多層光学薄膜の開発と太陽電池への応用に関する研究

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要　　旨

現在，世界で使われているエネルギー資源の約85％は化石燃料である。しかし，燃料の消費増大に伴い，化石燃料の枯渇や排出される二酸化炭素などの温室効果ガスの大気中濃度が増加して起こる地球温暖化などがエネルギー・環境問題として挙げられている。地球温暖化は環境のバランスを崩し，気温の上昇や海面の上昇，異常気象などを引き起こす。そのため地球温暖化への国際的な対応として，今後のエネルギー供給については，CO_2の主な発生源である化石燃料の割合を低減させるための配慮が不可欠になっている。そこで，環境にやさしい太陽電池が最も身近な新エネルギーとして注目されている。太陽電池はエネルギー源である太陽光はほぼ無尽蔵に得ることができ，騒音もなく，二酸化炭素などの排出がないなどの特長を持っている。

しかし，太陽電池の切実な需要がある一方で，その変換効率が15％前後と必ずしも高くはなく，さらに太陽光に含まれる赤外線領域の長波長光によって多結晶シリコン太陽電池の温度が上昇し，変換効率が減少するという欠点がある。また，太陽電池のカバーガラスによって発電に必要な入射太陽光が100％から92％までに減少し，太陽電池の発電量および変換効率を低減させる原因の一つと考えられている。

そこで，近年の光通信と薄膜プロセス技術の発展に伴い，性能の良い光学フィルター多層膜の開発が可能になっていることを踏まえた上で，本研究では，太陽電池の変換効率を向上させるために，太陽電池の表面温度上昇を防ぐことができる波長選択性透過多層膜と太陽電池発電に必要な可視光をより多く透過できる反射防止多層膜を設計・作製し，実際に太陽電池に応用して変換効率を向上させることを目的としている。

本論文は全6章で構成されており，以下のその概要をまとめて述べる。

第1章は序論であり，本研究の背景と目的について述べるとともに，太陽電池の発電原理，特徴，種類および多層光学薄膜について概説し，さらに波長選択性透過薄膜と反射防止膜について述べた。

第2章では，まずコンピューターシミュレーションソフトウェア（Essential Macleod）を用いた多層光学薄膜のシミュレーション設計方法について簡単に述べた。また，精密に多層光学薄膜を作製できる3元型RFスパッタリング法の成膜原理，特徴および成膜方法について説明した。

第3章では，作製した単層および多層光学薄膜の特性評価装置，多層光学薄膜付き太陽電池のソーラーシミュレーターによる電気的性能の測定方法などについて詳しく述べた。

第4章では，まず波長選択性透過薄膜と反射防止膜に用いる材料の単層膜を作製し，特性を評価した。様々な実験条件の下で作製した単層膜の膜厚，透過率，表面形状などを詳しく評価した。O_2ガス流量，Arガス流量，RF電力などのパラメーターを変化させて，それぞれの条件の下で作製した単層膜の中で，最も特性が良い薄膜の作製条件を明らかにした。次に，多層光学薄膜の理論式に

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より算出したパラメーターを基に波長選択性透過薄膜と反射防止膜を設計し，薄膜コンピューターシミュレーション最適化を行った。シミュレーション設計した波長選択性透過薄膜の透過率は可視光領域で平均98％に達しているとともに，赤外線領域で1％以下になっていた。シミュレーションによる片面と両面の反射防止膜の透過率は可視光領域で100％に達した。さらに，各材料による単層膜作製時の最適条件と多層光学薄膜デザイン理論に基づいたシミュレーション設計結果から，波長選択性透過薄膜と反射防止膜を作製するための各種パラメーターを決定し，ナノレベルの成膜制御を行いながらスパッタリング法により目標とする光学特性を持つ多層光学薄膜を作製した。作製した多層膜の表面形状をAFMを用いて評価した結果，十分に滑らかであり，入射光の反射や散乱が生じにくいと考えられる。また，透過特性を測定した結果，多層膜の透過率と波長の関係はほぼ設計通りであった。1,053 nm–1,365 nmの範囲の透過率は1％以下になっているが，483 nm–924 nmの範囲の透過率は高く，約88％に達していた。可視光領域の透過率は石英ガラスのみの場合より，平均約4％低いことがわかった。これらの結果から，作製した波長選択性透過薄膜は太陽電池の温度上昇を引き起こす長波長光をシャープに遮断するとともに，発電に必要な可視光を88％透過できることを明らかにした。また，作製した両面反射防止膜の透過率は可視光領域で98％に達して，450 nm–760 nmでの透過率は石英ガラスの透過率より高かった。作製した片面反射防止膜の透過率は460 nm–650 nmの範囲で96％に達して，450 nm–660 nmでの石英ガラスのみの透過率より高かった。二種類の反射防止膜の光学特性を分析した結果，反射防止膜は可視光領域の太陽光の透過率を上昇させるという目標を達成した。

第5章では，太陽電池のカバーガラスへ多層光学薄膜を成膜した場合としない場合の，太陽電池の表面温度と電気的な特性の変化を測定・検討した。測定は従来の通常のガラスと波長選択性透過薄膜と片面および両面反射防止膜を各々成膜したガラス，さらにこれらを組み合わせた光学膜を成膜したガラスをそれぞれ太陽電池に取り付けて行った。従来の太陽電池と多層光学薄膜を成膜したそれぞれの場合で，熱光源の加熱時間は0 minから20 minまで，太陽電池の表面温度と変換効率を測定した。測定した結果，波長選択性透過薄膜は赤外線を遮断することにより太陽電池の表面温度上昇を13.4℃～19.2℃抑制でき，また反射防止膜は可視光領域の太陽光の反射を防止することにより，太陽電池が発電に必要な可視光をより多く透過できることを明らかにした。そして，波長選択性透過薄膜と反射防止膜を組み合わせた多層光学薄膜を成膜することによって，太陽電池の変換効率を向上させることができることを確認した。

第6章は本論文の結論であり，本研究で得られた結果・結論，また今後の展望について述べた。

以上のように，本研究では太陽電池の変換効率上昇を目的として，温度上昇を防ぐ波長選択性透過薄膜と，可視光透過率を上昇させる片面および両面反射防止膜の設計および作製を行った。さらに，これらを組み合わせた多層光学薄膜を実際にカバーガラス上に成膜して太陽電池の電気特性を評価した結果，総合的な結果として太陽電池の変換効率を向上させることに成功した。本来太陽電池の変換効率を半導体自身に手を加えることなく向上させることは非常に難しい。本論文で述べたように，太陽電池のカバーガラスに多層光学薄膜を成膜するという簡単な方法で，変換効率を向上させるという手法は太陽電池の利用普及に対して大きく寄与でき，環境負荷の低減にもつながると考えられる。

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Research on Development of Multilayer Optical Thin Films and Their Application to Solar Cells

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Abstract

Of the energy sources currently consumed in the world, about 85 percent are fossil fuels. With the increasing fuel consumption, however, exhaustion of fossil fuels is becoming a concern and the concentration of emitted carbon dioxide and other greenhouse gases in the air is increasing to cause global warming, and these are cited as energy and environmental issues. Global warming upsets the environmental balance, raises the air temperature and sea level, and causes unusual weather. Therefore, as an international response to global warming, it is essential to pay attention to the energy supply from now on in order to reduce the proportion of fossil fuels, which are a major source of CO_2. Thus, the environmentally friendly solar cell is attracting attention as the most familiar new energy. The solar cell can obtain the almost limitless sunlight as its energy source, so it emits no noise, no carbon dioxide and no other gas and has these features. While there is a compelling demand for solar cells, their conversion efficiency is not necessarily high, or about 15 percent. As another drawback, the solar cell's polycrystalline silicon temperature is raised by the long wavelength components in the infrared region of sunlight, which further decreases the conversion efficiency. The cover glass on the solar cell decreases the incident sunlight necessary for power generation from 100 to 92 percent and this also is thought to be one of the causes of the decrease in amount and efficiency of power generation of the solar cell. With the recent progress of optical communication and thin film process technology, it has become possible to develop a high-performance optical filter with a multilayer film coating. Based on this and aiming at improving the conversion efficiency of the solar cell, this study designed and fabricated a wavelength selective transmission multilayer film that can prevent the solar cell's surface temperature rise and an antireflection multilayer coating that can transmit a greater amount of visible light necessary for power generation by the solar cell. It was intended to improve the conversion efficiency by applying them to an actual solar cell.

This thesis consists of a total of six chapters and they are collectively outlined below.

Chapter 1 is an introduction and describes the background and purpose of this study. Further, it outlines solar cells concerning their generation principles, features, types and multilayer optical thin films. It also describes a wavelength selective transmission thin film and antireflection coating.

Chapter 2 summarizes first a multilayer optical thin film simulation design method using

computer simulation software Essential Macleod. It also explains the deposition principle, features and deposition method of ternary type RF sputtering that can fabricate a multilayer optical thin film accurately.

Chapter 3 deals with the fabricated single-layer and multilayer optical thin films and details their characteristic measurement equipment. It also treats solar cells with a multilayer optical thin coating and a solar simulator for them and describes an electrical performance measurement method using the simulator.

In Chapter 4, single-layer films were fabricated using the materials used to make a wavelength selective transmission thin film and antireflection coating and their characteristics were evaluated first. Single-layer films were fabricated under various experimental conditions and their thickness, transmittance and surface topography were evaluated in detail. Various single-layer films were fabricated under varied conditions of O_2 gas flow rate, Ar gas flow rate and RF power to find the thin film fabrication conditions that would bring the best characteristics. Then, the parameters were calculated by a theoretical formula of multilayer optical thin film and a wavelength selective transmission thin film and antireflection coating were designed based on these parameter values and underwent optimization by a thin film computer simulation. For the wavelength selective transmission thin films designed by simulation, their transmittance attained 98 percent on average in the visible spectral region while it was held down to 1 percent or less in the infrared region. The transmittance of antireflection coating on one surface and both surfaces according to a simulation attained 100 percent in the visible spectral region. Further, from the optimum single-layer film fabrication conditions using various materials and results of simulation design based on multilayer optical thin film design theory, the various parameters to fabricate a wavelength selective transmission thin film and antireflection coating were determined. Then, a multilayer optical thin film was fabricated by sputtering with deposition control at the nanometer level to have the target optical characteristics. The surface topography of the fabricated multilayer film was evaluated using AFM and, as a result, it was so smooth that reflection or scattering of incident light was thought to hardly occur. The transmittance characteristics were measured to find, as a result, that the transmittance of the multilayer film versus wavelength was almost as designed. In the range from 1,053 to 1,365 nm, the transmittance was 1 percent or less while in the range from 483 to 924 nm, the transmittance reached as high as about 88 percent. It was found that the transmittance in the visible spectral region was about 4 percent lower on average than quartz glass without coating. From these results, it was made clear that the fabricated wavelength selective transmission thin film cut off long wavelength light sharply, which would cause a temperature rise of the solar cell, and could transmit 88 percent of visible light necessary for power generation. For the antireflection coating deposited on both sides, the transmittance reached 98 percent in the visible spectral region and was higher than that of quartz glass without coating in the 450 to 760-nm region. For the antireflection coating deposited on one side, the transmittance reached 96 percent in the range from 460 to 650 nm and was higher than that of quartz glass without coating from 450 to 660 nm. The optical characteristics of the two types of antireflection coatings were analyzed with the result that the objective of improving the transmittance of sunlight in the visible spectral region by antireflection coating was successfully achieved.

Chapter 5 deals with two cases where the cover glass of the solar cell is coated and is not coated with a multilayer optical thin film and measures and investigates the difference in surface temperature and electrical characteristics of the solar cell between the two cases. The measurement was performed on an untreated ordinary glass, a glass coated with a wavelength selective transmission thin film, glasses coated with an antireflection coating on one side and both sides, and a glass coated with a combination of these optical films, each attached to a solar cell. For a conventional solar cell and solar cell coated with a multilayer optical film, their surface temperature and conversion efficiency were measured while irradiating them by a hot light source for a heating time of 0 to 20 minutes. As a result of the measurement, it was found that the wavelength selective transmission film could cut off infrared rays and thereby hold the solar cell's surface temperature rise down to 13.4 to 19.2°C and the antireflection coating could prevent reflection of sunlight in the visible spectral region and thereby transmit a greater amount of visible light necessary for power generation by the solar cell. It was also verified that the conversion efficiency of the solar cell could be improved by depositing a multilayer optical thin film consisting of a wavelength selective transmission thin film combined with an antireflection coating.

Chapter 6 concludes this thesis and describes the results and conclusions obtained in this study and prospects for the future.

As stated above, this study aimed at improving the conversion efficiency of the solar cell and designed and fabricated a wavelength selective transmission thin film to prevent a temperature rise and an antireflection coating deposited on one side and both sides to improve the transmittance of visible light. Further, a multilayer optical film consisting of these films in combination was deposited on an actual cover glass and the electrical characteristics of the solar cell were evaluated. As a comprehensive result, the conversion efficiency of the solar cell was successfully improved. It is intrinsically very difficult to improve the conversion efficiency of a solar cell without modifying the semiconductor itself. As described in this thesis, however, the conversion efficiency could be improved by simply depositing a multilayer optical thin film on the cover glass of the solar cell. So, this technique is thought to be able to contribute much to the use and spread of the solar cell and lead to a reduction in environmental loads.

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