UNIVERSITI PUTRA MALAYSIA

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STRUCTURAL, MAGNETIC AND ELECTRICAL PROPERTIES OF La-A-Mn-O (A=Ca, Sr, Ba, Na AND K) IN BULK AND THIN FILM

By

NG SIAU WEI

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STRUCTURAL, MAGNETIC AND ELECTRICAL PROPERTIES OF La-A-Mn-O (A=Ca, Sr, Ba, Na AND K) IN BULK AND THIN FILM

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Chairman: Lim Kean Pah, PhD

Faculty: Science

Perovskite manganites oxide materials have attracted much attention due to their promising potential applications in the magnetic sensor or devices. In this research, $La_{1-x}A_xMnO_3$ (x = 0.33 when A=Ca, Sr and Ba; x= 0.2 when A=Na and K) in bulk form were prepared via solid state reaction method. $La_{0.67}Sr_{0.33}MnO_3$ (LSMO) and $La_{0.8}Na_{0.2}MnO_3$ (LNMO) are then converted to thin film via pulse laser deposition method (PLD) on different substrates (corning glass, fused silica glass and MgO (100)) and deposition duration. Rietveld refinement of X-ray diffraction data showed that all samples are polycrystalline having trigonal crystal structure except for $La_{0.67}Ca_{0.33}MnO_3$ (LCMO) and $La_{0.67}Ba_{0.33}MnO_3$ (LBMO) which are orthorhombic. From the Rietveld refinement, we observed that the lattice parameter, Mn-O bond length and bond angle changed in thin film which were influenced by the substrates type and deposition duration. The thickness of films was in the range of ~0.3-3.0 μm. The crystallite size for the thin film is between 15-22 nm. The grain size distribution for bulk samples are around 1.0-2.0 μm. A huge change of surface microstructure can be observed for thin film samples, where the grain size is reduced to ~50-150 nm.

Some nano-crack effects were observed in the thin film samples where this effect is due to the different coefficient of thermal expansion between the film and substrates during the annealing process. Thin film samples showed a much higher resistance (about 2-3 orders) due to the existence of disordered phase at the grain boundary and/or nano-crack barrier that causes higher scattering and/or tunneling effect when the electrons pass through them. The metal-insulator temperature (T_p) for LSMO shifted to lower values in the thin films, probably due to the change of Mn-O bond length and bond angle. Conversely, LNMO system showed greater T_p value in thin film suggesting that the grain boundary effect might also contribute to the T_p changes. Nonetheless, the deposition duration and substrates used also influence the T_p value. Overall, negative magnetoresistance (MR) have been obtained for bulk and thin films. The MR value increases with decreasing temperature at low applied magnetic field which known as Low Field Magnetoresistance (LFMR). In thin film form the %MR value has been improved with -25% for LS_M15 and -22% for LN_M20 as compared to that of the bulk LSMO (-16%) and LNMO (-21%) when a magnetic field of 1 Tesla was applied at 90 K.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Master Sains

SIFAT STRUKTUR, MAGNET DAN ELEKTIK BAGI PUKAL DAN FILEM NIPIS La-A-Mn-O (A=Ca, Sr, Ba, Na DAN K)

oleh

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March 2011

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Bahan perovskit manganites oksida telah menarik perhatian ramai disebabkan oleh potensinya dalam aplikasi sensor magnetik atau peranti. Dalam kajian in, $La_{1-x}A_xMnO_3$ (x = 0.33 bila A=Ca, Sr dan Ba; x= 0.2 bila A=Na dan K) disediakan melalui kaedah tindak balas keadaan pepejal dalam bentuk pukal. Kemudiaan, $La_{0.67}Sr_{0.33}MnO_3$ (LSMO) dan $La_{0.8}Na_{0.2}MnO_3$ (LNMO) telah ditukar menjadi filem nipis melalui keadah Mendapan Dedenyut Laser (MDL) di atas substrat (kaca Corning 7059, kaca fused silica and MgO (100)) dan tempoh mendapan yang berbeza. Kaedah Rietveld menunjukkan bahawa semua sampel adalah polihablur dan membentuk struktur trigonal kecuali $La_{0.67}Ca_{0.33}MnO_3$ (LCMO) dan $La_{0.67}Ba_{0.33}MnO_3$ (LBMO) yang membentuk struktur ortorombus. Daripada kaedah Rietveld, kami mendapati bahawa kekisi, panjang ikatan Mn-O dan sudutnya berubah dalam bentuk filem nipis dimana perubahan ini adalah dipengaruhi oleh jenis substrat dan tempoh mendapan. Ketebalan filem nipis adalah dalam julat ~0.3-3.0 μm. Saiz butiran bagi filem nipis adalah dalam julat 15-22 nm. Taburan saiz butiran bagi bentuk pukal adalah dalam julat 1.0-2.0 μm. Perubahan yang besar di

permukaan mikrostuktur dapat dikesan dimana saiz butiran berkurang menjadi ~50-150 nm bagi sampel filem nipis. Beberapa kesan nano-retakan dapat diperhatikan dalam filem nipis dan kesan ini adalah disebabkan oleh pekali pengembangan terma yang berbeza antara filem dan substrat semasa proses memijar. Sampel filem nipis menunjukkan kerintangan elektik yang jauh lebih tinggi (kira-kira 2-3 ganda) disebabkan oleh kewujudan fasa yang tidak tersusun di permukaan butiran dan/atau sekatan nano-retakan yang menyebabkan kesan serakkan spin dan/atau kesan penerowongan spin semasa elektron melaluinya. Suhu logam-penebat (T_p) bagi LSMO menganjak ke nilai yang lebih rendah dalam filem nipis, ini mungkin disebabkan oleh perubahan yang berlaku di panjang ikatan Mn-O dan sudutnya semasa bertukar ke filem nipis. Sebaliknya, sistem LNMO menunjukkan nilai T_p yang lebih besar dalam filem nipis mencadangkan bahawa kesan sempadan butiran mungkin juga menyumbang bagi pertukaran T_p. Walau bagaimanapun, tempoh mendapan dan substrat yang diguna juga mempengaruhi nilai T_p. Pada keseluruhannya, magnetorintangan (MR) negatif telah diperolehi bagi pukal dan filem nipis. Nilai MR bertambah dengan penyusutan suhu dalam keadaan medan magnet rendah yang dikenali sebagai kesan magnetorintangan medan rendah (LFMR). Sampel filem nipis telah meningkatkan nilai %MR dengan -25% bagi LS_M15 dan -23% bagi LN_M20 apabila berbanding dengan bahan pukal LSMO (-16%) dan LNMO (-20%) dalam medan magnet 1 Tesla pada suhu 90 K.

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DECLARATION

I declare that the thesis is my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously, and is not concurrently, submitted for any other degree at Universiti Putra Malaysia or at any other institution.

NG SIAU WEI

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