

Special Issue

# Consolidation of Thin-film Photovoltaic Technology: The Coming Decade of Opportunity

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*The previous decade, particularly its latter half, has been an exhilarating period for photovoltaics. Companies have entered a new era of profitability with photovoltaic company stocks now some of the most eagerly sought. The challenge for the next decade is for the industry to take advantage of the increased resources made available by the recent boom to position photovoltaics for an onslaught into wholesale electricity markets. It is argued that only the best of the thin-film technologies are likely to be able to lead such a charge. Criteria for the success of thin-film technology in the long term are discussed. Copyright © 2006 John Wiley & Sons, Ltd.*

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## 1. INTRODUCTION

The last decade has been an important period for photovoltaics (PV). Annual module production has increased 20 times while manufacturing costs have reduced enormously. If this performance is projected to the coming decade, the industry will be producing 30 GW/year of modules by 2015 with module prices approaching US\$1/Watt. The largest manufacturers may well by then have more than 5 GW/year capacities, consuming the entire production of a large float-glass factory. Photovoltaics should be competitive with retail electricity prices in most places of the world.

However, it is argued that even lower prices are needed for photovoltaics to make any significant impact upon wholesale electricity and energy markets. Only the best of the thin-films are likely to be able to reach the very low costs required for significant penetration of this market. Criteria for the long-term success of thin-film photovoltaics are discussed in the following sections. These are believed to relate to improved efficiency and the use of non-toxic, abundant and durable materials.

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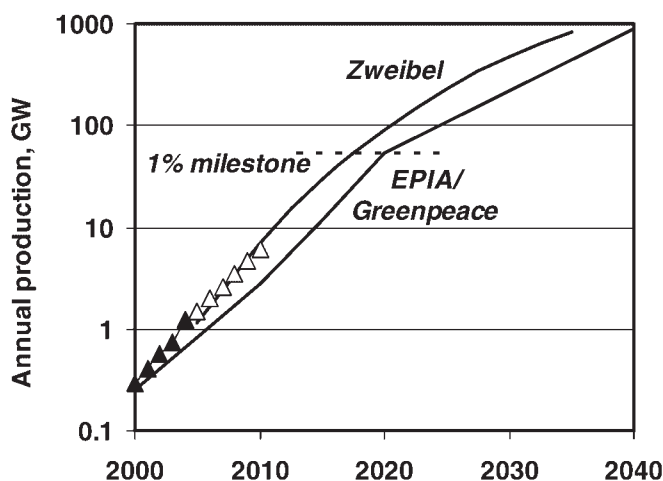


Figure 1. Annual production of photovoltaic modules showing actual data (▲) plus near-term (△) and long-term projections

## 2. FUTURE PRODUCTION VOLUMES

Figure 1 shows actual annual photovoltaic production volumes for the 2000–2004 period (closed triangles) together with various projections for the future. The first (open triangles) show recent estimates for production over the coming 5 years based on analysis of the current business situation.<sup>1</sup>

In a landmark study published in 2001, the European Photovoltaic Industries Association (EPIA), in conjunction with Greenpeace, published a forward-looking report highlighting the potential of photovoltaics to provide a significant part of the world's electricity requirements from 2020 onwards.<sup>2</sup> Future contributions were estimated by assuming 27%/annum production growth from 2000–2010, 34%/annum growth over the 2010–2020 decade and 15%/annum growth from 2020–2040. As can be seen from Figure 1, reality has overtaken conjecture and what initially seemed like quite optimistic projections have been shown to be conservative, at least for the first few years.

More recently, Zweibel<sup>3</sup> has made an even longer term projection, taking into account the rapid recent progress. He outlines a 60-year scenario, resulting in the installation of sufficient photovoltaics to supply essentially the entire world's energy requirements by 2065. The assumed annual growth rates seem quite reasonable based on recent experience. Zweibel assumes that growth peaks at 38%/annum around 2010, steadily relaxes to 20%/annum by 2020, 10%/annum by 2035, and finally settles down to 5%/annum by 2065.

Had it not been for the constraining effect of silicon supply, it seems that growth to 2010 would have outstripped even these updated estimates. As well as the need for material availability to grow as quickly as photovoltaic manufacturing capacity, a key requirement for attaining such volumes is, of course, a rapidly growing market for photovoltaic product. Substantially decreased cell costs are argued to be essential to sustain the growth rates projected in Fig. 1.

## 3. REQUIRED COSTS

A reference point for current costs is provided by the German PV feed-in tariff scheme, which has been responsible for much of the recent development of the photovoltaics industry. Here, a fixed tariff of about 50 Euro cents/kWh for PV-generated electricity is creating unprecedented demands for photovoltaics, at system installation prices of about 6 Euro/Watt. To generate a similarly enthusiastic demand if the tariff were set equal to a typical retail electricity price of 18 Euro cents/kWh would require installed system prices of about 2 Euro/Watt. With the tariff equal to the average wholesale electricity rate of about 5 Euro cents/kWh, installed system prices of 0.6 Euro/Watt would be required.

If the traditional ratio of about one-half is maintained between module and systems costs, this means that photovoltaic module costs have to be less than US\$0.50/Watt for any significant impact at the wholesale level in Germany, preferably a lot less.

Similar conclusions are reached from a study of photovoltaic generation costs across Europe and North Africa.<sup>4</sup> Costs similar to feed-in tariff values are calculated for residential systems in Germany in this study. In the sunnier regions of the Mediterranean and North Africa, calculated costs are about half these. With the economies of scale possible from larger ground-mounted systems, system costs of Euro 4/Watt were considered feasible at present for such large systems, with corresponding electricity costs reducing to Euro 0.15–0.20/kWh in sunnier regions.

It is concluded that the reduction in module prices from circa Euro 3/Watt to Euro 1/Watt that might be expected over the coming decade would make photovoltaics competitive with retail electricity prices over much of Europe. To progress to the next stage, photovoltaic costs must come down appreciably to be attractive compared to wholesale prices.

Another way to get the feel for the costs required for photovoltaics to make an impact upon wholesale markets is to compare to the case of wind electricity. Although photovoltaics have some advantages over wind, wind system costs are now similar to those expected of photovoltaics beyond 2015. At installed system costs of about Euro 1.00/W and in good wind areas where capacity factors might be 25–40%, wind energy is, at best, only grudgingly accepted as being marginally economic relative to fossil and even nuclear sources. Wind consequently has not been embraced as an option of choice by many large scale electricity producers.

China and India, where economic growth is presently contributing to the world's on-going demand for more energy, both have active wind programs. However, wind technology is still regarded as far more expensive than alternatives in such critical growth regions. For example, wind resources in the Chinese province of Inner-Mongolia are enormous but so are the coal resources. The recent rate of installation of coal stations in this province has outstripped the wind installation rate by many orders of magnitude.<sup>5</sup> Installed photovoltaic system prices of below Euro 1.00/W would be required to duplicate the present economic position of wind energy in good locations, given the lower capacity factor.

How low are possible costs? Low-emissivity glass that may contain eight sputter-deposited layers (but does not require patterning) apparently can be sold profitably in volume at US\$25/m<sup>2</sup>.<sup>6</sup> Photovoltaics is likely to be more expensive due to the need for patterning and encapsulation, but US\$30–45/m<sup>2</sup> seems, on this basis, to be a not unreasonable estimate of achievable prices, corresponding to US\$0.20–0.30/Watt at 15% module efficiency. Similar figures are estimated by a bottom-up approach.<sup>3</sup>

It seems unlikely that wafer or ribbon-based technology could ever reach such low costs due both to high material content and the large number of pieces that need to be handled, measured, inventoried and interconnected.

Until recently, it has seemed that the success of the wafer-based approach has inhibited rather than facilitated the commercialisation of thin-film technology. Such technological 'lock-in', where a technology arguably inferior in the long term to other options excludes these other options by gaining an early dominant market position, is thought to be not uncommon.<sup>7</sup> However, the recent silicon supply shortage has opened the door for thin-films and given them the opportunity to establish their credentials, an example of market forces working against 'lock-in' (although there seems to be no guarantee these forces will always do so).

Without this opportunity, thin films may have been kept in a perpetual state of immaturity. Meanwhile, the non-competitiveness of photovoltaics on the wholesale market would have led to increasing reliance on fossil fuels, notwithstanding the obvious negative greenhouse impact, and possibly to the spreading of nuclear plants to all corners of the globe, notwithstanding the concurrent weapons proliferation and waste disposal issues.

#### 4. POSTULATED LONG-TERM REQUIREMENTS

The present author has previously listed a set of postulated requirements for photovoltaic technology to be successful in the long-term. Based on the argument that high production volumes will automatically result in costs approaching material costs, these postulated requirements are that the technology be: (1) thin-film;

(2) high-efficiency; (3) non-toxic; (4) based on abundant materials, and (5) be durable. These postulated requirements are discussed in the remaining part of the paper.

## 5. THIN-FILM

The potential advantages of thin-film technology over wafer-based technology are fairly clear. Much less material is involved, the unit of production is much larger and cell interconnection occurs as an integrated part of module fabrication. However, the strengths of the wafer-based approach and problems associated with the immaturity of thin-films have, until recently, prevented these potential advantages from being exploited.

The strengths of the wafer-based approach are excellent durability, relatively high conversion efficiency, relatively low capital costs for capacity expansion, the relative maturity of the technology and its supply chain, and the potential that still exists steadily to reduce costs.<sup>8,9</sup>

As noted, the silicon supply shortage which became apparent in 2004 has given the opportunity for thin-film companies to expand market share. One industry analyst's view<sup>1</sup> is that the thin-film share will expand from 6% in 2004 to 12% in 2006 (from 70 MW/annum of thin-film product to 210 MW). The same analyst expects that this trend will reverse once silicon supply is again able to keep up with demand. Another possibility is that one or more thin-film technologies will be able to take advantage of this opportunity to establish a market presence and to expand to the production volumes required to capture inherent cost advantages.

For thin-films to maintain or enhance market share post-2006 would require a marketing advantage over the wafer-based approach, apart from the present advantage of availability. Lower cost is the most likely marketing incentive in the short-term, although some thin-films may have other advantages in the long-term, such as in durability.

## 6. HIGH-EFFICIENCY

The expected market area for most thin-film product over the coming few years is in large grid-connected systems being installed in Germany and Spain.<sup>10</sup> The Japanese program is reportedly also about to encourage such systems.<sup>11</sup> Here, the lower efficiency of the thin-films is not a huge disincentive to their use. Improved efficiency of thin-films is essential not only so that thin-film product more directly competes with wafer-based technology in other market areas, but also because of the impact of efficiency upon long-term manufacturing costs.

For all the thin-films, efficiency should increase with more production experience and as production processes are more finely tuned. Stacked or tandem cells offer another approach to improving efficiency. This approach has been used commercially for several years by United Solar who uses a 3-cell stack consisting of an a-Si cell on top of two a-SiGe cells.

The alternative of a-Si/ $\mu$ c-Si tandems appears of interest to other manufacturers, with Kaneka, Sharp and Fuji either producing, or planning to produce, such modules.<sup>12</sup> Although such a 2-cell stack improves efficiency from the 6–7% range to the 8–10% range, it appears that present economies are not strongly in favour of the tandem design. As production volumes increase and deposition costs correspondingly decrease, advantages are expected to become more pronounced.

Chalcogenide-based tandem stacks are also under investigation. Here, there are quite severe challenges in terms of matching the different processing requirements of the upper and lower cell, with no guarantee of successful outcomes.<sup>13</sup>

Third-generation approaches, such as based on hot carrier or quantum dot solar cells may make laboratory progress, but are not expected to have made a commercial impact over the timescale of present interest.

## 7. NON-TOXICITY

Obviously, a non-toxic photovoltaic technology would pose fewer issues than a toxic technology and be a clearly superior choice, other aspects being equal. The present strengths of CdTe in terms of its manufacturing

robustness and the resulting low capital cost of deposition equipment are providing a test case for the impact of toxicity.

Present CdTe modules have the CdTe layer trapped between two glass sheets that is reported to keep the material out of harms way, even during fires<sup>14,15</sup> (fire performance testing was done with slow and uniform heating where the glass fused rather than cracking, as might be expected from rapid, non-uniform heating; if such testing is valid, this may constrain the technology to the heavy glass–glass encapsulation scheme which may be a disadvantage as module size increases). The presently largest manufacturer of CdTe modules, First Solar, announced an insurance policy approach to funding the estimated future costs of reclaiming and recycling First Solar modules at the end of their use.<sup>16</sup> Such an approach addresses concerns about large numbers of modules containing toxic materials accumulating in the field and being left to an uncertain fate should the technology become superseded.<sup>17</sup>

Under present European legislation, both CIS and CdTe modules contain sufficiently concentrated Cd (greater than 0.1% by weight of Cd in a unit that cannot be mechanically disjointed) to be banned from sale on the European market from 1 July, 2006. While photovoltaic consumer products are covered by the impending legislation, photovoltaic modules are exempted with such exemption to be reviewed at least every 4 years.<sup>18</sup>

Exemptions may be given when it is likely that the elimination or substitution of the cadmium causes an increase in the environmental health and/or consumer safety impact.<sup>18</sup> While Cd is produced as a by-product of Zn mining, which can have rather severe environmental consequences including substantial Cd emissions, proponents of CdTe technology argue that the bulk of these undesirable consequences are not legitimately attributable to Cd production but to Zn production. Furthermore, they argue that sequestering the Cd in photovoltaic modules combined with good recycling practices is a far sounder environmental outcome than leaving the cadmium in the waste stream from Zn mining.<sup>19</sup>

The same argument could undoubtedly be applied to many other products incorporating toxic materials. However, the lack of resolution shown by European legislators in addressing the issue of NiCd batteries in power tools<sup>20</sup> suggests there might be at least a near-term window of opportunity for Cd-based photovoltaic technologies.

Notwithstanding the present situation, the looming threat of unfavourable legislation must act to some extent as a disincentive to unfettered large scale investment. The toxicity of cadmium will also likely to make CdTe product less desirable on the market, making it likely to attract lower market prices than a non-toxic competitor, other features being similar.

## 8. ABUNDANT MATERIALS

The recent silicon wafer supply shortage has given the industry first-hand experience of the effects of a widespread shortage of critical material. However, while silicon is, after oxygen, the most abundant element in the earth's crust, some of the other materials that are being investigated for photovoltaics are amongst the rarest. Ga, Ge, In, Ru, Se and Te, all classified either as 'scattered' or 'precious' metals, are all mined in smaller quantities than the archetypically scarce metal, Au.

The low concentration of these elements, even when concentrations are enhanced in mineral ores, means that large quantities of ore must be mined to extract them. This would be prohibitively expensive (and environmentally disastrous) except for the fact that these rarer elements frequently occur in combination with concentrates of materials already mined in larger volume, specifically bauxite (Al), Zn, platinum group metals, and Cu. This allows these scarce elements to be extracted as by-products of the mining of metal used in larger volume.<sup>21</sup>

One result is that most of the economic and environmental cost of extracting the by-product is borne by the primary product. Zn mining is dirty and dangerous but, as noted, it has been argued that Cd, a by-product, should not be held responsible for this, since Zn mining would go ahead regardless of Cd demand.

A similar situation prevails with the pricing of these by-products, where the law of supply and demand is seen at its most basic. When demand falls below supply, prices for these by-products drop, sometimes to below the costs of extracting them from the primary waste-stream, and stockpiles increase. When demand picks up, prices increase, skyrocketing once inventories are drained and all existing 'by-product' production capacity is brought

on-line. Even in periods of high prices, the ability to bring new production facilities to bear is limited. Moreover, the rapid fluctuations in the price caused by changing demand increase the financial risk of any undertaking designed to increase supply.

These trends are illustrated by recent price experience with In, Se and Te. After all three dropped to low prices in 2002 after a period where supply exceeded demand, the price of each of these metals has since risen quite dramatically.

Indium is produced, along with Cd, as a by-product of Zn mining. Demand from the flat-panel display industry has been responsible for the recent pressure on indium prices. Since 2002, the demand for indium has increased by 40% each year,<sup>22</sup> a figure not dissimilar to the PV industry growth rate. This has pushed indium prices to 65-year highs as shown in Figure 2. The ability of the supply chain to respond has been quite limited (primary production is expected to increase by only 10% from 2003 to 2007, from 355 tonnes to 396 tonnes.<sup>22</sup>) Closures of several Zn refineries, due to the high levels of pollution they were creating, have compounded the supply problem. Were it not for fervent recycling of indium from sputter targets, where most is being used, prices would be substantially higher.

Even at present prices, the value of the indium contained in each tonne of zinc ore mined is still much less than that of the Zn content. Decisions about Zn production and hence indium production will be made based on demand for Zn and not that for indium. The true cost of extracting indium due to its low concentration is, without assigning value to the Zn 'by-product', extremely high due to its low concentration (similar to that in an Au or Pt ore).

A similar situation holds for Se and Te which are produced as by-products of Cu refining. Increasing Chinese demand for Se for glass, steel and fertilizers put pressure on inventories and available production from 2003.<sup>23</sup> According to an industry commentator, all available Se was finding its way to the market by May, 2005, when it reached record prices. A subsequent relaxation in price may be due to substitution of Se in some of its uses, given the record prices.

A similar increase in demand in Te for use in metal alloying and in thermoelectric devices has similarly pushed this metal to record prices. Again, there is limited scope for increasing the production of Te in the near-to-medium term.

Allowing for present inefficiencies in use, the present requirement for these metals is about 0.1 g/Watt.<sup>24</sup> The present price of indium at \$1,000/kg would therefore contribute about \$0.10/Watt to module costs. However, this estimate overlooks the fact that the grades of metal represented in Figure 1 are standard, essentially

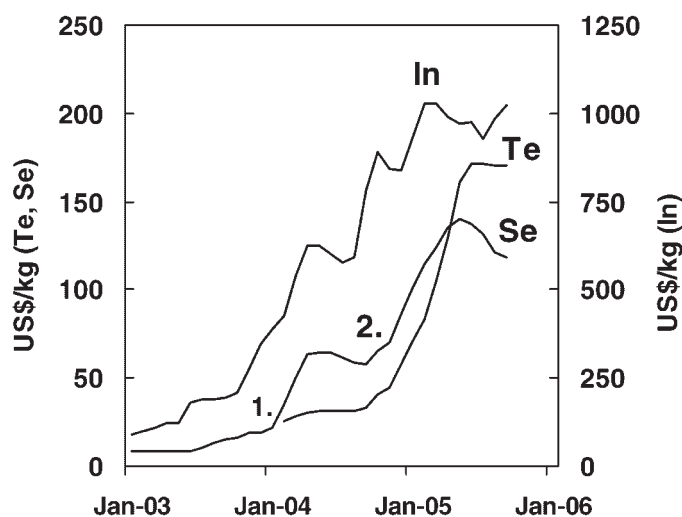


Figure 2. Prices for In (99.99%), Se (99.5%) and Te (99.95%) over the 2003-2006 timeframe (www.metalprices.com). Ref. 23 attributes the steep rise of Se curve at point 1 to 'depletion of Cu slimes' and that at point 2 to 'depletion of Ni slimes'



'metallurgical', grades. Semiconductor grades would normally be required in cell fabrication. Historically, semiconductor grades of In, Te and Se have been 5–10 times more expensive than standard grades.<sup>25</sup>

A manufacturer producing 5 GW of photovoltaic panels using these materials in 2015 would require 200–500 tonnes of these materials per annum, depending on improvements in utilisation efficiency implemented over this timeframe. A single large manufacturer would therefore put considerable price pressure on these materials by this time, particularly In and Te, considering that 2003 total production volumes were 355 tonnes, 304 tonnes and 2125 tonnes for In, Te and Se, respectively.<sup>22,26</sup>

Basing future cost estimates upon historical costs for these elements may not be appropriate. Once demand exceeds the availability from the preferred by-product stream, a new, more expensive, source must be found. The cost of extracting these elements directly from ores would be very high, probably not too dissimilar from the cost of extracting Au and Pt, given the similar concentration levels.

The recent price rises for these metals may actually be a positive situation for photovoltaic technologies dependent on them. If present prices are sustained, and if these are sufficiently high to encourage either increased extraction efficiencies or even primary production, a stable supply of intermediately priced product able to support a growing photovoltaics industry may become available.

However, as previously mentioned, this means that material costs of technologies relying on scarce or scattered materials, if these technologies are successful, are likely to be higher than estimated based on historical prices of such materials.

## 9. DURABLE

It has been argued elsewhere that it is the lack of a demonstration of a similar level of durability that has worked against the acceptance of thin-film photovoltaics in the past.<sup>27</sup> The present market situation is finally giving thin-film technologies the chance to establish their credentials in this area. With this chance, of course, comes a risk that one or more will not perform as well as expected.

Wafer-based modules have demonstrated an impressive level of field reliability. Loss in output is generally less than 1% per year with increasing contact resistance, cracked cells, delamination and the discolouring of the polymeric encapsulant between the cell and the glass cover the main sources of such loss. There are many examples of modules that have performed well after more than 20 years in the field.

There is, of course, much less data available for the thin-films. A well-monitored test system for modules was installed near the German town of Koblenz-Gondorf on the Moselle River in 1988 with significant quantities of early a-Si modules provided by 3 manufacturers. (All three had reported establishing manufacturing facilities in 1984 and earlier modules from these facilities had undergone several years of testing by this point in time<sup>28,29</sup>). One of these module types performed respectably at Koblenz-Gondorf, degrading to 70% of nameplate rating after 10 years, showing that reasonable performance was possible. The other two module types performed badly (below 10% of nameplate rating after 6 years showing it was also possible to get very poor performance).<sup>30</sup> The first large a-Si system was installed at Davis, California in 1992. Originally operating consistently at 479 kW<sub>ac</sub> rating, the system was recently re-rated to 290 kW<sub>ac</sub>, or 60% of the original rating after 12 years,<sup>31</sup> suggesting a similar degradation rate to the earlier array. A system at the same site based on ribbon silicon was rated at 80% of its original rating after 11 years in the field while one using monocrystalline silicon seems to have retained its original rating.<sup>31</sup>

Testing of a 1.8 kW a-Si triple junction array reportedly was commenced by NREL in 1994. The performance after 10 years in the field was within 10% of nameplate with performance over the last 4 years of testing deteriorating quite predictably at 0.74%/year, similar to wafer-based modules.<sup>32</sup> Similar rates of 1.1%/year have been measured for subsequently installed systems.<sup>33</sup> Some problems have been reported for other module designs that use conducting oxides on the glass surface.<sup>34,35</sup> Despite some initial problems early in their history, it would seem from such data that a-Si modules at least have the potential to give a level of reliability not too dissimilar from that of wafer-based silicon, once the initial 20% stabilisation loss is accounted for.

Historically, the rear contact of CdTe cells has given rise to stability issues. Possibly for this reason, there seems to be limited long-term field data for this technology. Data have been reported for a 1 kW array installed

in 1995.<sup>36</sup> After 5.5 years, array output had dropped 3.3%, again not dissimilar to that expected from wafer-based modules. The performance of individual modules has, however, been quite unusual. Some had decreased in performance by up to 15% while others had improved by over 20%. These changes were largely the result of fill-factor changes, which suggests changes in contact properties. Rear contact problems also may have been the source of a sudden drop in output of one of the first large CdTe systems to be installed in 2002.<sup>37</sup> However, it now seems that this problem can be addressed by appropriate materials and processing selection.<sup>38</sup> Other recently installed commercial systems seem to have performed well, although available data extend over only a few years.<sup>39</sup> At least using the glass/glass encapsulation scheme, which may be required to contain the Cd in any case, it seems that reasonable stability may now be possible. Similarly to the case of a-Si, the use of tin-oxide coated glass may give rise to corrosion problems under some conditions.

Field testing of CIS modules has given mixed results. Testing on two small 8% efficient modules commenced at NREL in 1988. After 8 years in the field, these had degraded by only 10–20%, quite a reasonable result given the immaturity of the technology at that stage.<sup>40</sup> Subsequent field testing of more recent product has also given good results,<sup>41</sup> although degradation has been reported in more humid climates.<sup>42,43</sup> It appears that unencapsulated cells are unstable under exposure to moisture<sup>44,45</sup> so that modules must be designed to prevent moisture ingress. Glass–glass modules help achieve this, although less moisture-permeable non-glass backing layers are also under development.

It would appear from this survey that the best of the thin-films may be able to match the excellent durability of standard wafer-based product, or at least be able to do so in the not too distant future. The lack of more convincing data in this area has, in the author's view, acted to discount the value of thin-film technology in the past. The present expanded field-use of thin-films provides an opportunity to generate more data.

There is no intrinsic reason why thin-film cells have to be less durable than wafer-based cells. A sufficiently rugged thin-film design could, in fact, eliminate some of the weak-spots in present wafer-based modules (aging of solder joints, cracks within cells, discolouring of the polymer layer between cell and glass required to accommodate thermal expansion mismatch, etc.). Encouraging in this regard is that the CSG (Crystalline Silicon on Glass) thin-film modules have outperformed wafer-based modules in accelerated testing,<sup>40</sup> although this potentially improved durability is still to be confirmed in the field.

## 10. CONCLUSION

The recent silicon shortage has given thin-film photovoltaic technologies to establish a firm market presence. Market share is expected to grow from 6% in 2004 to 12% in 2006.<sup>1</sup>

What happens then is likely to depend on how well these thin-films perform. If all goes well, the present opportunity may allow one or more thin-film technologies to reach a critical mass, allowing the inherent cost advantages of thin-film technology to be captured.

Such a result may be good for the long-term evolution of photovoltaics. It is argued that, although there remains plenty of potential for further cost reductions,<sup>8,9</sup> wafer-based technology is unlikely to reach the very low costs (below US\$0.50/Watt) required for photovoltaics to compete on wholesale electricity markets. There is, however, a chance that a well-balanced thin-film technology could do this. The sooner that market forces are focussed on such an outcome, the more quickly photovoltaics may make large-scale impact.

The key requirements for a thin-film technology to be successful in the long-term are believed to be that it be high efficiency and be based on non-toxic, abundant and durable materials.

Improved efficiency is important, not only so thin-films are not at a disadvantage when compared to wafer-based modules, but also to reduce material costs. Non-toxicity will ensure that future progress of the technology is not impeded by what is likely to become increasingly stringent legislation relating to the use and disposal of toxic material. Abundancy is important in ensuring stable and affordable material prices. Technologies that are based on elements that are so scarce or scattered that direct mining from the ore is not feasible (at reasonable costs) are likely to be subject to a series of price shocks as increasingly expensive by-product streams are successively exhausted.



The lack of convincing long-term data on the durability of thin-films relative to wafer-based modules undoubtedly has been a former disincentive to their widespread use. The next few years will provide the opportunity for large quantities to be field-tested which may help remove the quite natural prejudice in this area.<sup>27</sup> Ultimately, thin-films could have superior durability to wafer-based technology since they have the potential to eliminate key degradation modes. Such superior durability may then allow a less expensive encapsulation scheme than required for wafer-based technology, giving cost savings additional to those usually assumed.<sup>9</sup>

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