during the fabrication process as well as during the use of the solar cells.

First tests consisted in observing the evolution of optical and electrical properties of the LP-CVD ZnO layers stored in the air and in a dessicator. Results show that ZnO behaves differently in the air if it is doped or not.
Second tests were done in the same way in an atmosphere with variable humidity level.

The main conclusions of this study are that LP-CVD ZnO is sensitive to oxygen and the water present in the air, and that ZnO degradation strongly depends on temperature.

On the basis of these results, a EUREKA project has been started on 1.11.2004. This project will investigate the measures (protective encapsulation material for PV modules or modification of ZnO deposition) that can be undertaken to avoid/reduce the degradation of LP-CVD ZnO.

## 5. Material characterization and device physics studies.

### 5.1 Nucleation and growth of microcrystalline silicon layers

As was already known from previous studies of our own group, and has subsequently been confirmed by many other groups, μc-Si :H is not a straightforward homogeneous material, but rather a complex mixture of nanocrystalline phase, amorphous phase, grain boundaries and voids. It is now being increasingly clear that the microstructure of the material incorporated in μc-Si :H solar cells is a very important factor that influences cell performances and stability. Especially with a view of applying in future new high-rate deposition techniques and with the hope of further optimizing cell efficiency, it is indeed useful to gain more understanding about the growth of μc-Si :H layers on different substrates.

Thereby, we have used transmission electron microscopy (TEM) as an imaging tool to look at the microstructure of layers deposited on different substrates (different susbstrate roughness, different chemical nature). Our observations indicate that :

- **Nucleation** of μc-Si :H depends mainly on the chemical nature of the underlying layer
- **Growth** of μc-Si :H is influenced by the substrate roughness.

As the crystalline domain grow perpendicular to the local substrate plane, they collide over each susbtrate’s groove. This results in the accumulation of amorphous material at the bottom of the grooves and in a grain boundary (and even cracks or voids) over each substrate’s groove. It is not clear yet how this effect affects the electrical properties of the resulting solar cell.
We have in this context also been able to introduce a simple numerical model that reproduces most of the features of the complex microstructure of μc-Si :H. A typical microstructure is presented in Fig. 9.a. The model developed is a three dimensional discrete model involving a cubic lattice where each grey level in Fig. 9.b correspond to the crystallographic orientation of a growth unit ( a particle representing the statistical behavior of a large number of adatoms involved in the growth process). An amorphous domain is, thus, filled with particles of random grey levels, whereas a crystalline domain is filled with

particles in the same crystallographic orientation. The result of these simulations ils a discrete representation of the microstructure of the layer that compares directly with TEM micrographs. This model is the first one that renders realistically the microstructure of μc-Si :H and the evolution of layer roughness with accumulated layer thickness.

(a)

(b)

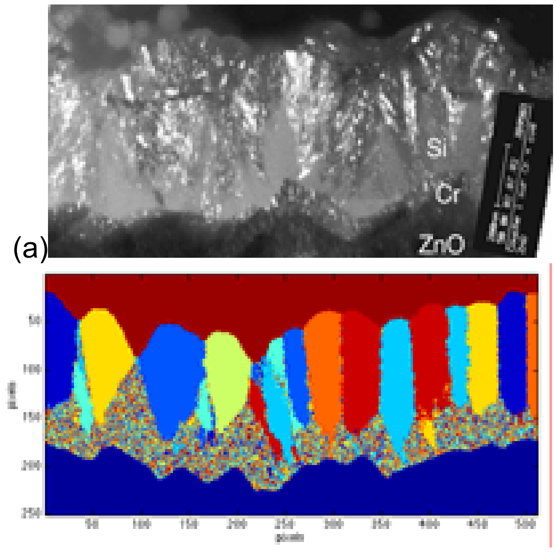



Fig. 9 : μc-Si :H layer grown on a rough substrate : (a) TEM micrograph ; (b) Simulated layer.

## 5.2 Tools for material and cell analysis

**Material analysis**: Sofar, the most useful techniques for the study of μc-Si :H layers were micro-Raman spectroscopy which gives a reliable and non-destructive indication for layer crystallinity as well as the Constant Photocurrent Method (CPM), which enables us to measure sub-bandgap absorption, and, thus, to obtain a relative evaluation of the quality of intrinsic (i-) layers. CPM is, however, a relatively cumbersome method. We have therefore introduced in 2004 a new tool that gives us the same information as CPM but in a much faster way : This is the method of Fourier Transform Photoconductive Spectroscopy (FTPS) that was recently invented by the Prague group. This method is based on a commercial Fourier transform infrared spectrometer in which the layer to be characterized is configured as an external photodetector. The FTPS method allows for a rapid, non-destructive evaluation of the i-layer quality within a full solar cell: It allows for the relative evaluation of defect (recombination center) density through the evaluation of the absorption coefficient value at 0.8 eV photon energy and also for the evaluation of material « disorder » (bandtail states shown as Urbach slope i.e. as exponential decrease at the absorption edge). FTPS measurements have already been performed on several series of μc-Si :H solar cells and the results have been proven to be very useful.

**Cell analysis** : µc-Si :H solar cells often suffer from low values of Fill-Factor (FF) (50% to 60%) and it is important to be able to identify the causes of such « substandard » cells. Low values of FF can be, in general, traced to the following causes : low i-layer quality (this shows-up in the FTPS measurement), contamination of i-layer through boron (this shows up in spectral response measurements), shunts due to cracks in the µc-Si :H layer, due to particles (dust) or finally due to cell structuring problems. It is not very easy to detect shunts, so we investigated a new method (Variable Illumination Measurement, VIM) in this context. Indeed, the measurement of the I(V) characteristic under variable illumination is a powerful tool to characterize shunts and also to identify collection problems. An automated VIM set-up has been implemented at our laboratory and can now be used for cell analysis.

## 5.3 Optimisation of Voc in µc-Si :H solar cells

The open circuit voltage $V_{oc}$ is a key parameterfor increasing the efficiency of µc-Si :H solar cells. It is well known that the $V_{oc}$ increases as one deposits the i-layer with values of the silane concentration close to the µc-Si :H/a-Si :H transition. The fundamental reason for this behavior was not known sofar. Recently, we have shown that the value of $V_{oc}$ increases linearly with the average amorphous volume fraction at the p-i and n-i interfaces. The Fourier Transform Photoconductive Spectroscopy (FTPS) measurement technique newly installed in our laboratory has allowed us since to gain further information on the role of the amorphous phase in increasing the value of $V_{oc}$. Indeed, thanks to this measurement technique, we have been able to show that density of deep defects, that act as recombination centers, decreases with increasing amorphous volume fraction (see Fig.10) It seems, thus, that the amorphous material plays the fundamental role of passivating defects in µc-Si :H.

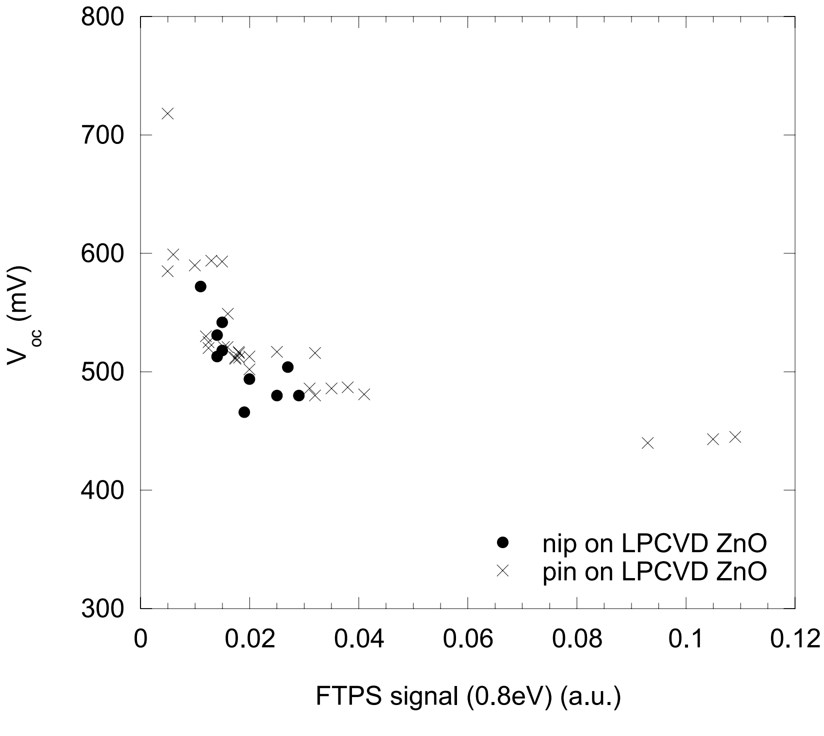



Fig. 10 : Values of the open-circuit voltage $V_{oc}$ of pin-type and nip-type solar cells as a function of defect-related absorbance at 0.8eV, measured by FTPS.

## 5.4 Stability of µc-Si :H cells under light exposure

Contrary to earlier conclusions, not all µc-Si :H solar cells are stable when exposed to full sunlight. FTPS has been used as well to study defect generation by light-induced exposition of nip-type and pin-type μc-Si :H solar cells. These studies have revealed that μc-Si :H cells with a high crystalline volume fraction are quite stable with respect to light-induced degradation ; but these cells do not possess high enough open-circuit voltage for optimum conversion efficiency. On the other hand, μc-Si :H cells with optimum conversion efficiencies show a mild light-induced degradation effect, that has been related by FTPS measurements to an increased defect density (see Fig.11) . Finally, μc-Si :H solar cells with a high amorphous volume fraction (more than 70%) degrade more than 10%, a behavior similar to that of un-optimised fully amorphous solar cells.

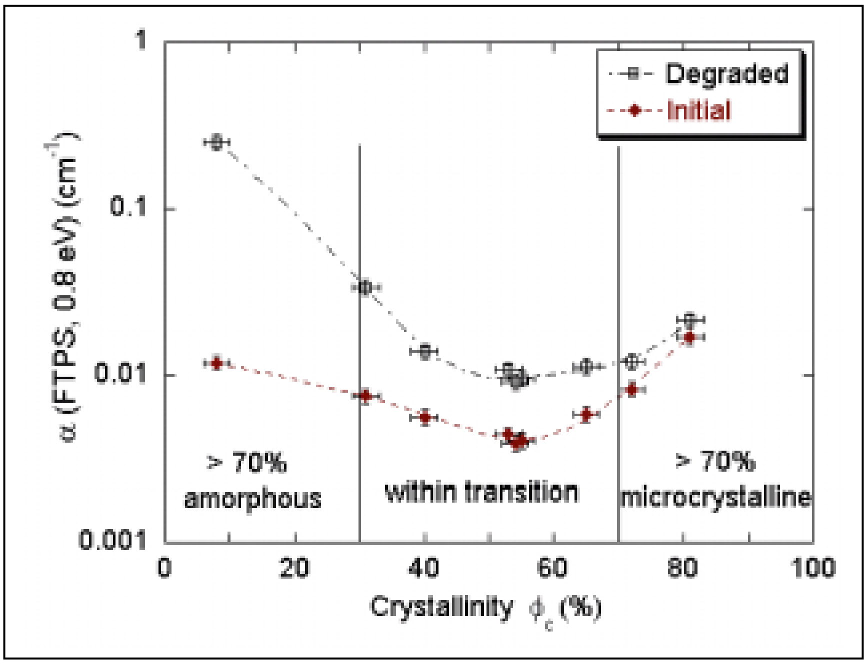



Fig.11 : Defect density before and after light-induced degradation as measured from the absorption coefficient at 0.8 eV with the FTPS technique ($\alpha_{FTPS0..8\ eV}$). The best solar cells obtained within the transition remain the best cells after light-induced degradation, even if their defect-related absorption increases by a factor of 3 to 5.

When μc-Si :H cells are used as bottom cells in the micromorph tandem, they are not subject to the full impact of incoming light, as only the red component of light is transmitted to the microcrystalline cell, the high-energy component of the spectra being absorbed by the amorphous top cell. Thus, the microcrystalline bottom cell remains indeed quite stable in this configuration. However, our recent results do mean that the stability of μc-Si :H layers and cells needs to be carefully checked in future, whenever new deposition methods /parameters are used.

# 6. Conclusions and Outlook

IMT obtained its major innovations in an early period of research work on microcrystalline silicon and micromorph tandem cells. At that moment it was clearly world-wide the leading laboratory that introduced and successfully proved the validity of many new concepts.

Since then, these concepts have been taken up by many other laboratories that are now pursuing the same goals as IMT with a much more powerful equipment base. It is not surprising that these other laboratories have caught up and, in some cases, even overtaken IMT.

One should definitely not forget that IMT is doing the work on self-built reactors and that we have to invest a lot of time and efforts in making our technological processes reliable and reproducible, whereas most other laboratories have large equipment credits that allow them to purchase commercial multi-chamber reactors. These are ready-to-use systems that are installed and serviced by professional companies. This difference in the mode of operations is not a serious handicap for the pioneering phase, but becomes very important for the phases of consolidation and industrialization.

The consequences for IMT are the following:

- Remain on the forefront of innovative ideas with at least a part of the team
- Invest another substantial part of IMT’s future work for consolidating further the technological equipment base and standardizing fabrication procedures
- Try to obtain in the medium term substantial equipment credits that allow IMT to selectively renew and strengthen its equipment base
- Intensify close collaboration with Industry (UNAXIS, VHF-Technologies)
- Intensify collaboration at the European level with other laboratories working in the same field.

The latter point will probably become possible from 2005/2006 onwards thanks to two European (FPG) projects that have been submitted and where IMT is strongly involved. It is very likely that at least one of these European projects will be approved for financing by the EU. This would definitely give a big boost to future research work at IMT.

# 7. Project goals and achieved results for the period 03-04

| Goals 03-04 | Achievements |
|---|---|
| **p-i-n solar cells on glass substrates** | |
| Optical optimisation for enhancing $J_{SC}$: | Fully achieved.<br>a) single-junction µc-Si:H p-i-n cells: an increase of the $J_{SC}$ value from 10mA/cm$^2$ for flat front TCO up to a $J_{SC}$ value of 13mA/cm$^2$ for highly-textured front TCO is measured within the spectral range of interest (550-1000nm).<br>b) Micromorph tandem with intermediate reflector: a well balanced $J_{SC}$ value of 11mA/cm$^2$ is achieved.<br>Known issues: low values of $V_{OC}$ and FF for the micromorph device. |
| Introduction of new doping gas into the process of µc-Si:H and a-Si:H cell manufacturing: a-Si:H cell efficiency of 10.1% / **8.2**% (initial / light-soaked) | Fully achieved on operational level.<br>Not fully achieved re. results: conversion-efficiency of the light-soaked amorphous solar cell reaches only values of 10.1% / **7.4**% initial/degraded, respectively. |
| High rate deposition of µc-Si:H absorber layers: combination of $\eta$>5% AND deposition rate > 10Å/sec. | Partly achieved:<br>State of the art solar cells deposited in the dedicated high-rate reactor) reach now a conversion efficiency $\eta$=6.5% but still at low deposition rates (4 Å/sec). |

| Microcrystalline silicon n-i-p solar cell on glass substrate | |
|---|---|
| Optimisation and consolidation of process windows for n- and i-layers of microcrystalline solar cells | Fully achieved:<br>Development of n-, i- and p- layers in a new double chamber system led to solar grade quality layers that were incorporated into the solar cells |
| Study of the back reflector shape | Fully achieved:<br>Glass/Cr/as-grown textured Ag/sputtered ZnO was found to be the best back reflector, leading to current density as high as 24mA/cm$^2$ [14] [28] |
| **Micromorph n-i-p solar cell on glass substrate** | |
| n-i-p type "micromorph" tandem solar cells on glass substrates at 10% stabilized efficiency | Not achieved:<br>Best initial efficiency of 10.3% on glass/Cr/as-grown textured Ag//sputtered ZnO substrate. ($V_{OC}$=1.4V, FF=66%, $J_{SC}$=11.1mA/cm$^2$) |
| **Microcrystalline silicon n-i-p solar cell on flexible substrate** | |
| Reproducible deposition of single junction µc-Si:H nip cells on glass/stainless steel and PET:<br>7% efficiency on PET | Fully achieved:<br>Microcrystalline solar cells were optimised on glass/as-grown textured Ag/sputtered ZnO.<br>The cell was then optimized on a periodically textured PET substrate covered with Cr/Ag/sputtered ZnO. On that substrate, 7% efficient solar cells were obtained. |

| Micromoph n-i-p solar cell on flexible substrate | |
|---|---|
| 8% efficiency on PET or another polymer | Fully achieved:<br>8.3% stable efficiency (after 1000h of light soaking) on periodically textured PET substrate covered with Cr/Ag/sputtered ZnO. |

| Goals 03-04 | Achievements |
|---|---|
| **Transparent conducting oxides** | |
| • Enhanced chemical stability of LP-CVD ZnO | Work started:<br>A pre-study has established a strong electrical vulnerability of the ZnO layers in hot and humid environments. Based on this pre-study, a EUREKA project has been started on 1st of November 2004. |
| • ZnO doping by e.g. F or Al<br>• Preparation of plasma-assisted decomposition of alternative doping gases, such as $CF_4$ for F-doping to improve layer quality. | Almost fully achieved for F-doping:<br>Transparent and conducting ZnO layers doped with Fluorine have been obtained. However, the quality of the layers was not improved compared to the ZnO layers doped with Boron. |
| • Improved vapor distribution system for ZnO reactors.<br>• Particle (flakes) reduction.<br>• Reactor and process full optimization (only small reactor, $8x8cm^2$). | Fully achieved:<br>Cooling systems have been installed on both reactors, leading to a strong reduction of flakes (which could induce shunts in the solar cells), and also to a reduction of the cleaning time. |
| • Systematic layer characterization of ZnO layer series: completion of characterization, Hall mobility, photo- and dark conductivities. | Fully achieved:<br>See [5] (thesis) and [24][32]. |
| • Preparation of PE-CVD reactor for ZnO | Not started:<br>Priority has been given to the plasma-assisted pre-decomposition of $CF_4$ for F-doping. |
| • Preliminary experimental study of plasma cleaning for ZnO reactors. | Not started:<br>Priority has been given to solutions for reducing the time required for the mechanical cleaning-cycle (cooling system, new design of gas distribution …). |
| • Optimization of doping level, thickness and roughness of LP-CVD ZnO layers, within nip- and pin-cells (a-Si:H, µc-Si:H and “micromorph” small-area cells).<br>• Increase of present cell current by 5%. | Partly achieved:<br>Work has been focused on pin solar cells [24][26]. Using a highly rough ZnO front contact allowed one to increase the photo-generated current by almost 40% compared to a cell deposited on a quite flat ZnO front contact. Further work has now to be done, to get high values of $V_{oc}$ and FF for pin solar cells deposited on highly rough front contacts. |
| • Optimization of nucleation layers on glass and other substrates, with a goal to increase the carrier mobility within the ZnO layers and roughness and to decrease free carrier absorption<br>• Reach a mobility value of 40 (at present 30). | Partly achieved:<br>The mobility has not been drastically increased (~ 35 $cm^2/Vs$), but we obtained ZnO layers which were at the same time conductive enough and very rough, without increasing the free carrier absorption. |

| Goals 03-04 | Achievements |
|---|---|
| **Material Characterization and device physics studies** | |
| Transmission electron microscopy study of growth of μc-Si :H on different substrates. | Fully achieved : Demonstration of the dominant role of substrate surface chemistry on nucleation and growth of μc-Si :H [34] |
| C(V) and CPM measurements on μc-Si :H cells | Partly achieved: C(V) measurement technique : test only on a single device, not definitively installed.<br>Fully achieved : CPM measurement refurbished in summer 04 and fully operational. CPM and FTPS yield identical results. |
| Raman measurements of series of solar cells | Fully achieved : Observation that the open-circuit voltage of μc-Si :H solar cells decreases linearly with average device crystallinity [20]. |
| Etablishment of reliable value for transport properties of μc-Si :H | Fully achieved : published in [3] |
| Measurement of $R_p$ and $R_s$ in μc-Si :H cells and their link with cell FF. | Partly achieved : measurement of $R_p$ and $R_s$ automatized in a new Variable Illumination Measurement (VIM) set-up, operational since October 04. Link with FF not yet fully established. |
| p-layer characterization | Partly achieved : p-layer crystallinity calibration by Raman achieved, but relationship between $R_s$, $R_p$ and spectral response not yet fully established. |
| i-layer stability | Fully achieved : observation of a mild degradation of μc-Si :H solar cells exposed to AM1.5 illumination as a function of layer crystallinity .[35] |
| Development of new characterization technique for μc-Si :H i-layers | Fully achieved : Fourier transform photoconductive spectroscopy (FTPS) installed, fully automatized. FTPS yields identical results as CPM, but is a much faster technique. |

# 8. Publications

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