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Silver grid finger corrosion on snail track affected PV modules - investigation on degradation products and mechanisms

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Abstract

The silver grid corrosion phenomenon of c-Si solar cells, often called “snail trails” or “snail tracks”, which occurs mainly shortly after installation of PV modules in the field, was first observed approximately one decade ago in the southern part of Europe. It was recognized quickly that these discolorations on the metal grid fingers are always accompanied by cell (micro) cracks.

From the herein presented investigations, it can be shown that several different chemical mechanisms and material involvements can lead to the visual phenomenon of silver grid finger discoloration. It must therefore be clarified that the term “snail track” (or “snail trails”) only describes a failure class and not a single degradation mechanism.

This work focuses on a bottom up approach, which includes the chemical analyses of the affected cell parts extracted from various industrial modules which have been installed in the field. In addition to the analytical investigations after field failure, the modules are exposed to artificial stresses to investigate the development, formation or further reaction of the snail track degradation products. Since the phenomenon is always accompanied by diffusion open areas in the module (cell gaps, cell cracks), the diffusion of reactant partners from the rear side of the module, e.g. atmospheric gases or additives from the back sheet material, has been the most suspected root cause from the beginning of the observation. The role of these reaction partners is therefore highlighted within the results. Four different snail track degradation products have been identified until now: Silver carbonate, silver sulfide, silver phosphate and silver acetate. Silver sulfide snail tracks were only found after damp-heat test (only artificially induced), all other types were found from field failed PV modules and respectively partially from stress tests.

The different snail track types, materials and module components considered being involved and the formation mechanisms are presented in this work.

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1. Introduction

One main challenge in terms of PV module failure analysis is the fast changes in the usage of materials and thus a transfer of results to all PV modules. Over the last years, materials changed tremendously with respect to materials' additives, fillers and quality with growing price pressure. These changes induced not only a new pricing segment, mainly for the polymer materials, but also changed the physical properties of these materials significantly. Fillers in back sheet materials e.g., which are used to lower the pricing, can have a clear impact on the permeation properties.

The silver grid corrosion phenomenon of c-Si solar cells often called "snail trails" or "snail tracks" was named according to its visual appearance. This kind of discoloration is always accompanied by the existence of a cell (micro-) crack, whereas the discoloration of the silver grid takes place along the cell crack, even mapping it. This fact triggered speculations concerning diffusing reaction partners from the rear side of the PV module already in early stages of the observation. Amongst others, mainly diffusing humidity and additives from the back sheet material seemed to play a significant role. In literature, several authors all over the world stated to have found the degradation product which should cause the discolorations. However, these results differed widely in terms of chemical composition and mechanism proposed (see chapter 3).

The impact on the performance output is negligible, since in the beginning of the degradation these effects can be stated as purely visual and surface related. However, visual defects are often listed as possible reasons for claims by PV power plant owners. In addition, it can still not be ruled out that certain types of snail tracks might progress to more severe failure, which might for instance lead to safety problems. Therefore, it is of high concern how to eliminate snail track corrosion and with this the question of which materials are involved. In the herein presented work, a bottom up approach was conducted. With the detection of the chemical composition of the degradation products and the known material combination used, degradation mechanisms can be explained. With the help of accelerated ageing stress tests, further aspects which are used for mechanism determination have been revealed.

2. Experimental

2.1. Selection of modules and further artificial stress

The following work focuses on the investigation on snail track affected c-Si PV modules, which have been exposed in field locations in different climates and climate zones for different time scales. A number of modules of each "bill of material" (BOM) are analyzed by means of the analytical methods described in section 2.4. The type of sample preparation is always destructive, so that only non-analyzed modules are further investigated by artificial stress. The following stress tests are employed: UV light exposure using fluorescence tubes with a UV-A to UV-B ratio according to standard IEC 61215 [1] and Damp-Heat testing (85 °C, 85 % RH) also according to standard IEC 61215 [1].

2.2. Visual inspection

The modules' appearance is investigated by a detailed visual inspection. Each single cell is therefore photographed with the same distance and resolution with respect to the camera gaining comparable images of the cells within the modules before and after stress testing.

2.3. Preparation of samples

The extraction of samples (cell parts with grid finger and encapsulation parts) of the affected cells is performed mechanically without any chemicals involved. Cell parts with diameters of about 1-2 cm with intact grid finger structures are extracted from the full-size PV modules. The corresponding ethylene vinyl acetate (EVA) encapsulation material can also be extracted separately with some further preparation steps to identify if the discoloration is also part of the encapsulation material or only seen as a grid finger surface discoloration.

2.4. Analytical and experimental methods

Different analytical methods are applied for the characterization of the snail track corrosion products. For the Energy Dispersive X-ray (EDX) spectroscopy measurements, the extracted cell parts are mounted by graphite pads on the sample carriers. The EDX measurements are carried out at an Environmental Scanning Electron Microscope ESEM (FEI Quanta 400) with integrated EDX spectroscopy system (EDAX Genesis).

The Raman spectroscopic measurements are performed at a WiTec Alpha 500 Raman microscope. The excitation source is a laser with a wavelength of 532 nm, which is obtained using a frequency doubled Nd:YAG laser with a maximum excitation power of around 17 mW. As the majority of silver salts are strongly light sensible, the measurements have to be performed with lower laser intensities. A CCD detector, thermoelectrically cooled to -67 °C, was employed in the measurement set-up. The confocal microscope enables the use of different objectives with different magnifications.

The identification of phases is facilitated by the measurements of reference products. Two of these silver salts, silver carbonate and silver acetate, were obtained by the reaction of silver nitrate and the corresponding potassium salt as counter anion. The references for silver phosphate, silver sulfide and silver oxide Raman measurements are obtained by commercially available products. The results of these measurements are shown in the graphs in section 3.1 as bar diagrams.

3. Literature review

Several publications reported findings on snail track product in the last years which differed in the applied analytical methods as well as in the compositions of products found. The following section will give a literature review about some of the most popular theses.

Peng *et al.* showed in a study from 2012 the existence of silver carbonate looking at snail track affected cells [2]. The degradation product was investigated by EDX and Raman spectroscopy. A degradation mechanism of diffused oxygen, carbon dioxide, humidity and silver together with UV irradiation is proposed. In addition, they investigated the accelerating factor by the silicon material in terms of micropore arrays.

Chou *et al.* also detected silver carbonate in the snail track affected area of the EVA material by XPS measurements accompanied with an organic compound, acetone, identified exclusively in the affected areas in the EVA by Py-GC/MS analysis [3]. An experiment revealed silver as a main factor for the discoloration in the EVA material. From the observations a degradation mechanism is proposed; organic components from EVA material and silver react to form silver carbonate after some time in outdoor exposure. In addition, the statement is given that neither a potential nor a current flow is responsible or needed for snail track formation.

Several papers are published by Meyer *et al.* showing different analytical approaches united by the finding of silver nanoparticles inside the EVA material [4,5,6]. The papers focus mainly on the EVA material and investigated the degradation products in terms of SEM, TEM, EDX, ToF-SIMS or XPS measurements. In [4], Meyer *et al.* showed that the detected silver nanoparticles, which are stated to origin the discoloration of the snail track phenomenon, are present in the polymer film above the discolored area. Experiments on different combinations of back sheet and encapsulation materials with varying additive compositions, detected by laser ablation ICP-MS, investigate their contribution to the discoloration. Especially the phosphorus-containing additives are regarded as critical to the reaction mechanism, because they enable a redox mechanism to form the silver nanoparticles. In addition, the significance of humidity inside the PV module is discussed. In [5], experiments of mini-modules exposed to Damp-Heat condition with additional current flow are shown. In the paper, it is stated that “preliminary results on the phosphorus content of the foils are in good correlation to the formation of snail trails”. The investigations in [6] focus on field exposed modules and mini-modules after Damp-Heat testing. In addition, experiments with the sole encapsulation material and a silver nitrate solution are introduced.

Several publications focus on the impact of snail tracks on the performance of affected PV modules, as e.g. Dolara *et al.*, which mostly resulted in the statement that “the snail trails phenomenon can indicate cell cracks on the PV module: Therefore, it can be considered as a serious defect for PV modules” [7].

First results from our investigations showed already in 2013 (Philipp *et al.*) that there must be more than one snail track degradation product due to different behavior to artificial stress from field failed modules [8].

4. Results

4.1. General aspects and visual appearance

The formation of snail tracks is strongly related to diffusion processes into and inside the PV module. The formation of the detected silver salts, which are responsible for the silver grid finger discoloration, is therefore dependent on diffused reactants such as environmental gases or other molecules coming from the rear side of the module e.g. from the highly breathable, polymeric back sheet. In addition, reaction stress factors as e.g. UV light or temperature are required for a reaction mechanism, as they function like an activator in the reaction and/or as a root cause (photochemical degradation of EVA).

The thesis of diffusing reactants is supported by a second phenomenon which affects the silver grid fingers at the cell edges and is named “framing” due to its visual appearance (see Fig 1b + c). For field failed PV modules, it is often observed that the degradation products of framing and snail tracks differ or that no framing is observed even when snail tracks are present. Therefore, one of the most interesting tasks next to finding the root causes of the single degradation products is to investigate the difference between framing and snail tracks in terms of reaction conditions.

The main differences between the degradation of the cell edge metallization and the cell metallization near a cell crack are:

- Framing can possibly appear already from beginning of exposure, while cell cracks are mostly formed in the aftermath due to environmental stresses (if not process-induced)
- Wider gap between the cells in comparison to (micro-) cracks,
- Cell edges are electrically insulated in comparison to induced cell cracks,
- Temperature effects of cell cracks in terms of shunting,
- Electrical potential of shunts, non-insulated cell cracks,
- Dependence of electrical potential to acidity level (pH), smaller gaps of cracks lead to decreased diffusivity of acetic acid (see first fact).

Snail tracks can strongly differ in terms of visual appearance in the PV module. In addition to the differentiation done before (framing vs. snail tracks, see section 4.1), also the single snail track phenomena can show different characteristics. Snail tracks occur single lined (see Fig 1a), double lined, worm-like (see Fig 1b) or even more extensively (see Fig 1c).

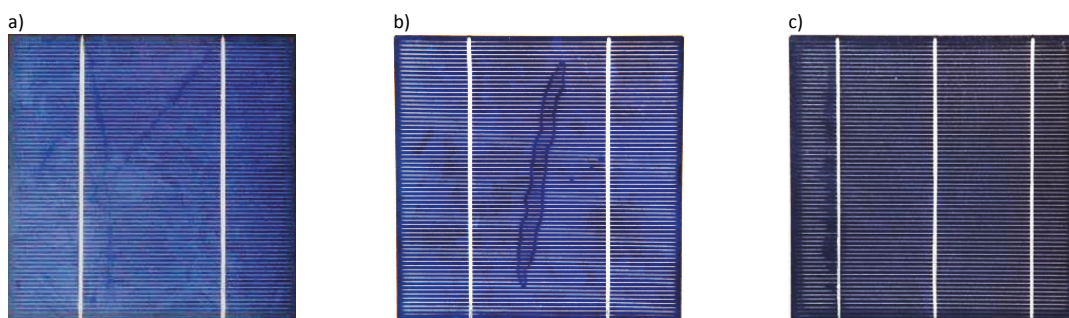


Fig 1 Visually different snail track discolorations; (a) single lined, (b) double lined and (c) extensive discoloration.

As described in section 2.3., a destructive sample preparation has to be done in order to get cell samples for the subsequent degradation product determination. After preparation, an additional differentiation of visual appearance of the single snail track types could be observed (see Fig 2). For two types of snail tracks (a and d), the silver grid fingers show a strong discoloration while the corresponding EVA material from the front of the cell does not show any discoloration on top or inside the material. The other two types of snail tracks showed a completely different behavior; both the silver grid finger and the EVA material show strong discoloration (see Fig 2b and c).

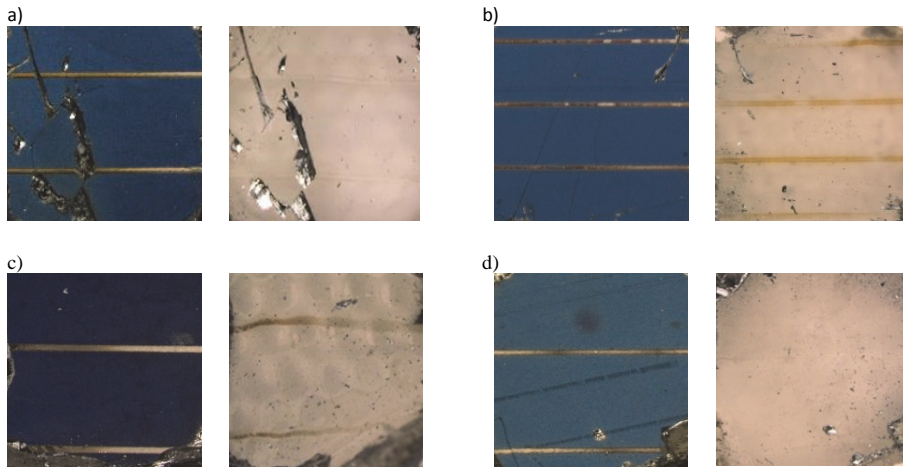


Fig 2 Cell and EVA sample from module affected with (a) silver carbonate and (b) silver acetate, (c) silver phosphate and (d) silver sulfide snail tracks.

The discoloration which can be seen on the silver grid finger is mainly contributed by small silver salt precipitates on the grid finger surface, which is exemplarily visualized for a silver acetate snail track type by Scanning Electron Microscopy (SEM) in Fig 3b. In addition, the degraded surface shows a significantly increased surface roughness in comparison to the reference grid finger surface (see SEM image of reference grid finger surface Fig 3a), which contributes to the visual discoloration as well.

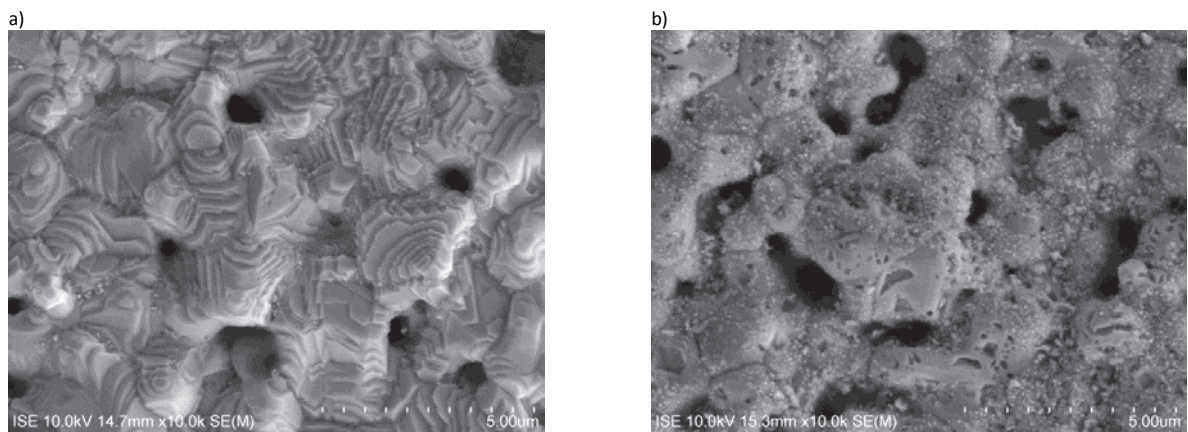


Fig 3 SEM image of reference (a) and snail track affected (b) silver metallization grid finger surface.

4.2. Snail track product A - Silver carbonate

Referring to Fig 2, on the cell sample (a) silver carbonate was detected on the discolored silver grid finger. The Raman measurement together with the bands measured from the reference material of silver carbonate is shown in Fig 4. In addition to silver carbonate (Ag_2CO_3), silver hydroxide (AgOH) may be detected as a second phase, which indicates the formation reaction reported in literature [9], resulting in a basic silver carbonate.

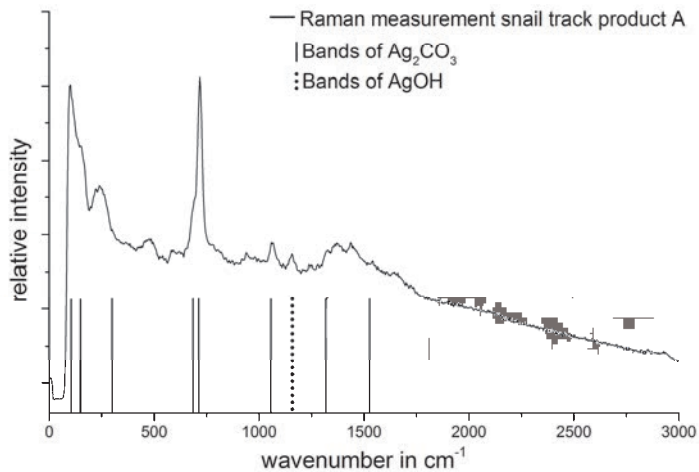


Fig 4 Raman spectrum of degradation product A at low laser intensity with the main vibrations assigned and the bands given from the silver carbonate reference sample.

The finding that silver carbonate is formed under basic conditions supports the presumption that carbon dioxide (CO_2) as a reaction partner is mainly present due to permeation from the rear side of the module (from atmospheric gas). CO_2 may also be present due to a photochemical degradation of EVA, which is described in the so-called Norrish reactions (type I). However, this would most likely also result in the formation of acetic acid (Norrish reaction type II), which can clearly be seen contradictory to the detection of the basic silver carbonate degradation product.

Wiesinger *et al.* reported a reaction mechanism of silver and carbon dioxide under the formation of basic silver carbonate ' $\text{Ag}(\text{OH})\text{Ag}_2\text{CO}_3$ ', which was observed under a certain amount of humidity and UV irradiation, whereas UV irradiation is stated to be the main acceleration factor in the reaction investigated. Without the presence of UV irradiation, the basic silver carbonate dissolves after a short time in the moisture film on the silver surface [9]. This finding supports our herein presented results perfectly, since our investigations showed disappearing (dissolving) silver carbonate snail tracks under Damp-Heat conditions, which include high humidity and high temperatures, but no UV irradiation. The UV irradiation seems to play the crucial factor for this reaction. It can be stated that there is a need for an activation of the silver grid finger, which could be realized by the UV irradiation together with oxygen forming a reactive silver surface due to the interaction of adsorbed oxygen on the silver surface or the formation of small amounts of ozone, which could oxidize the silver surface.

With this knowledge of the reaction partners, silver, carbon dioxide, moisture and UV irradiation, the essential question, why isn't every module or at least more modules affected, arises. The answer could lay in the reaction mechanism itself. In addition to the Raman spectroscopic measurements, energy dispersive X-ray (EDX) spectroscopic measurements are performed. Zinc (Zn) was found all over the affected cell metallization, also on the non-affected cell parts, when silver carbonate was detected on the discolored cell areas (see Fig 5). This has also been observed by Peng *et al.*, who also detected zinc oxide (ZnO) accompanying the silver carbonate snail track product [2].

A good explanation for the ZnO -supported reaction mechanism of silver carbonate is found by Freund *et al.* [10], where a metal oxide supported adsorption of the carbon dioxide on the silver surface is detected. ZnO is a common glass additive used in most of the standard screen printing metallization pastes for c-Si cells, besides other metal oxides for the contact formation as e.g. lead oxide. Normally, the metal oxide containing glass additive melts during the firing process of the screen printed paste accompanied by a reduction of the metal oxides caused by the reductive atmosphere.

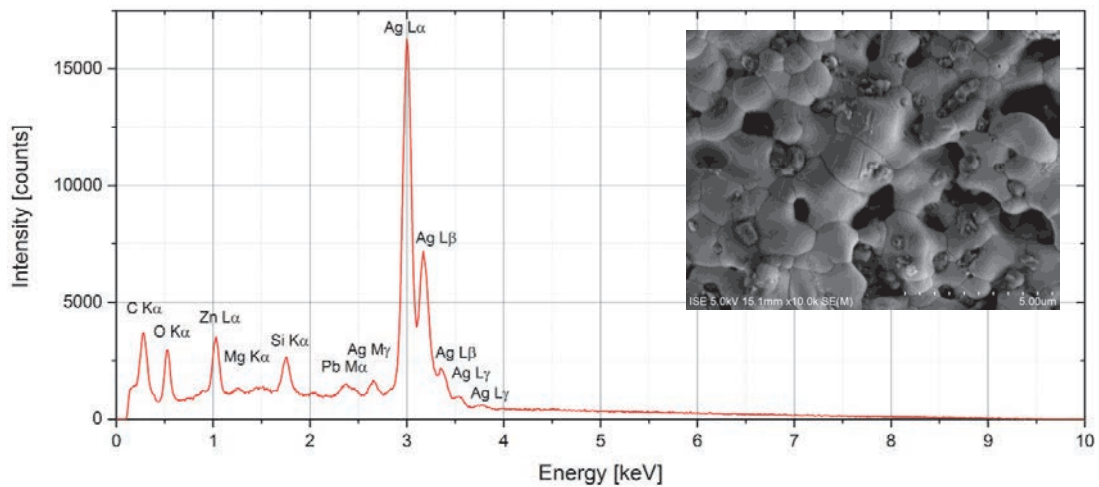


Fig 5 EDX measurement and SEM image of silver carbonate affected cell showing zinc carbon, oxygen, lead, silicon, magnesium and silver.

The theory of the present ZnO supporting the CO₂ adsorption on the silver grid finger, and therefore coordinating the carbonate together with the silver, could additionally explain the slight shifts in the Raman bands in comparison to the bands of the reference, Ag₂CO₃, spectrum.

Often this kind of degradation product (Ag₂CO₃) is only found for snail tracks. The question why no framing is formed or why the snail tracks do not evolve further can only be speculated. But since this kind of degradation product is formed in the first part of the outdoor exposure, it seems like the ZnO reacts further to be inactive for CO₂ adsorption after a definitive time and temperature or electrically induced effects further support the reaction.

In Fig 2a it is shown that the EVA material above the silver carbonate affected cells does not show any discoloration. This indicates that the encapsulation material is not directly correlated to this kind of snail track and therefore this effect can be observed for all types of encapsulation (not limited to EVA material). The reaction forming silver carbonate snail tracks is dependent on the metallization composition of the grid finger surface (presence of ZnO), the carbon dioxide and water vapor diffusion property of the back sheet material and the presence of UV irradiation.

4.3. Snail track product B - Silver sulfide

Referring to Fig 2 on the sample (d), silver sulfide (Ag₂S) was detected on the discolored silver grid finger. Silver sulfide as a snail track degradation product was so far only observed from accelerated aging tests including high temperatures and high humidity (as e.g. Damp-Heat test 85 °C and 85 % RH, according to IEC 61215 [1]). The formation and reproduction of this type of snail track product is, however, simple. Laminates including a back sheet material with a high amount of sulfur-containing additives show framing or snail tracks, if cracks are present, already after approx. 1000 h of Damp-Heat testing.

These organosulfur additives are mainly used as secondary antioxidants, which are needed for processing and/or further stabilization of the back sheet material, together with hindered amine additives, during thermolysis up to 100 °C – 150 °C. At high temperatures, sulfur oxide or organosulfur acids are formed [11].

As sulfur-containing secondary antioxidants the usage of different compounds, such as thiols, thioesters or thioethers are reported [12,13,14]. Thioesters may form thiols by hydrolysis, while thioethers show a certain inertness. The present or formed thiols are able to react with silver to silver thiolates. A further reaction to silver sulfide under the hot and humid conditions is possible and the most probable explanation for the detected degradation product.

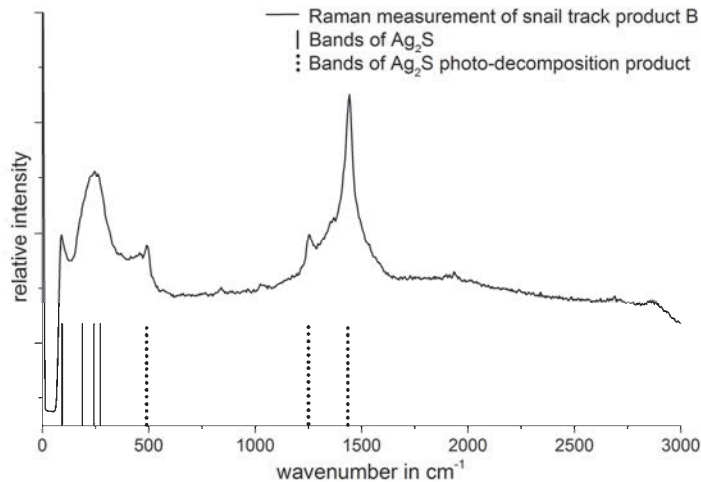


Fig 6 Raman spectrum of degradation product B at low laser intensity with the main vibrations assigned and the bands given from the silver sulfide and its photo-decomposition product.

The Raman spectrum of the degradation product measured on the affected silver grid finger, the reference bands of silver sulfide and additional bands of a silver sulfide decomposition product [15] are shown in Fig 6. It shows that the main vibrations assigned to the degradation product silver sulfide, measured at low laser intensity, are in good agreement with the bands given from the silver sulfide reference and its photo-decomposition product.

From atmospheric silver corrosion (Ag_2S), an acceleration factor by oxygen is known [16]. The influence of oxygen on the silver sulfide snail track degradation product is also observable. Back sheet materials with higher oxygen transmission rates do show a faster formation of silver sulfide snail tracks in experiments.

To summarize it can be stated that the formation of silver sulfide as a snail track product depends on the additive composition of the back sheet material. In addition, the oxygen and water vapor diffusion property of the back sheet material plays a significant role. Humid and high temperature conditions seem to be necessary to induce the reaction. The extracted encapsulation material from the top of the affected cells does not show any discoloration (see Fig 2d). Therefore, it can be concluded that it does not directly contribute to the reaction which forms silver sulfide, although some encapsulation materials may include sulfur-containing additives, but in much lower concentration.

4.4. Snail track product C - Silver phosphate

Referring to Fig 2, on the sample (c) silver phosphate was detected on the discolored silver grid finger. The Raman spectroscopic measurement of the degradation product found on the affected silver grid finger and the corresponding reference peaks (data from [15]) can be seen in Fig 8. Two different sets of reference bands and therefore degradation products could match. On the one hand silver phosphate (Ag_3PO_4) and on the other hand silver oxide (Ag_2O) (see Fig 8) [9,15,17]. EDX measurements on the affected silver grid finger showed the presence of phosphorus (P) (see Fig 7), so that we conclude that silver phosphate is the right assignment to this snail track type.

From literature, it is known that phosphorus-containing additives (phosphite or phosphonite compounds) used as secondary antioxidants are often utilized as stabilizer against peroxide degradation in encapsulation materials. In contrast to the sulfur-containing antioxidants discussed before, these additives are used mainly for process stabilization. Therefore, the influence on long-term stability is limited, since the hydrolysis or oxidation reactions easily transform the compounds into the corresponding acids or phosphates [11]. These phosphates are then likely to form silver phosphate on the silver grid finger when oxygen and humidity is present.

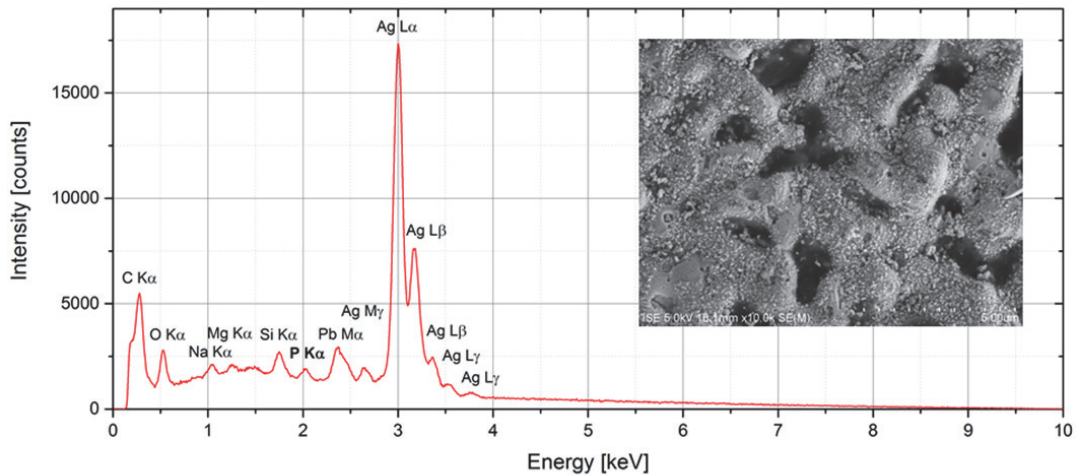


Fig 7 EDX measurement and SEM image of silver phosphate affected cell phosphorus, carbon, lead, silicon, silver and sodium, magnesium and aluminium most likely as counter ions.

Meyer *et al.* also discussed the contribution of phosphite additives used for encapsulation material in terms of snail track formation. In contrast to our findings of silver phosphate on the affected cell as degradation product, it is stated that the phosphite additives react over a silica phosphate transition compound to form phosphates after oxidation by silver ions, which are themselves reacted to silver nanoparticles in the EVA material [7].

Looking at the extracted cell and EVA samples from silver phosphate snail track affected PV modules, it can be seen that the EVA material from the top of the affected grid fingers shows significant discoloration (see Fig 2c). This supports our thesis well that the EVA material or every other encapsulation material including phosphorus-containing antioxidants directly contribute to this reaction and therefore show also a discoloration.

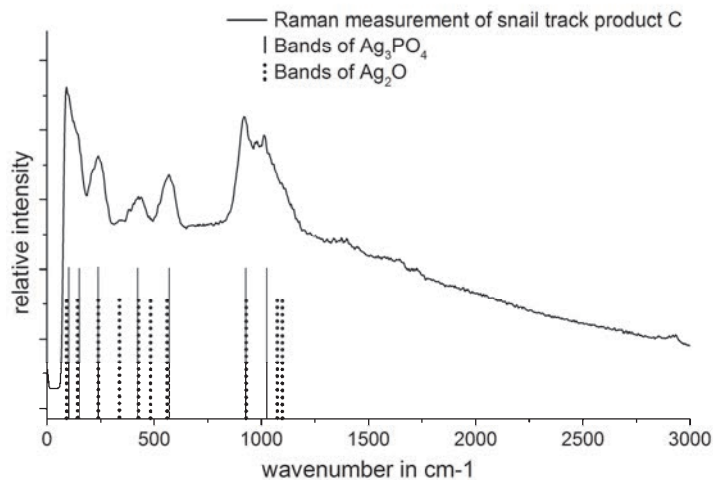


Fig 8 Raman spectrum of degradation product C at low laser intensity with the main vibrations assigned and the bands given from the silver phosphate or silver oxide reference spectra.

4.5. Snail track product D - Silver acetate

Referring to Fig 2, on the sample (b) silver acetate was detected on the discolored silver grid finger. The Raman measurements of the snail track affected cell is shown together with the bar diagrams of the silver acetate ($\text{Ag}_2(\text{CH}_3\text{COO})_2$), which is abbreviated in the following as Ag_2Ac_2 (“Ac” = Acetate CH_3COO^-), reference measurement in Fig 9.

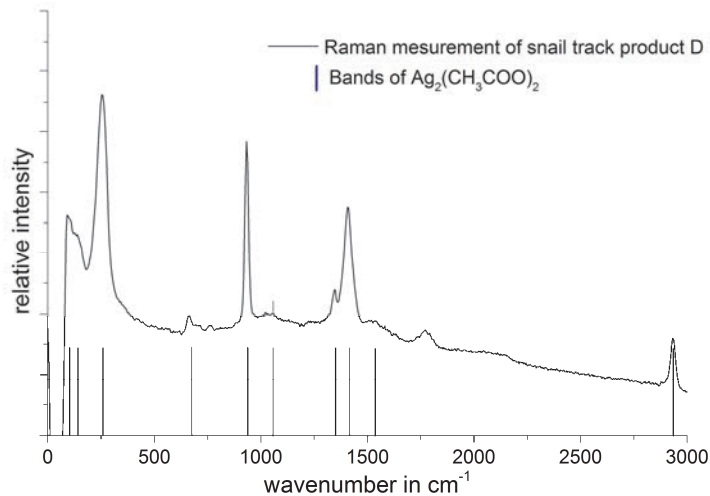


Fig 9 Raman spectrum of degradation product D at low laser intensity with the main vibrations assigned and the bands given from the silver acetate reference sample.

The formation of silver acetate snail tracks on already affected PV modules from the field could be reproduced by exposure to UV irradiation after the induction of new cell cracks. Silver acetate must therefore be formed by the reaction of silver with acetic acid, which is a photochemical degradation product of EVA by the Norrish reaction II. The UV irradiation is most likely not only the source for acetic acid, but also an important activator for the reaction together with oxygen as described for the silver carbonate formation (see section 0). This is additionally supported by the observation that silver acetate snail tracks are most often found for back sheets with high oxygen transmission rates (OTR).

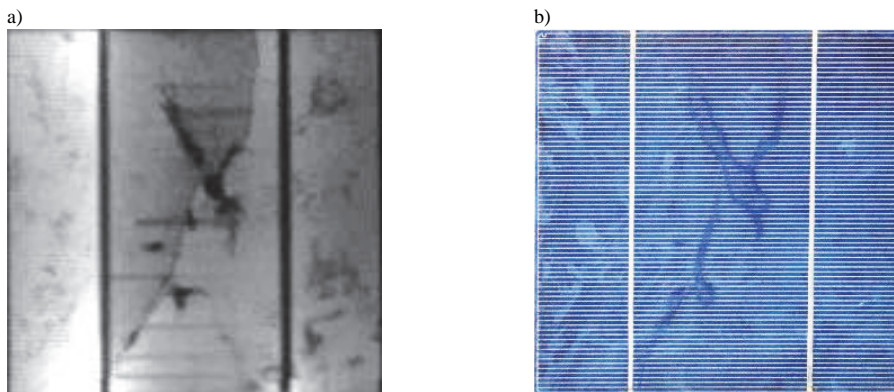


Fig 10 After the induction of new cell cracks and UV testing: EL image (a) and silver acetate snail track affected cell (b)

The fact that silver acetate is most often seen for snail tracks and less frequently observed for the framing phenomenon indicates the fragile reaction equilibrium which is needed for this reaction. In the area of the cell edges, acetic acid is able to diffuse out of the module because of the big cell gaps in comparison to small micro-cracks,

which hinder the acetic acid out-diffusion. Additionally, oxygen must be present in a defined concentration to activate, together with the UV irradiation, the silver grid finger surface.

Fig 10 supports this assumption in an imposing way. The electroluminescence image shows the characteristics of the cell crack (wider cell crack or several cell cracks in parallel = darker, thicker black line). In the area where the cell crack is wider, acetic acid may diffuse out of the module; therefore no silver acetate formation is observable directly on wide cell crack parts. But in some distance to this wider cell cracks, where most likely both the acetic acid formation and the oxygen diffusion from the rear side of the module is still sufficient, the reaction takes place. Therefore, the worm-like, double lined snail tracks appear.

5. Summary and conclusion

The herein presented work demonstrates the existence of at least four different degradation products which are found in affected PV modules which show the degradation phenomenon called snail track. It therefore can be shown that not only 'the one' degradation mechanism, but four different types of snail track degradation products exist and thus different reaction partners, PV module components and stress factors are involved. It must therefore be clarified that the term "snail track" only describes a failure class.

Silver carbonate, silver phosphate and silver acetate have been found in the investigation of field failed PV modules from different module manufacturers by Raman spectroscopy. EDX spectroscopy was additionally performed to support the findings. Silver acetate is reported the first time as a snail track degradation product. New silver acetate snail tracks have been produced by exposure to UV irradiation of the field failed modules with the silver acetate snail track type. Silver sulfide was only observed from artificial stress (Damp-Heat condition) and not in field exposed modules so far.

To prevent snail track degradation, it is inevitable to optimize materials and material combinations which are prone for snail track formation. The encapsulation material plays a significant role for the formation of silver acetate (formation of acetic acid) and of silver phosphate (additive composition). The formation of silver sulfide and silver carbonate seems to be independent from the used encapsulation material.

The back sheet material with its permeation properties is most essential for all types of snail track products. For silver carbonate the carbon dioxide, water vapor and most likely oxygen transmission rates are crucial. For silver acetate as snail track degradation product the oxygen transmission rate most likely influences the formation of snail tracks. For silver phosphate and silver acetate mainly the water vapor transmission rate should be analyzed in terms of reaction partner (hydrolysis) or diffusion supporting agent.

The cell metallization seems to play an important role only for the silver carbonate formation, since it is assumed that zinc oxide of the glass additive supports the carbon dioxide reaction with the silver surface.

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