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Analysis of back contact layers for flexible CdTe/CdS photovoltaic structures

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ABSTRACT

The article shows the comprehensive results of the experiments, conducted in order to select the appropriate metal layers, for back absorber contacts, to apply in flexible, thin-film photovoltaic cells based on cadmium telluride. Preliminary selection of investigated materials was made on the basis of general knowledge and physical data. Deposition techniques, as well as layer parameters, were adjusted to the specific flexible solar cell's needs. Selected metal layers were deposited either by physical vapour deposition (PVD) or screen-printing method (SP) and tested in terms of their flexibility, thermal resistance, as well as adhesion to CdTe layer. Practical verification of selected configurations is proved by the complete construction of the device.

1. Introduction

Base contacts in photovoltaic (PV) structures are made as metal layers deposited generally on the whole back area of the device. Such construction offers low series resistance and it has a function of mirror for photons, which were not absorbed in the total volume of the structure. This allows reusing their energy in the photoconversion process. Elaboration of the proper base contacts for thin film, flexible solar cells are a significant and complex technological problem, which cannot be solved by direct transferring of the processes established for rigid cells. In the photovoltaic cell construction, base contact layers are placed directly between the substrate from one side and semiconductor structure from the other. This means that research on the properties of these layers, as elements of solar cells, should be conducted in specified conditions, taking into consideration the influence of the interface layers' connections on the contact quality. In this case, polyimide foils have been used as substrates for an investigated CdTe/CdS semiconductor PV structures.

The challenge to produce flexible base contacts for CdTe/CdS solar cells is also related to the specific material properties of cadmium telluride layer, which has a crucial contribution to the generation of photocurrent in the PV structure [1]. The contact must be formed properly to the low doped p-type material, with relatively wide bandgap and high work function.

During the manufacturing of the ohmic contacts for CdTe-based solar cells, there are two general types of technological problems. The first concerns the requirement for metal used for the process, which should have a work function value higher than the sum of the electron affinity and the energy bandgap of the semiconductor base. Such formulated requirements are intended to place the top of the valence band on the Fermi level of contact metal, which allows to avoid blocking of the carriers' flow between semiconductor and metal contact layer. Taking into account the parameters of cadmium telluride, this criterion is not easy to meet. The electron affinity of CdTe is equal to 4.3 eV [2] at the energy bandgap of 1.45 eV, which determines the value of the work function of the metal contact at the level above 5.7 eV [3]. This condition is therefore practically impossible to meet, which leads to the formation of the blocking junction in the vicinity of the close-contact layer. Consequently, this is a requirement for formation of the high-doped p + layer on the back surface of the semiconductor, near the back contact layer. This is a second problem which may be described as a condition for achieving a suitable quality of base contact. Introduction of high doped p + zone leads to narrowing of the Shottky junction, which is being created inevitably in the vicinity of the back contact. This consequently allows tunnelling of holes in its direction. Nevertheless, high doping level is difficult to achieve due to the compensation of the dopants which occurs in CdTe. These technological difficulties have been widely described in the literature [4]. In order to increase the doping level of the base region, several layer's treatments are being implemented, such as introducing various types of dopants, e.g., Sb, Te or Zn-Te to the base region or forming the intermediate layers made of $Cd_xHg_{1-x}Te$ compositions [5]. However, the solution which is most frequently applied, is cop-

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Table 1
Parameters of selected metals for flexible back contacts.

Metal	Molybdenum (Mo)	Silver (Ag)	Copper (Cu)
Melting temperature (°C)	2623	962	1085
CTE (ppm/°C)	4.8	18.9	16.5
Resistivity (nΩ·m)	53.4	16.1	17.1
Work function (eV)	4.36 – 4.95	4.52–4.74	4.53–5.10
Density (g/cm³)	10.25	10.50	8.96

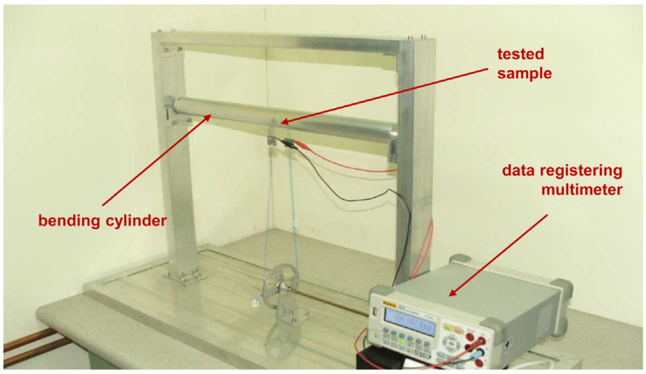

Fig. 1. Measuring system for testing of the material resistance to dynamic bending.

per doping [6]. Although also this approach causes some difficulties as dopant inter-diffusion into the PV structure, which occurs alongside the grain boundaries of CdTe [7]. Copper doping, in case of a deep diffusion, can positively influence the quality of the CdTe/CdS heterojunction, unless it causes the internal short circuit in the PV cell structure, and as a result, abrupt reduction of its parallel resistance value. Atoms of Cu are usually introduced into the cell structure as base contact dopants which diffuse inside the base region during the heating process.

In terms of the restrictions described above, the problem of the base contacts for CdTe PV cells was considered at several research centers, resulting in different solutions using the materials or compounds such as Mo [8], Au [9,10], Cu/Au [11] and Ag, HgTe, ZnTe: Cu and Cu_2Te [12,13]. In order to improve the quality of the base contact layers, among others, D. Bonnet [14] proposed the introduction of an intermediate layer. In this role, an additional non-stoichiometric cadmium telluride region with the predominance of Te can be used, as well as a buffer layer made of the compositions based on carbon, such as graphite.

2. Investigated materials and methods

In further research, three materials have been deposited on polyimide foils to be verified in the function of CdTe/CdS flexible base contacts. Copper, molybdenum and silver have been chosen. Table 1 presents their basic physical properties.

Investigated samples, deposited on flexible polymer substrates, have been tested in terms of their electromechanical strength. These tests included the influence of dynamic bending on samples' electrical resistivity. The measuring equipment for this purpose is shown in Fig. 1. It enables conducting durability tests according to the A-De Mattia method and consistent with the PN-EN ISO 7854 Standard [15], which defines the conditions of bending resistance determination.

Mechanical durability measurements have been conducted as 200 bending cycles (one cycle: flexion and straightness) of each sample. Cylinder diameter was set as 25 mm at a constant tension of the sample. During these tests the resistance of the sample was monitored and recorded automatically by the Digital Rigol multimeter.

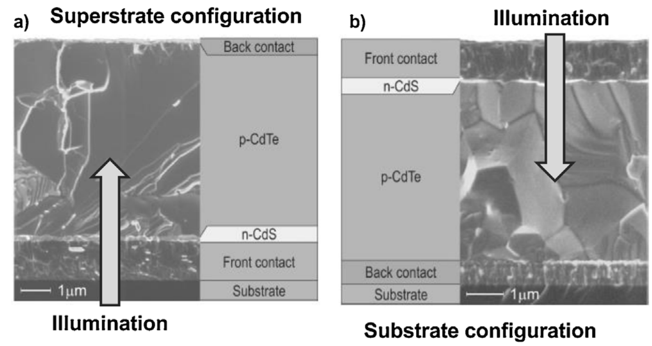

Fig. 2. SEM micrographs and schematics of the cross-section of CdTe/CdS solar cell a) in superstrate and b) substrate configurations [16].

3. Obtained results

Metal layers have been deposited using two different techniques, namely PVD evaporation and screen printing. Selected layers have been used for manufacturing of flexible CdTe/CdS solar cells in substrate or superstrate configuration. In the superstrate configuration of CdTe/CdS structures, the subsequent layers are grown onto the transparent substrates (such as glass or high-temperature polymer), while for substrate configuration metal foils or metal coated glass or polymers are used. Figure 2 shows the schematic images of these two possible configurations.

3.1. *Thin copper layers*

The first considered material for flexible CdTe/CdS structure back contact was copper (Cu). It is an easily available metal of $17.12 \cdot 10^{-9}$ Ω·m resistivity and relatively high work function in the range of 4.53 to 5.10 eV [17], depending on the atomic configuration in the material's surface. These properties make it possible to obtain the proper contacts to the CdTe layer, which have been investigated in the experiments with standard CdTe/CdS cells [18]. Copper is also a material with very good mechanical properties and high corrosion resistance which combined with good thermal conductivity makes it a serious candidate for using in flexible photovoltaic structures based on CdTe.

As it was already mentioned, copper is also used as a component of the base contacts' intermediate layer in CdTe/CdS solar cells because of the acceptor character of its dopants in a CdTe layer. There is a copper diffusion inside the CdTe base layer which takes place in the annealing process of the PV structure with graphite contact enriched with Cu. Through this exertions p + type layer is formed in the function of Back Side Field (BSF). The level of the obtained surface barrier is dependent on the concentration of introduced dopants. Thus, by using a complex contact construction, one can have a better control on the diffusion depth of copper, which always proceeds unevenly in the polycrystalline structure, with the risk of a short circuit of the p-n junction.

There are also reports [19] indicating an additional, very important advantage of the use of copper contacts in flexible cells CdTe/CdS. It concerns the protection of emitter structures made of CdS within cells in a superstrate configuration. Test investigations for PV cells on glass substrates show that the copper additive in the CdTe base reduces the phenomenon of tellurium agglomerates formations, thereby decreases the influence of the layer's non-stoichiometry on its optoelectronic parameters. This phenomenon allows to lower the temperature of the deposition and recrystallization processes of CdTe layers which is extremely important for the implementation of photovoltaic structures on polymer substrates. Hitherto, one of the main undesired effects occurring during such attempts was non-stoichiometry of obtained layers, which,

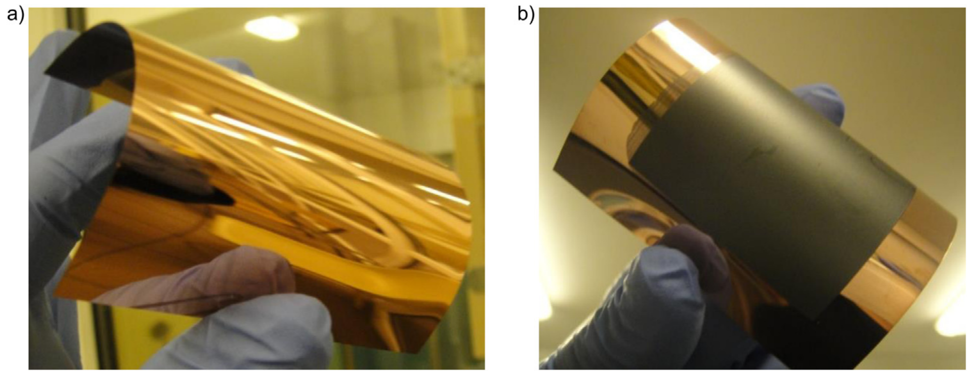

Fig. 3. Pictures of back contact layers' samples: a) copper, b) copper with graphite.

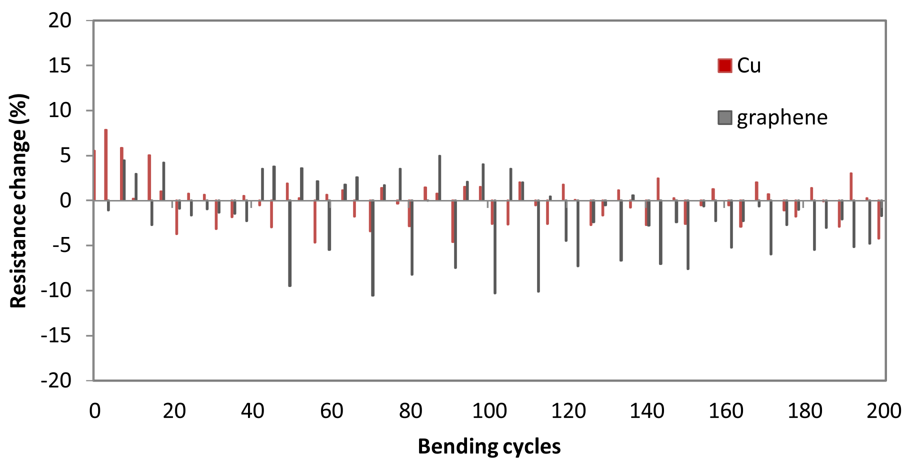

Fig. 4. Percentage change of the electrical resistance during bending on 25 mm diameter cylinder for Cu and graphite layers.

together with the small size of grains in polycrystalline CdTe/CdS structure, influenced in a significant way on the photoconversion efficiency reduction.

In order to verify the practical possibilities of CdTe/CdS photovoltaic structures' production on flexible substrates using copper contact base, relevant experiments have been conducted. In the first stage, only back contact layers have been deposited on flexible substrates. Copper films were obtained by physical vapour deposition (PVD) method using a laboratory grade Cu of 99.99% (in pellet form) evaporated in vacuum with no substrate heating. One part of the samples was additionally equipped with the screen-printed graphite layer as a buffer between the CdTe semiconductor and Cu metal, in order to improve the ohmic contact. For graphite layers' deposition, a commercial screen-printable paste has been used. Samples were prepared in two variants, as follows:

- Upilex-S foil (by UBE America) of 75 μm thickness– Cu layer of 200 nm thickness deposited in PVD process;
- Upilex-S foil (by UBE America) of 75 μm thickness– Cu film of 200 nm thickness deposited in PVD process and screen-printed graphite layer of 2 μm thickness.

Figure 3 shows the pictures of samples in two version described: a) evaporated copper and b) evaporated copper with additional, printed graphite layer. The surface resistance of investigated layers on these samples was on the average approx. 0.1 Ω/square for copper and approx. 40 Ω/square for graphite.

All samples have been tested in terms of electromechanical resistance using A-De Mattia method according to the PN-EN ISO 7854 [15] standard. Figure 4presents the results of measurements demonstrating the percentage change of the electrical resistance during bending.

As shown in Fig. 4, the resistance change is negligible, especially considering copper layer. The resistance of both samples is slightly increasing and decreasing during the process which is caused by the tension forces in dynamic bending and the structure of the material.

Both copper and graphite layers are fully resistant to mechanical stress, which allows using them in flexible PV cells constructions. Therefore, further technological steps for manufacturing CdTe/CdS cells were undertaken.

A series of experiments have been conducted including the deposition of CdTe base active layers, as a technological step of the PV cell manufacturing process. Cadmium telluride layers, may be deposited by several methods, including PVD, chemical bath deposition (CBD), chemical vapor deposition (CVD), with the variant of plasma enhanced CVD (PECVD), atmospheric pressure chemical vapor deposition (APCVD) and most importantly metal organic chemical vapor deposition (MECVD) [24]. Very important role in the present industry manufacturing processes of CdTe deposition plays the close space sublimation (CSS), which in the variant of isothermal close-space vapor transport process (ICSVT) was extensively tested by the authors, and described elsewhere [25]. For the current small-scale experiment though, the PVD technique was selected as the simple, fast and reliable technique with high compatibility with the contact deposition processes.

Cadmium telluride layers have been deposited in a PVD vacuum evaporation process on two sample types including copper, which are described above. For these samples, processes required to form the structures of photovoltaic CdTe/CdS cells in a substrate configuration have been implemented. In the first step of

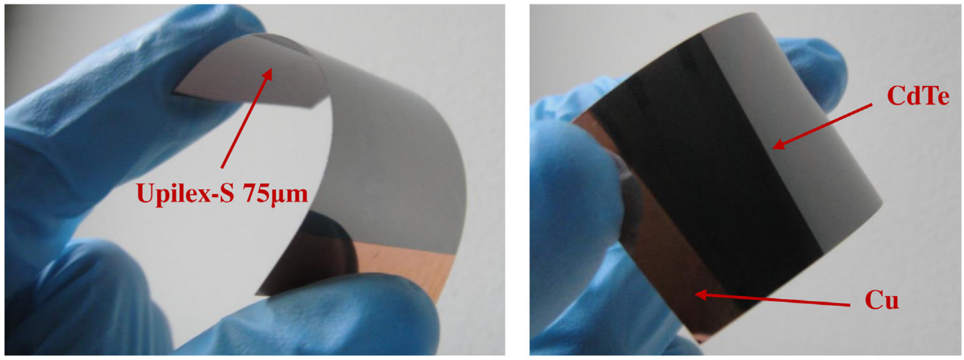

Fig. 5. The CdTe base deposited on Cu contact layer obtained on the flexible polyimide Upilex-S foil.

these investigations, CdTe base layers have been deposited. For this experiment, Sigma-Aldrich powdered cadmium telluride of the 99.99% purity has been used as a target. The semiconductor base material has been evaporated in a vacuum PVD method using special closed tantalum R.D.Mathis® sources. The PVD process was conducted in the same conditions for all tested samples. For increasing the level of adhesion of semiconductor material to the substrate, the samples were placed on a heated carrier. The temperature of substrates' structures during the deposition of CdTe was at the average level of 350 °C. Basing on the other authors work [11,20–23] and own experience, this temperature was accepted as an optimal. Samples deposited on polyimide Upilex-S foils were characterized by a good adhesion of CdTe layer both for copper and copper-graphite layers. Obtained layers had high flexibility allowing bending of the structure without the risk of the delamination or disintegration. Moreover, deposited CdTe layers showed high homogeneity over its entire surface. Figure 5 presents the sample flexible structure composed of the polyimide Upilex-S foil as a substrate and evaporated Cu and CdTe layers.

Typical further technological step of the flexible CdTe/CdS manufacturing process is high temperature recrystallization of the semiconductor structure with a $CdCl_2$ catalyst. The process causes aggregation and growth of the structure's crystallites, thus the improvement of the active layer's parameters is observed. Recrystallization process parameters in the experiment have been adjusted to the substrate thermal resistance capabilities and standard recrystallization temperature profile. Maximum temperature for Upilex-S treatment is approximately 500 °C. According to the reports [22,23] and previous own research, the temperature of 420 °C is the minimum value for the possible recrystallization. Thus, in these investigations the temperature range of 420–500 °C was set as a testing range for CdTe recrystallization. This process was carried out using a programmable high temperature PEO601 furnace.

Unfortunately, a significant problem occurred during the realization of this procedure. Namely, disintegration of semiconductor structures and contact layers in all tested samples followed after each recrystallization process. This leads to the conclusion that copper contacts do not withstand the recrystallization process, therefore other materials such as Mo were tested in the further steps on this investigation.

In the further stage of the copper made base contacts analysis, one more experimental series has been carried out. In this case superstrate configuration of the flexible photovoltaic CdTe/CdS structure was used. The manufacturing of the cell was started by the selection of an appropriate substrate, which should be both high temperature resistant and sufficiently transparent in the spectral range of solar radiation, corresponding to the absorption spectrum of a CdTe/CdS structure. Basing on the previous authors' research, polyimide Kapton PV9102 foil has be chosen for this purpose. Selected foil is 38 μm thick and its average optical transmittance in the wavelength range of 400-800 nm was measured at the level of 71%. As emitter contacts, aluminum doped zinc oxide (ZnO:Al or AZO) layers, with surface resistance equal to 45 Ω/ square have been used. Optical transmittance measured in the range of 400÷800 nm was determined at the average level of 92%. On such prepared structures, two collecting silver front contacts have been deposited using screen-printing technique. Further steps were analogical as in the previous configuration. Layers of CdS and CdTe have been respectively deposited using PVD technology and recrystallized with $CdCl_2$ assistance. Several experiments have been conducted in different temperatures of the recrystallization, as previously in the range of 420-500 °C. In general, all deposited layers were stable and homogenous in all temperature variants before and after the thermal processes. The last stage of the photovoltaic structure manufacturing in the superstrate configuration is back contact layer formation. In this case, copper base contacts have been successfully deposited using thermal evaporation in PVD vacuum process. The obtained Cu layers show homogeneity on the whole surface, as well as good adhesion to the semiconductor CdTe layer. Complete flexible CdTe/CdS photovoltaic structure, with copper as a base contact layer, is shown in Fig. 6.

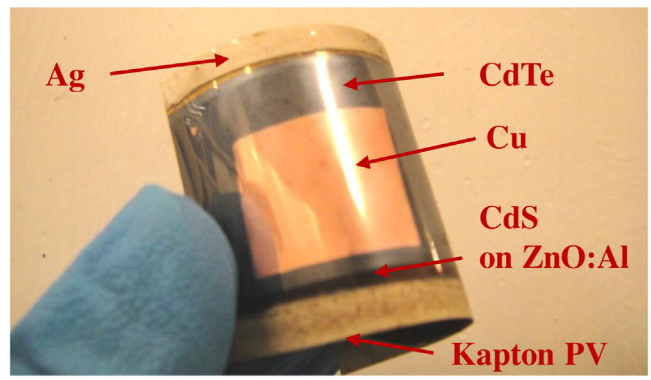

Fig. 6. Example of the flexible CdTe/CdS PV structure formed on the polyimide Kapton PV9102 foil with Cu base contact layer.

The obtained structure's quality allows for the surface elastic deformation without its degradation or destruction. The resistance of such prepared Cu layer is the same as it was in previously considered configurations. These experiments proved the suitability of PVD evaporated copper, as a proper base contact material for flexible CdTe/CdS structures manufactured in the superstrate configuration.

3.2. Molybdenum contacts

Molybdenum (Mo) is a second considered material as a back ohmic contact to the base CdTe layer of flexible solar cell, as it

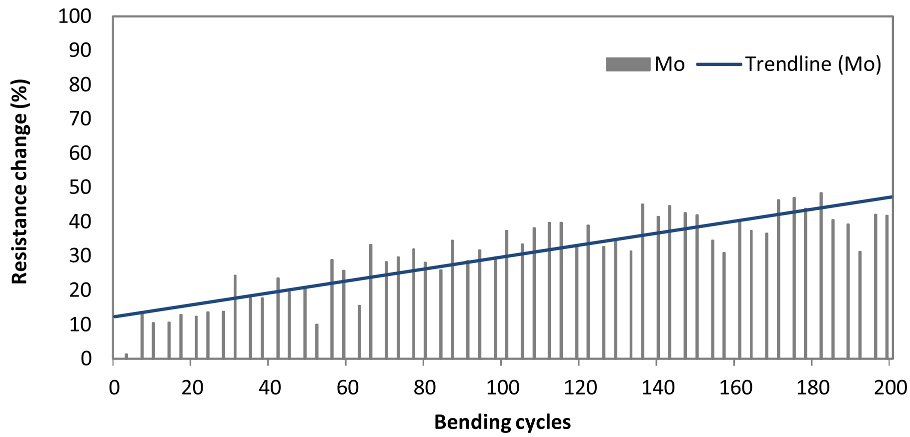

Fig. 7. Percentage change of the electrical resistance during bending on 25 mm diameter cylinder for Mo layers.

has many advantageous properties for this role. The most interesting properties of molybdenum as back contact for CdTe/CdS photovoltaic structure are as follows:

- appropriate linear coefficient of thermal expansion CTE (4.8 ppm/°C comparing to the value of 5.9 ppm/°C for CdTe in 20 °C temperature);
- relatively high work function (4.36–4.95 eV) in relation to the value of 5.7 eV for forming the optimal base contact to CdTe layer;
- the possibility to obtain high purity material, which helps to prevent undesirable dopants' diffusion to the base layer of cadmium telluride during thermal processes;
- the highest degree of corrosion resistance.

One can expect that with these features, molybdenum could form a proper contact to the CdTe/CdS structure, however layer's flexibility is equally important in the considered solutions. Research concerning verification of Mo suitability as base contact material for flexible PV structures was conducted analogically as for the copper layers.

For this purpose, a set of samples was prepared on 75 µm thick Upilex-S substrates. Molybdenum layers of 300 nm thickness have been obtained using vacuum electron gun evaporation in the PVD technique. Kurt Lesker Mo target pellets of 99.95% purity and also Mo crucible source have been used for the process. Surface resistance of such deposited thin films has been measured at the level of 2.8 Ω/ square.

Such prepared samples have been subjected to endurance tests, determining the level of the electrical resistance stability in the dynamic bending process. Dynamic bending was conducted according to the procedure described in Section 3.1 for copper layers. Figure 7 presents the results of measurements demonstrating the percentage change of the electrical resistance during bending.

Measurements show a trend of increased sheet resistance of molybdenum under the influence of mechanical stress in the form of a dynamic bending process. However, according to a relatively low initial value of these layers' resistance, these results can be considered as satisfactory and not exclude Mo layers from further research for the implementation as flexible base contacts to the CdTe/CdS solar cells.

The key step of the manufacturing process of this kind of cell is to apply a proper base structure of CdTe on a molybdenum layer, simultaneously retaining its both crystalline and mechanical parameters, as well as high adhesion to the substrate. Therefore, semiconductor CdTe base layers were deposited on Upilex-S substrates covered with previously evaporated thin Mo layer. CdTe deposition procedure was analogical as described in point 3.1. Cadmium telluride layers obtained in this experiment were homogenous, had a good adhesion to contact structures and high flexibility allowing for bending without degradation of their structures. Further step was the recrystallization process which was carried out at temperatures in the range of 420 to 500 °C in an inert nitrogen atmosphere (as in previous experiments). In this case, according to the expectations, thermal recrystallization has not influenced negatively on the structures' quality in any of investigated variant. Therefore, the realization of the further stage of research has been possible. CdS emitter layers were deposited in the same process as CdTe layers, however the substrate temperature during the process was much lower, in the range of 150-165 °C. As a result of this process, also correct semiconducting layers, with high adhesion and flexibility have been obtained. These structures have been subjected to the recrystallization procedure again, achieving almost complete flexible test PV structures grown in a substrate configuration.

Figure 8 shows the examples of a molybdenum layer evaporated on polyimide Upilex-S foils and flexible CdTe/CdS structure with Mo back contact.

Obtained results confirm the suitability of described technology and molybdenum usage as a base contact layer in a flexible photovoltaic CdTe/CdS structure formed in substrate configuration.

Some of the samples have been used for the final stage of this experiment, which was a construction of the complete flexible CdTe/CdS PV structure. Emitter contact in this case does not have to be high temperature resistant as it is almost the final layer in the structure. For this purpose, PEDOT transparent conductive polymer has been used. Additional collective emitter contacts have been made of a silver grid. Both PEDOT and silver were deposited using cheap and easy, screen-printing technique. A sample of the test flexible CdTe/CdS photovoltaic structure formed in the substrate configuration using the described technology is presented in Fig. 9.

Conducted measurements of the presented flexible photovoltaic structures show their proper operation, and their efficiencies are comparable to those obtained in the same technology on rigid substrates.

Presented investigations indicate the usefulness of the proposed solutions and processes developed for producing flexible photovoltaic structures based on CdTe and CdS semiconductor compounds. It has been shown that both, proposed substrate configuration and materials selected for the base contact are correct and can successfully lead to an efficient flexible photovoltaic cell construction.

3.3. Printed silver layers

The last investigated material for back contact was silver (Ag). This is an element, similarly as copper and molybdenum, belonging

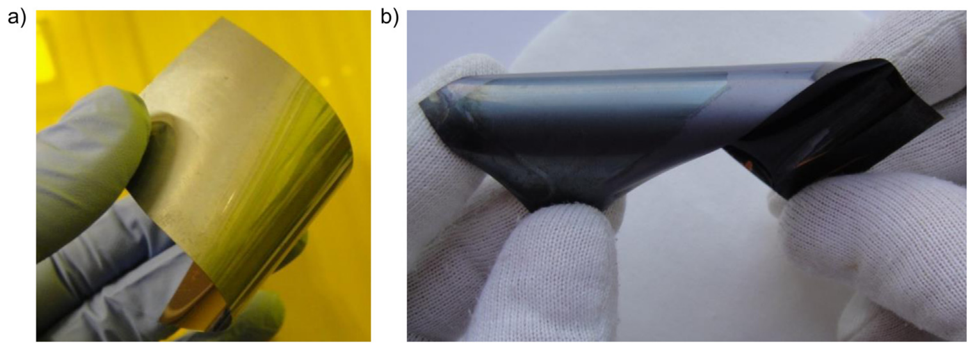

Fig. 8. Pictures of samples: a) Mo on foil, b) CdTe/CdS structure with Mo contact.

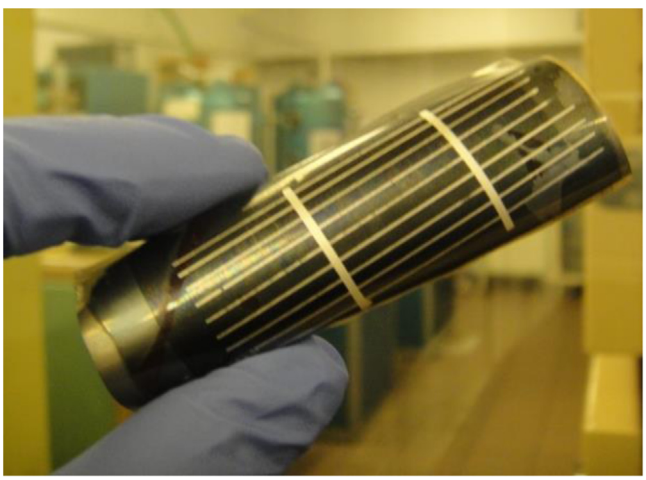

Fig. 9. Complete flexible CdTe/CdS photovoltaic structure, formed on Upilex-S foil with Mo back contacts.

to the group of transition metals. Silver has the highest, among all metals, values of the electrical and thermal conductivity. Parameters of Ag indicate the possibility of using it as an ohmic contact to the CdTe-based photovoltaic cell.

However, it can by only considered analogically as Cu, in superstrate configuration or as collective front contact material in substrate configuration. This is because of the high difference between silver and cadmium telluride thermal expansion coefficients (18.9 ppm/°C for Ag and 5.9 ppm/°C for CdTe).

In this case, for samples preparation a screen-printing technology has been used. Silver pastes have been deposited on Upliex-S polyimide foils in the form of a regular layer, as well as on more transparent Kapton PV substrates as nets of tiny "fingers" for application as front collective electrodes.

For all these layers' applications, a commercial silver paste Electrodag Acheson from Henkel has been used. Layers applied in this technique, are by definition much thicker than those made in vacuum evaporation technologies. Their thicknesses are in the range of several micrometers. After the process of screen printing, the layers are dried in 120 °C in order to obtain the stability of the structure. Such prepared silver samples were about 6 μm thick and the average surface resistance was 0.1 Ω/ square. Figure 10 shows the example samples.

Mechanical strength and electrical stability measurements have been conducted in the same conditions as for previously investigated materials. Figure 11 presents the results of measurements demonstrating the percentage change of the electrical resistance during bending.

Obtained results show that the dynamic bending has some effect on the electrical resistance values in both cases. On the average, the resistance increases linearly with the number of dynamic bending cycles. This increase is maximum of 20% (after 200 cycles) comparing to reference.

However, it should be noted that due to a very low initial resistance value, at the level of 0.1 Ω/ square, these changes, although the percentage may seem significant, are not very important from a practical point of view. Therefore, it can be concluded that the screen-printed silver layers may also be used for preparation of effective contacts in flexible CdTe/CdS photovoltaic structures. Notwithstanding, it should be noted that in these structures only superstrate configuration is suitable for Ag base contacts.

4. Conclusions

Basing on the conducted experiments, several test flexible CdTe/CdS photovoltaic structures were manufactured, both in substrate and superstrate configuration. All investigated materials are suitable for the second configuration where back metal contact is deposited as the final layer. Though, for a substrate design only

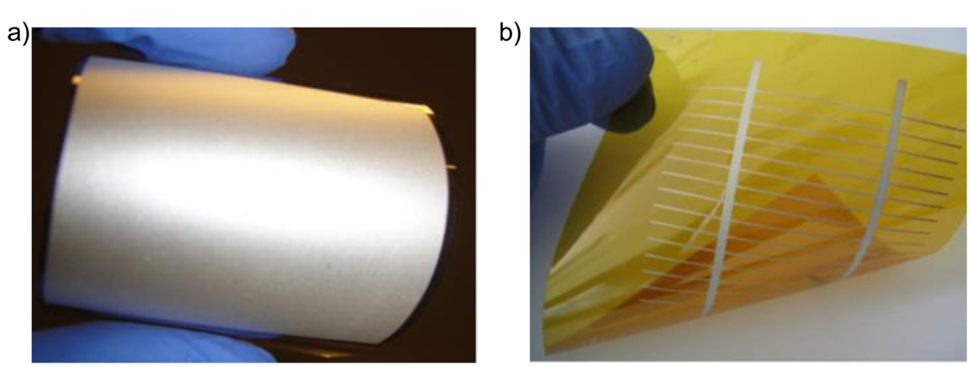

Fig. 10. Pictures of screen-printed samples: a) Ag layer on Upilex-S foil, b) Ag net on Kapton PV foil.

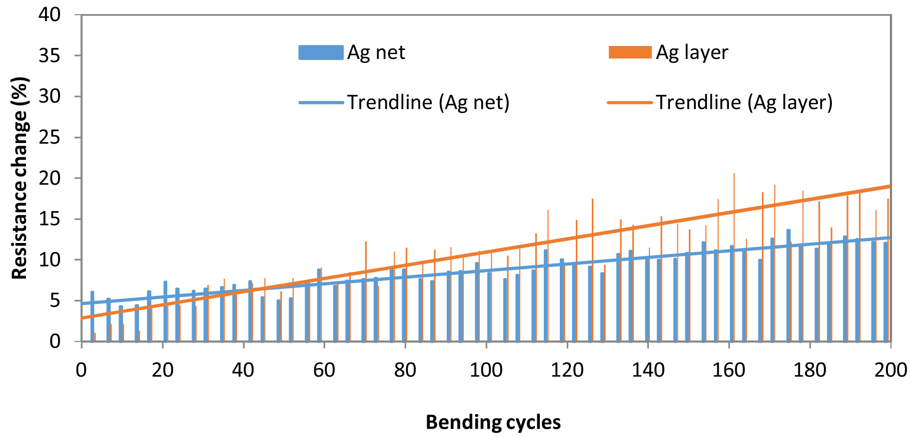

Fig. 11. Change of the electrical resistance during bending on a 25 mm diameter cylinder for Ag layers.

molybdenum is an appropriate material which meets the requirements of further technology steps.

For copper layers, in substrate configuration, disintegration of all structures was observed after the recrystallization process and further steps were not possible because of too high CTE differences between CdTe and Cu materials. Complete solar cell structures were achieved in the superstrate configuration, where copper was deposited as the last layer. There are even higher differences in thermal expansion coefficients of Ag and CdTe which excludes this material from further investigation in a substrate configuration.

However, the general conclusion is that after the defined limitations, all investigated materials can be used as base contacts in flexible CdTe/CdS photovoltaic technology. Moreover, all investigated samples showed a relatively good electrical resistance stability under the influence of dynamic bending, which is an important parameter in the research work of flexible photovoltaic devices.

Author statement

Katarzyna Znajdek: Conceptualization, Methodology, Validation, Formal Analysis, Investigation, Data Curation, Writing – Original Draft, Writing – Review & Editing, Visualization, Supervision, Project Administration, Funding Acquisition.

Maciej Sibiński: Validation, Formal Analysis, Writing – Original Draft, Writing – Review & Editing.

Andrzej Kubiak: Methodology, Investigation.

Łukasz Ruta: Methodology, Investigation.

Zbigniew Lisik: Supervision, Project Administration.

Daniel Janczak: Methodology, Investigation.

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