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Titolo CARBON-BASED HYBRID PLATFORMS FOR NOVEL PHOTOVOLTAIC DEVICES: BURIED INTERFACE CHEMISTRY AND CHARGE CARRIERS DYNAMICS [Tesi di dottorato]

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Sommario

Current photovoltaic (PV) market is strongly dominated by an intense use of silicon. Although it is the second most abundant element on the Earth crust, after oxygen, Si is never present in its pure form but always bounded with other elements, and relatively complex and expensive purification procedures are needed in order to have clean, crystalline and optimally doped pure silicon. This issue, joined with the ever-increasing demand of clean Si by almost all the technological modern applications, led scientists all over the world to look for suitable alternatives. One of the most promising options, is to try to substitute silicon with carbon, essentially for two reasons: (i) pure C not only exists in nature but can also be obtained and purified through easy and low-cost processes, (ii) carbon can behave as a metal or a semiconductor without being doped, depending only on the particular allotrope. Moreover, carbon allotropes capability of arranging in various geometry allows C-based materials to assume different dimensionality, starting from the quasi zero-dimensional fullerene to three-dimensional diamonds. This makes carbon nanomaterials excellent candidate for a wide range of electrical and technological devices, offering the possibility to chose the suitable allotropes depending on the particular task that is needed to be

fulfilled. For photovoltaic application, a semiconducting material which can provide dissociation sites for excitons is necessary. To accomplish this role, the mono-dimensional form of C, carbon nanotubes (CNTs), revealed to be a perfect substitute of p-type silicon, on one side of the junction because CNTs are naturally p-doped in air. Moreover, thanks to their peculiar geometry and extraordinary electrical conductivity, they are able to provide excellent transport path for the dissociated carriers with a very good transparency (which allows a relevant amount of incident light to reach the depletion region). In the first chapter of this thesis, carbon nanotubes will be introduced, emphasizing the properties which make this nanostructured materials optimal for PV applications. Then, the different types of carbon/silicon heterojunctions will be analyzed, starting from the classical semiconductor theory, to a more complex and realistic model. At the end of the chapter CNTs solar cells state of the art will be presented, highlighting the open questions at which this thesis is aimed to answer. The experimental techniques, such as angle-resolved X-rays photoelectron spectroscopy (AR-XPS) and transient reflectivity (TR) measurements, used to reach this goal will be presented in Chapter 2, together with the description of the manufacturing processes that yielded to the creation of three different series of PV devices, with an improvement of the efficiency from 0.1% to 12.2% in three years. In the third chapter, we will show how the complex buried interface between CNTs and Si can be investigated and modelled by means of photoelectron spectroscopy techniques. A complex oxide interface, composed by silicon dioxide and non-stoichiometric silicon oxide, has been unveiled and possible effects on the power conversion efficiency of PV devices are outlined. A systematic study on the chemical and physical properties of the buried interface will be presented in Chapter 4. Oxides have been alternatively removed and regrown using suitable acids and the effects on the PV performances will be discussed in detail in this chapter. The doping effects of acids on the carbon nanotubes will also be investigated through Raman spectroscopy. Acid effects on the heterojunctions will be unambiguously shown by the XPS measurements, and the matching of these data with the electrical PV measurements allows us to discuss the nature of the heterojunction in more detail. In order to properly address the operation mechanism of these devices, which can be either a conventional p-n or a metal-insulator-semiconductor (MIS) junction, the dynamics of charge transfer processes at the interface will be investigated in Chapter 5 with time-resolved pump-probe reflectivity measurement. The aim is to find a correlation between the thickness of the buried SiOxlayer and the carriers photogeneration and transport, comparing the device electrical parameter with the ultrafast behavior, analyzed by time-resolved reflectivity. These last findings, along with several improvements in the CNTs dispersion and deposition, have led to the creation of optimized third-series solar cells with a record efficiency of 12.2%, which will be fully characterized at the end of this last chapter through a combination of suitable experimental techniques, in order to highlight the factors which contributed to this huge jump in the power conversion efficiency. The stability in time of this optimized PV devices will finally be discussed.