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Thin film solar cells using impure polycrystalline silicon

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R. Kishore (²) (**), J. L. Pastol (²), R. Mertens (³), M. Caymax (³) and M. Eyckmans (³)

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Résumé. — On étudie les photopiles solaires épitaxiques dans le but d'utiliser comme matériau-substrat du Si métallurgique amélioré (UMG). On confirme que les éléments de transition ont ségrégé lors de la croissance du lingot et que B, P ou Al ne diffusent pas du substrat vers la couche épitaxique, qui a donc une qualité (résistivité ajustable, longueur de diffusion électronique élevée) adaptée à la production de bonnes photopiles. On a obtenu un rendement de photopile de 10,3 %. Des mesures de réponse spectrale, interprétées par un modèle simple, montrent que la combinaison des pertes à l'absorption et à la collection dans la couche épitaxique mince est responsable de la limitation du rendement. On conclut par une brève analyse économique et une comparaison de cette technique avec les autres aptes à l'utilisation de Si-UMG bon marché.

Abstract. — Epitaxial solar cells have been studied with the view of using rather impure upgraded metallurgical grade (UMG)-Si as a substrate material. It is confirmed that transition elements have segregated during ingot growth and that impurities such as B, P, Al do not diffuse from substrate to epilayers, so that the latter have resistivity and electron diffusion length adequate to produce good solar cells. 10.3 % efficiency cells have been obtained. By spectral response measurements, interpreted through a simple model, it is shown that this efficiency is limited by both absorption and collection losses in the thin active epilayer. With the help of a brief economic analysis, this technique is compared to the other ones able to make use of cheap UMG-Si.

1. Introduction.

Solar cell industry has now largely substituted poly-crystalline Si to single crystal as a material for solar cells. However the chemical quality of this raw material is still of the electronic grade (EG), or rather « off-grade » EG, i.e. EG-Si rejected by the electronic industry, which costs about 20 $/kg. Up to now, all attempts to deliberately produce a « solar grade » Si by halogenide or silane synthesis and cracking, through simplified processes to reach a cost of 10 $/kg, have failed, both in France and USA. As a result, the material cost still amounts to 30 or 35 % of the total module cost, at least in factories where cell and module production steps have been largely automatized, leading to a module cost limit of 5-6 $/Wp, which is still too high for very large scale applications.

The solution to this problem has been clearly indicated as early as 1980 [1], by the Wacker-Heliotronic company, which has a world leading position in the field. It is to reject the halogenide or silane steps and to purify metallurgical grade (MG)-Si by simple processes (gas leaching of molten Si, slag-melt reactions, acid etching of solid powder). This upgrading step may take several forms [2] and leads to a material called UMG-Si (Fig. 1) whose cost is only 3 to 6 $/kg.

It may be possible, in future, to use directly UMG-Si to produce conventional solar cells. However this goal may imply a combined use of severe selection of quartz and carbon sources and/or pre-purification of these raw materials [3] and/or sophisticated steps in UMG-Si processing [4] and/or double ingot crystallisation steps [5]. These production steps may well be too expensive when extrapolated to the high throughputs necessary for megawatt/year size solar cell factories.

We have chosen another way, which consists of using a rather impure UMG-Si, such as the one

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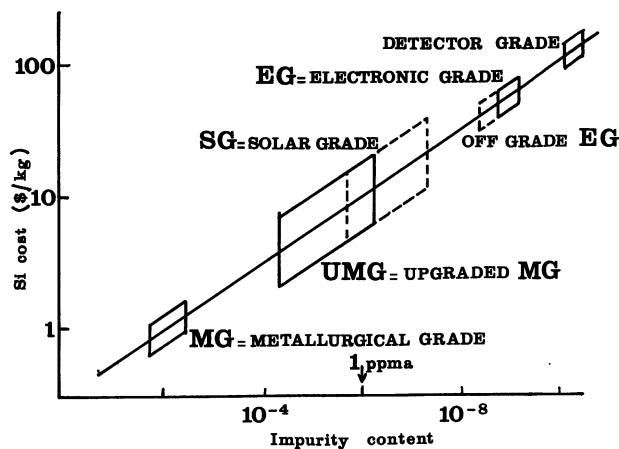(**) On leave from National Physical Laboratory, Delhi, India.

Fig. 1. — Purity and cost of different classes of Si.

studied by Péchiney [6], as a substrate on which an epitaxial layer is built. This epilayer is the active material absorbing solar radiation. After Chu [7], many teams have made such epitaxial solar cells (see Refs. in [2a, 8]). Our most recent results, published in [8], indicate a 10.3 % efficiency ; they will be summarized in part 3 of this paper, confirming the expectations exposed in part 2 about the impurity behaviour in the successive production steps.

Part 4 will be devoted to a particular aspect of epitaxial solar cells, i.e. the fact that these cells are really thin film cells : their active layer thickness is smaller than the minority carrier (electron) diffusion length. In a previous paper, some of us [9] have shown that the quality of these thin layers could not be assessed by the conventional LBIC measurement of this diffusion length L_n. We shall show it again for the spectral response measurements, which are a usual technique to measure L_n in completed solar cells.

This fact is correlated with the finding, by various authors [10], that the optimal epilayer thickness for such cells is near 20 μm, a value surprisingly lower than the thickness necessary to absorb most of the solar radiation : without light trapping, 20 μm thick layer absorbs about 80 % of the « useful » photons. We shall show that a reduced absorption efficiency is indeed the price to pay for the reduced quality of the substrate material, and explains essentially the maximum efficiency of 10-11 % found by all teams who made epitaxial solar cells.

Then part 5 will both discuss the ways to improve epitaxial solar cells and compare, on a technico-economic basis, this technology with other ones using UMG-Si.

2. Principles of the epitaxial solar cell production.

Our main problem is to get epitaxial layers which have both large grain size and low impurity content.

The combination of ingot growth by directional

solidification of molten Si and conventional epilayer growth by CVD at 1 120 °C should ensure centimeter-size crystals, so that the main difficulty is relative to impurities.

Figure 2 shows the sequence of processes that we use. The raw material is rather impure UMG-Si, which is transformed into ingots by directional freezing. Wafers cut from these ingots are coated with epilayers, on which complete solar cells are built. As shown in figure 2, different properties of the various impurities are used to get rid of their nocive effects.
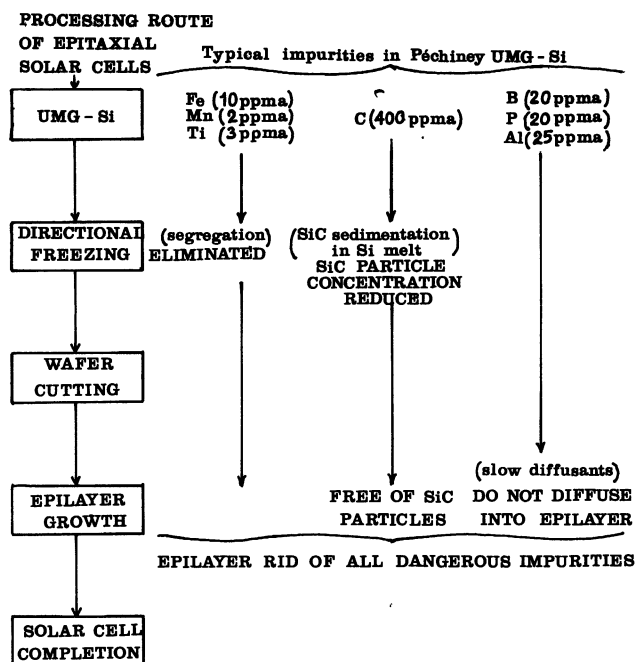

Fig. 2. — Processing steps of epitaxial solar cells and evolution of main impurities along these steps.

The transition metals, which appear in the ppm range in the raw material, should be eliminated during the ingot freezing, due to their very low segregation coefficients $(< 10^{-6})$.

Carbon exists as silicon carbide particles, which are expected to be sedimented in the Si melt, and thus to be absent from the largest part of the ingots, as well as the strains and dislocations induced by them.

Finally boron and phosphorus, which are abundant in the raw material and also in the ingots because they segregate only little, should not diffuse into the epilayers, owing to their low diffusion coefficients. Al, with a segregation coefficient of 2×10^{-3} and a coefficient of diffusion slightly higher than that of B, should be also eliminated by these two steps in series : ingot growth and epilayer growth.

The latter affirmation that B, P and Al diffusions can be neglected at 1 120°C has to hold, of course,
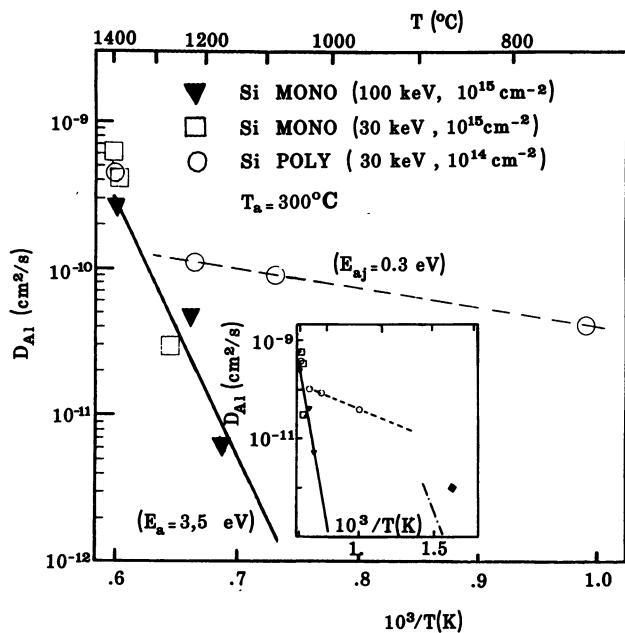
Fig. 3. — Diffusion coefficient of Al into single crystal and fine-grain polycrystalline Si [11b]. Al has been implanted (as indicated in parenthesis), then laser-annealed. The insert shows also the low temperature values of D^{Al} found by Hwang [12a] (——) and Nakamura [12b] (♦).

for polycrystals, while diffusion coefficients are currently known for single crystals. It was thus necessary to ensure that the intergranular diffusion coefficient was not much higher than the bulk one at 1 120 °C. This was done by a special study of the Al diffusion in Si, the results of which are shown in figure 3. This study used laser annealing to diffuse Al into Si single crystal or fine grain polycrystal, and SIMS to measure the diffusion profile [11a] ; by modelling the thermal and diffusion regimes, it was possible to deduce $D_{Al}(T)$ from the diffusion profiles induced by laser annealing [11b]. The model was validated by the results on single crystals, and then, the study of polycrystals showed that the intergranular contribution D^i_{Al} is dominant below 1 300 °C, but does not exceed 10^{-10} cm²/s at 1 120 °C. After our study, the apparent contradiction between our results and those of Hwang [12a] and Nakamura [12b] at 300-400 °C was resolved by Kazmerski [13]. For $D_{Al} = 10^{-10}$ cm²/s, the penetration of Al into polysilicon during a 40 min diffusion should be of order 5 μm. The cases of B and P are assumed to be similar, though no such detailed study is available.

For comparison, the corresponding diffusion range for Fe at 1 120 °C is in the cm-range. This shows that Fe (and other transition elements) should necessarily be eliminated during crystal growth, as mentioned above, whereas B, P and Al can be tolerated in large quantities in the substrates (maximum : 10 ppm Al, 20 ppm B and P).

3. Summary of experimental work by the CNRS-Vitry, CNRS-Meudon and IMEC-Leuven teams.

We give hereafter the main results of the collective work which has been presented at the recent PVSEC II Conference in Beijing [8].

Ingots were grown by a directional freezing technique, as explained in the original papers [14], one of which appears in this issue of *Revue Phys. Appl.* The crystals have centimeter-size except in the first five millimeters or so, which contain many carbide particles, inducing fine grains. The transition elements were found to have effective segregation coefficients (at the growth rate of 1 cm/h) often 10 to 100 times larger than the equilibrium ones, but yet low enough to obtain the following result : all these elements were, in the ingots, at contents very similar to those obtained for ingots grown from EG-Si. Their concentration were found smaller than the detection limit of nuclear activation analysis (e.g. for Fe : < 0.02 ppma) except for Ni (0.015 ppma), Cu (0.01 ppma), Co and Cr (some ppba).

These findings validate the expectations of paragraph 2 above on impurity segregation during ingot growth.

The resulting Si still contains large quantities of B and P. It is p-type, with a resistivity near 0.04 Ωcm and an electron diffusion length equal to 13-18 μm [6] : these values are consistent with a boron content of 20 ppma.

Epilayers and solar cells were made, using the processes developed by IMEC. After careful polishing and cleaning, the wafers were coated with pure Si layers grown from SiH_2Cl_2 pyrolysis at 1 120 °C, at a rate of 30 to 60 μm/h. Layer thickness was adjusted between 20 and 86 μm and layer resistivity between 0.1 and 3 Ωcm. As shown in [9], the quality of these layers can be assessed by LBIC measurements, only if the value of L_n to be measured is smaller than layer thickness ; for layer thickness 86 μm, we found L_n to be of the order of 60 μm. This is smaller than found in polysilicon ingots grown from EG-Si (80-100 μm), but yet sufficient in principle to build 12 % efficiency solar cells.

In order to demonstrate the effect of the epilayer as a screen for low-diffusing impurities contained in the substrate, we also used an ingot especially grown from EG-Si added with 1 560 μg/g of Al. Different wafers of this ingot were coated with epilayers ; the values of L_n in these epilayers are given by table I. While the traps brought about by Al reduce drastically L_n for Al concentrations as low as 10^{17} cm⁻³, reasonably good values of L_n can still be found in epilayers built on substrates doped 100 times more. This illustrates the aptitude of epitaxial cells to use impure substrates, as expected.

We chose substrates grown from UMG-Si and EG-Si + Al, coated with epilayers of thickness 25 to

Table I. — *Quality of epilayers grown on impure polycrystalline substrates.*

Substrates grown from	Al content	L_n of substrate	L_n of thick epilayers grown on substrate
UMG-Si	$< 10^{15}$ cm^{-3}	13-18 μm (*)	62 μm
EG-Si + Al EG-Si + Al EG-Si + Al	$1 . 10^{17}$ cm^{-3} $7 . 10^{17}$ cm^{-3} $5 . 10^{19}$ cm^{-3}	< 5 μm < 5 μm < 5 μm	50 μm 30 μm 25 μm

(*) Limited by boron doping : B content $\simeq 10^{18}$ cm^{-3}.

Table II. — *Best characteristics of epitaxial solar cells built on impure polysilicon substrates.*

Starting charge	EG-Si	UMG-Si from Péchiney 20 ppma B, 20 ppma P, 10 ppma Fe 25 ppma Al, 3 ppma Ti, 400 ppma C	EG-Si + 260 ppma Al (in principle : no other impurity)
Ingot growth	CZ mono-crystal	Directional freezing (1 cm/h) of polycrystalline ingots : ∅ 10 cm, height 9 cm	
Wafer preparation		Cut from middle and top parts of the ingots CP 4 polish, followed by RCA-cleaning	
Epilayer growth	No	At 1 120 °C, from SiH$_2$Cl$_2$ + B$_2$H$_6$ + H$_2$ growth rate : 30 μm/h	55 μm/h
Epilayer properties	—	Thickness 35 μm, $\rho = 1.6 \,\Omega$cm, $L_n = 62$ μm	Thickness 24 μm, $\rho = 2.2 \,\Omega$cm, $L_n = 50$ μm
Solar cell production	Screen-printing for junction, contacts and AR coating. Total area 3.24 cm^2, including grid (12 % coverage).		
Solar cell properties (for *total* area)	T2A $J_{sc} = 28.4$ $V_{oc} = 531$ FF = 0.74 $\eta = 11.2$	P2 R15/54 $J_{sc} = 25.3$ mA/cm^2 $V_{oc} = 561$ mV FF = 0.72 η(AM1) = 10.3 %	R36 H45 $J_{sc} = 22.7$ mA/cm^2 $V_{oc} = 529$ mV FF = 0.71 η(AM1) = 8.5 %

35 μm and resistivity 1.6 to 2.2 Ωcm, to build solar cells by the integral screen-printing technique. As explained in the original papers of the IMEC-Leuven group [15], this technique implies heat treatments below 900 °C, is inherently cheap, and gives reference cells (for single crystal EG-Si without epilayer) of maximum efficiency 12.5 %. Table II summarizes the properties of some of our solar cells : a reference one with a pure crystal and no epilayer, and two epitaxial cells made on impure substrates.

A first conclusion at this point is that a rather high efficiency of 10.3 % has been obtained, using a very impure UMG-Si substrate (containing B, P, C in large quantities). A 35 μm thick epilayer of pure Si

has played here the expected role of high quality active layer deposited on the impure substrate. The efficiency approaches the best values quoted in the literature for epitaxial cells : 11 % by the Spire group [16], 10.5 to 12 % by the Crystal System-RCA group [17], both working with a more sophisticated cell processing technique ([17] quotes 13.4 % for reference cells).

A second conclusion is that the Al-rich substrate gives a somewhat lower efficiency. Here the layer thickness has been reduced to 24 μm, and the epilayer quality is also reduced ($L_n \simeq 50$ μm for $N_{Al} \simeq 1 \times 10^{17}$ cm^{-3} in the substrate, cf. Table I), which may account in part for the reduced efficiency. A complementary explanation is that, even in single

crystals, heat treatments at 900 °C are known to seriously degrade the diffusion length of Al-doped Si [18], so that the active layer diffusion length after cell completion may be much smaller than before, at least in the 5 μm nearest to the substrate.

There is a more general factor which prevents to correlate simply the efficiency η of epitaxial cells with the quality L_n of their active layer ; we shall discuss this problem in the next paragraph, in conjunction with the study of solar cell spectral response.

4. Spectral response of epitaxial cells, viewed as « thin film » solar cells.

The correlation between active layer quality and solar cell properties is well established in conventional cells. It can be summarized in figure 4, which shows the decrease of short circuit current due to : i) impurities in single crystals, according to Davis [19], ii) grain boundaries, according to Sopori [20]. It is striking to note that all these different imperfections reduce the short circuit current in a similar way ; the relation between J_{sc} and η is also the same for all impurities, as stated in [19].

The usual way to determine L_n in completed cells consists of measuring the spectral response and tracing $1/Q$ vs. $1/\alpha$, where Q is the internal quantum efficiency (corrected for grid area and surface reflectivity) and α the absorption coefficient of Si (depending on wavelength). However it is easy to see that this technique is not valid generally for epitaxial cells. For the same reason figure 4, which is valid for an infinite thickness d of the active layer, cannot be valid for epitaxial cells.
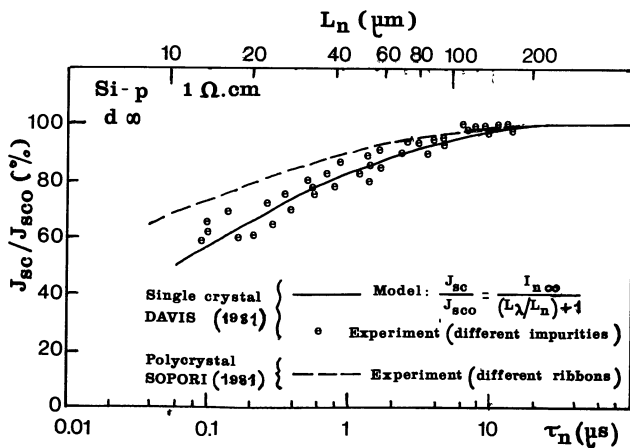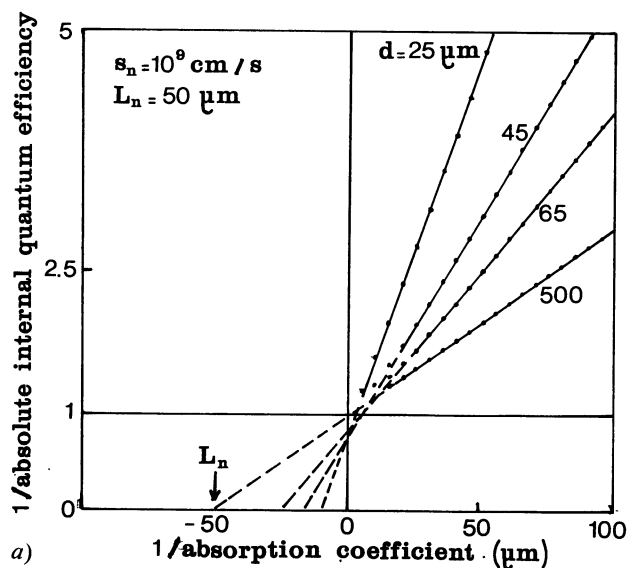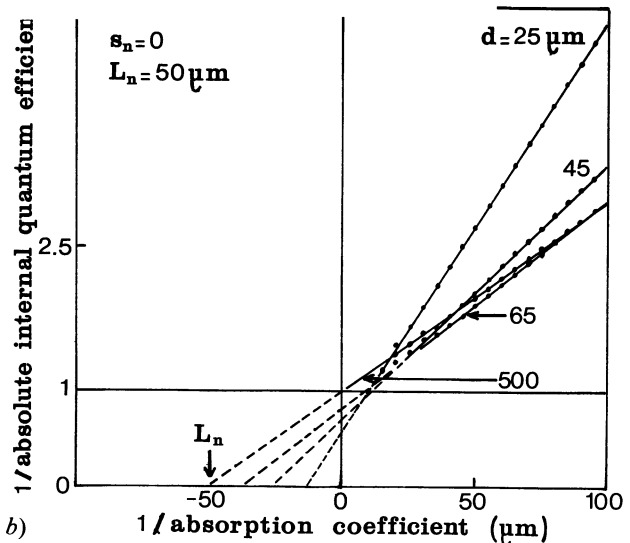
The argument is as follows. We are interested in the infrared photons $(0.8 < \lambda < 1.1 \, \mu m)$ which are absorbed in the bulk active layer. We take the absorption coefficient value α that is given by Runyan [21]. The complete expression of the quantum yield Q is written as a function of d, L_n (thickness and diffusion length of the active layer) and S_n (surface recombination of the back face of the active layer) ; we use for this equation (19) in the book of Hovel [22]. It is well known that, for $d/L_n \gg 1$, Q can be approximated by the function $Q = \alpha L_n / (\alpha L_n + 1)$ independent of d and S_n. But this is no more true when $d/L_n < 1$. The result of the exact calculation is illustrated in figure 5a and 5b for the respective cases of $S_n \, \infty$ and $S_n = 0$. Contrary to

a)

b)

Fig. 4. — Solar cell short circuit current J_{sc} vs. active material quality described by electron lifetime τ or diffusion length L_n. Signs e show Davis's experimental results for $J_{sc}(\tau)$ in single crystals doped by various impurities ; the full line describes a simple model for this case [19]. The broken line shows the mean $J_{sc}(L_n)$ variation found by Sopori [20] in polycrystalline ribbons. The scales of τ and L_n were fitted together using the electron diffusion coefficient $D_n = 20 \, cm^2/s$.

Fig. 5. — Internal quantum efficiency vs. absorption coefficient, for fixed L_n and variable layer thickness : a) for back surface recombination coefficient $S_n \, \infty$; b) for $S_n = 0$. In the latter case, the curve crossings are due to back surface field (cf. [10a]).

the case of infinite thickness which allows to deter-mine L_n unambiguously, then the extrapolation of $(1/Q)$ vs. $(1/\alpha)$ does not give L_n any more, but a value which is more or less correlated to d.

This result looks like the one already obtained experimentally by LBIC measurements [9] ; however the calculation would be more difficult in that case because the problem is two-dimensional.

Another way of looking at the results in figure 5 is to draw Q as a function of L_n for fixed α and d. It is clear from figure 6 that the value of Q depends principally on the smallest of the two quantities (d, L_n).

In other words, collection losses reduce the cell efficiency only for imperfect and thick active layers $(L_n < d)$. They are suppressed when $d < L_n$, but

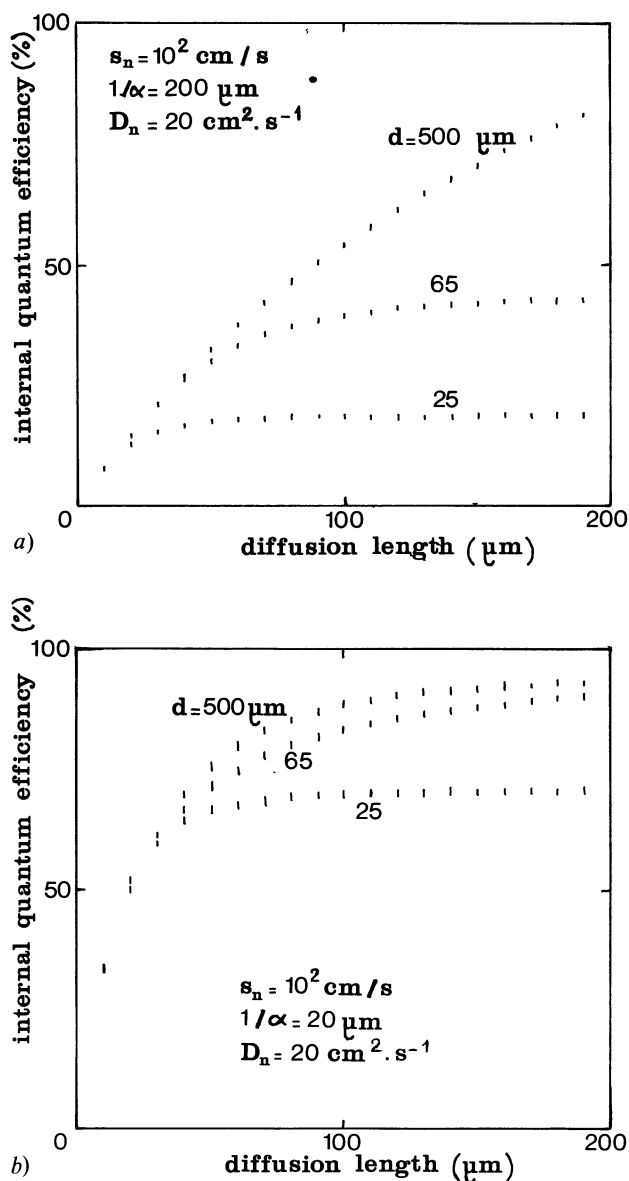may be replaced by absorption losses if d becomes lower than $1/\alpha$, i.e. in the infrared part of the spectrum, near the absorption edge.

Let us now come to the experimental measure-ments of $Q(\alpha)$. Figure 7 shows the spectral re-sponses of the three cells described in table II (measured at low injection rates). All cells have similar low quantum efficiencies for high energy (blue) photons, because of high recombination at the front surface and in the volume of the P-doped layer. The infrared quantum efficiency is high in the reference cell T2A of quasi-infinite thickness : it is reduced in cell P2R15/54 made on UMG-Si and still more in cell R36H45 made on Al-doped EG-Si ; both collection and absorption losses may contribute to this reduction of Q. Since in both cases $d < L_n$ before cell processing, collection losses can only arise from L_n degradation during cell processing : absorption losses are expected to be more important in the R36H45 cell because of thinner epilayer and are likely to explain most of the efficiency reduction.

In other words, the active layer of epitaxial cells is thin enough to decrease, or even suppress, collection losses due to layer imperfections, but so thin that absorption losses begin to be important in the infrared, near the Si bandgap energy. For such cells, the J_{sc} vs. L_n curve should lie well above the ideal curves in figure 4 (valid for infinite thickness).

It can be concluded that epitaxial solar cells are indeed « thin film » solar cells. The optimal value of epilayer thickness d has been found of order 20 μm by previous authors [10], only because they used very impure substrates ; in such cells the determi-

a)

b)

Fig. 6. — Internal quantum efficiency vs. material quality for fixed wavelength and variable layer thickness : a) $\lambda = 1.02$ μm ; b) $\lambda = 0.86$ μm.

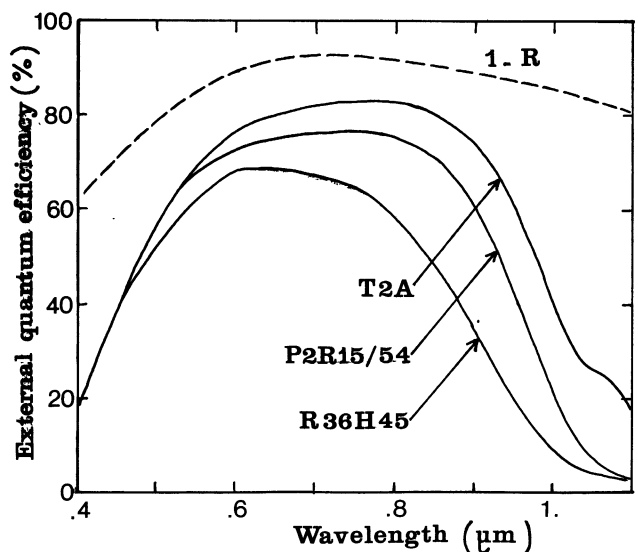Fig. 7. — Spectral response of three solar cells described in table II, corrected for grid area but not for front surface reflectivity R : T2A = reference cell (single crystal, without epilayer) ; P2R15/65 = UMG-Si + epilayer ; R36H45 = Al-doped EG-Si + epilayer. Also shown by... is the maximum quantum yield due to reflectivity losses.

nation of L_n, made by authors [16] and [17], may be incorrect : they have found values of 13 μm and 30 μm respectively, which may describe their epilayer thickness (18 and 20 μm respectively) rather than L_n. If higher quality epilayers were obtained, like in our P2R15/54 case, the optimum thickness would be displaced towards higher values, say 50 μm : but this has no interest since, as we shall see below, the cell cost would then become prohibitive.

Thus the natural application range of epitaxial cells is the use of very imperfect Si substrates and thin (20 μm) epilayers, to obtain 10-12 % efficiency solar cells, the efficiency being limited by the absorption losses in the thin epilayers. In order to reach 12 %, two ways seem possible :

— reduce absorption losses by light trapping, using a back reflective surface, but this seems difficult in our case where the back surface is a Si/Si interface,

— improve the cell processing technique, which seems to be feasible since the industrial process in use to-day for polycrystalline cells gives higher efficiencies than the integral screen printing technique and may even be further refined [23].

of epitaxial solar cells is to compare them with those offered by other proposed ways to use UMG-Si. Such comparisons can only be qualitative.

UHM-Si twice recrystallized was shown by the Exxon group [5] to be pure enough for 13 % efficiency cells to be built on it. However a second crystallization may be more expensive than a CVD epilayer growth step. Up to now, the Pragma study [2d] seems to fall in the same category of processes demanding a double crystallization.

The same is true, in a sense, of the process developed by Amouroux [4] at ENSCP (France), in which the last step of MG-Si upgrading is Si melting in a plasma furnace, prior to ingot growth by some directional freezing technique. This technique seems to have two advantages : possible use of rather impure MG-charge, and high speed of the plasma treatment. However this technique has still to be upscaled to the range allowing industrial production of solar cells (yearly production of at least 1 MWp).

Another interesting technique, developed by Siemens [3], implies a very clean arc furnace reaction of prepurified C and SiO_2, but there again upscaling and cost have still to be studied.

5. Economical analysis and comparison with other technologies presently being developed.

The competitivity of epitaxial solar cells has been discussed in 1983 [6] by comparing its main asset — lower cost of raw material — to its main liability — the introduction of the CVD step. For instance an overcost of 250 F/m² (38 $/m²) due to the latter was shown to just compensate a cost advantage of 100 F/kg (15 $/kg) due to the former.

Today industrial cells are made from offgrade EG-Si, which is much cheaper (12 to 24 $/kg) than the material currently used in 1983. For the UMG-Si to be used in epitaxial cells, at least one commercial product has appeared on the market, as a result of the Elchem group work [5], and its cost seems to be lower than expected in 1983 (3 to 6 $/kg). Consequently it is realistic to admit an initial cost advantage of 15 $/kg for the raw material, in favour of epitaxial cells.

The corresponding allowed overcost for the CVD step would be 38 $/m² (0.4 $/wafer) according to the 1983 calculation (made for a wafer thickness of 300 μm) ; with the current decrease of wafer thickness, the limit should be put at 30 $/m² (0.3 $/wafer). Whether or not this limit can be reached with an industrial CVD reactor of sufficient capacity remains to be seen. If we assume this condition to be fulfilled, a wafer cost near 1 $/Wp would be obtained.

Another way of evaluating the future possibilities

6. Conclusions.

As long as there is no demonstrated process to produce conventional solar cells from UMG-Si at the required scale, the epitaxial route seems to be a possible alternative, provided that a new type of CVD-reactor is designed. Epitaxial cells are essentially thin film cells ; their best range of application is in conjunction with very imperfect Si. The adapted raw material may be rather impure UMG-Si ingots as mentioned in the present paper, or eventually very imperfect ribbons, such as those produced by the plasma torch process [24].

Apart from these practical conclusions, it should be stressed that the evolution towards thin film cells can be noted presently in several areas of the Si solar cell technology. For instance R. Hezel [25] has shown that a new type of « back-MIS cell » is much better ($\eta = 13$ %) for active layer thickness of 80 μm than for 300 μm, and his explanation uses a model similar to that of paragraph 4. Also A. Barnett [26] uses only 20 μm of solution-grown Si to build 9.5 % efficiency solar cells on stainless steel and hopes to take profit of the steel/Si interface to decrease the absorption losses by light trapping. Even the field of amorphous Si offers a similar evolution : the use of very thin (< 3 000 Å) intrinsic layers is the main solution to the collection degradation by the Stäbler-Wronski effect and the resulting absorption losses are currently reduced by light trapping and tandem structure.

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