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Electrophysical characteristics of nanodimensional cobalte-cuprate-manganite $\text{LaNa}_2\text{CoCuMnO}_6$ and nickelite-cuprate-manganite $\text{LaNa}_2\text{NiCuMnO}_6$

The temperature dependences of the electric capacity, dielectric constant and electrical resistance of cobalte-cuprate-manganite of lanthanum and sodium of $\text{LaNa}_2\text{CoCuMnO}_6$ and nickelite-cuprate-manganite of lanthanum and sodium of $\text{LaNa}_2\text{NiCuMnO}_6$ were investigated on the LCR-800 serial device (manufactured by Taiwan) at the operating frequencies of 1 kHz, 5 kHz, and 10 kHz in interval of 293–483 K through 10 K continuously in dry air. It was determined that $\text{LaNa}_2\text{CoCuMnO}_6$ in interval of 293–483 K shows the semiconductor conductivity. A band gap (ΔE) is 0.54 eV. The compound has the high values of the dielectric constant, which are equal $2.17 \cdot 10^6$ (1 kHz), $2.31 \cdot 10^5$ (5 kHz), $8.22 \cdot 10^4$ (10 kHz) at 293 K and $8.49 \cdot 10^8$ (5 kHz), $7.87 \cdot 10^7$ (10 kHz) at 483 K. $\text{LaNa}_2\text{NiCuMnO}_6$ in interval of 293–483 K demonstrates the semiconductor conductivity ($\Delta E = 0.48$ eV), at 433–443 K — the metallic conductivity and at 453–483 K — the semiconductor conductivity ($\Delta E = 2.33$ eV). The values of the dielectric constant are $4.97 \cdot 10^3$ (1 kHz), $9.2 \cdot 10^2$ (5 kHz), $5.1 \cdot 10^1$ (10 kHz) at 293 K and $1.02 \cdot 10^6$ (1 kHz), $1.98 \cdot 10^5$ (5 kHz) and $9.76 \cdot 10^4$ (10 kHz) at 483 K. The compounds can be classified as the narrow-band gap semiconductors and they are of interest for the semiconductor and microcapacitor technologies.

Keywords: electrophysics, cobalte, nickelite, cuprate, manganite, lanthanum, sodium, dielectric constant, electrical resistance.

Introduction

The compounds, based on the manganites, nickelites, cuprates and cobaltites of the rare-earth elements doped with oxides of the alkali and alkaline earth metals, have the unique physicochemical properties such as semiconductor, superconductive, ferroelectric, para-, ferro-, antiferromagnetic and other properties [1–4].

It should be pointed that the compounds based on lanthanum and strontium nickelates have the high values of the dielectric constant [5]. The Chemical-Metallurgical Institute performs the systematic and targeted studies on the synthesis and investigation of the physicochemical characteristics of manganites, ferrites, chromites, cuprates, cobaltites, nickelites of the rare earths doped with oxides of the alkali and alkaline earth metals with the valuable properties [6–10].

The purpose of this paper is to investigate the electrophysical properties of nanodimensional cobalte-cuprate-manganite of lanthanum and sodium of $\text{LaNa}_2\text{CoCuMnO}_6$ and nickelite-cuprate-manganite of lanthanum and sodium of $\text{LaNa}_2\text{NiCuMnO}_6$. The mentioned compounds synthesized with the ceramic technology in the interval of 800–1200 °C by the interaction of stoichiometric amounts of La_2O_3 , CoO, NiO, CuO, Mn_2O_3 and Na_2CO_3 . By milling of polycrystalline samples on Retsch MM301 vibration mill (Germany) were obtained their nanodimensional (nanocluster) particles, which sizes were defined on the atomic-force microscope JSPM-5400 Scanning Probe Microscope «JEOL» (Japan). It was found that both compounds are crystallized in a isometric system in the structure with the following parameters of grid: $\text{LaNa}_2\text{CoCuMnO}_6$ $a=14.43 \pm 0.02 \text{ \AA}$; $V^o=3005.5 \pm 0.06 \text{ \AA}^3$; $Z=4$; $V^o_{elec.cell}=751.38 \pm 0.02 \text{ \AA}^3$; $\rho_{roent.}=3.86$; $\rho_{pick.}=3.72 \pm 0.01 \text{ g/cm}^3$, $\text{LaNa}_2\text{NiCuMnO}_6$ — $a=14.19 \pm 0.02 \text{ \AA}$, $V^o=2859.42 \pm 0.06 \text{ \AA}^3$, $Z=4$, $V^o_{elec.cell}=714.86 \pm 0.01 \text{ \AA}^3$, $\rho_{roent.}=3.38$; $\rho_{pick.}=3.29 \pm 0.02 \text{ g/cm}^3$ [11–13].

Results and discussion

The measurements of the electrophysical properties were carried out according to the procedures [14, 15].

The investigation of the electrical characteristics (dielectric constant and electrical resistance) was made with the measuring of the electric capacity of the samples on the LCR-800 (Taiwan) serial device at the operating frequency of 1 kHz, 5 kHz and 10 kHz continuously in dry air in a thermostatic mode with holding time at each fixed temperature.

Some plane-parallel samples were preliminarily made as disks with 10 mm diameter and 5–6 mm thickness with a binder additive (~1.5 %). The pressing was carried out under a pressure of 20 kg/cm². The received disks were fired in a silit furnace at 200 °C for 6 h. Then they were thoroughly double-sided grinding.

The dielectric constant was determined from the electric capacity of a sample at the known values of thickness and a surface area of electrodes. The Sawyer-Tower circuit was used to receive correlations between the electric induction D and electric field E. The visual observation of D (hysteresis loop of E) was made on the C1–83 oscilloscope with potential divider consisting of resistance of 6 mOhm and 700 kOhm and a reference capacitor of 0.15 µF. The frequency of generator is 300 Hz.

The samples were placed in the furnace in all temperature studies. Temperature was measured with a chromel-alumel thermocouple connected to B2–34 voltmeter (± 0.1 mV). The rate-of-change of temperature is ~5 K/min. A value of the dielectric constant at each temperature was defined from formula:

Then they were thoroughly double-sided grinding.

$$\varepsilon = \frac{C}{C_0}, \quad (1)$$

where $C_0 = \frac{\varepsilon_0 \cdot S}{d}$ — a capacity of capacitor without a test substance (aerial).

The band gap (ΔE) of the test substance was calculated under formula:

$$\Delta E = \frac{2kT_1T_2}{0.43(T_2 - T_1)} \lg \frac{R_1}{R_2}, \quad (2)$$

where k — Boltzmann constant equal to $8.6173303 \cdot 10^{-5}$ eV·K⁻¹, R_1 — resistance at T_1 , R_2 — resistance at T_2 .

The dielectric constant of a standard substance — barium titanate (BaTiO₃) was measured at frequencies of 1 kHz, 5 kHz, and 10 kHz to confirm the received data. Values defined in an experiment of the dielectric constant at 293 K at frequencies of 1 kHz and 5 kHz are satisfactory with its recommended value of 1400 ± 250 [16–18]. In addition, the observed changes in the electrical conductivity of BaTiO₃ at 383 K in all frequencies (1 kHz, 5 kHz, and 10 kHz) are consistent with its transition from the perovskite cubic phase $Pm3m$ to the tetragonal (polar) ferroelectric phase with space group $P4mm$ [16–18].

It should be pointed that despite the lower values of the dielectric constant of BaTiO₃ in frequency of 10 kHz and at T equal to 293 K, 303 K, 313 K, all ε values of BaTiO₃ in all three frequencies (1 kHz, 5 kHz and 10 kHz) at 313–483 K have approximately the same values up to 2150 which testifies that a change in frequency does not particularly influence on the temperature dependence of the dielectric constant of BaTiO₃ at 313–483 K.

Figure 1 and table 1 below demonstrate the results of the electrophysical characteristics of the studied compounds.

Table 1

Dependence of electrical resistance (R) and dielectric constant (ε) on temperature of LaNa₂CoCuMnO₆ and LaNa₂NiCuMnO₆ at various frequencies

T, K	ε	lg ε	lg R	ε	lg ε	lg R
1	2	3	4	5	6	7
LaNa ₂ CoCuMnO ₆			LaNa ₂ NiCuMnO ₆			
Measurement frequency 1 kHz						
293	2171210	6,34	4,28	4971	3,70	5,72
303	1803567	6,24	4,23	5883	3,77	5,67
313	1404926	6,15	4,12	7746	3,89	5,57
323	1576730	6,20	3,96	11857	4,07	5,43
333	2049087	6,31	3,82	17811	4,25	5,29
343	2677028	6,43	3,70	27406	4,44	5,14
353	3203737	6,51	3,62	39710	4,60	5,00
363	4104119	6,61	3,54	54742	4,74	4,87
373	5784119	6,76	3,46	71452	4,85	4,77
383	8776221	6,94	3,38	97729	4,99	4,64

1	2	3	4	5	6	7
393	13976221	7,15	3,28	131335	5,12	4,52
403	26967304	7,43	3,13	165215	5,22	4,43
413	60578344	7,78	3,01	229605	5,36	4,30
423	242335457	8,38	2,86	339679	5,53	4,16
433	404076433	8,61	2,77	377230	5,58	4,16
443	604526539	8,78	2,67	343902	5,54	4,28
453	742734607	8,87	2,59	435548	5,64	4,23
463	849248408<	8,93<	2,51	608350	5,78	4,12
473	849248408<	8,93<	2,41	762270	5,88	4,04
483	849248408<	8,93<	2,35	1016889	6,01	3,93
Measurement frequency 5 kHz						
293	230921	5,36	4,26	920	2,96	5,49
303	210887	5,32	4,20	1079	3,03	5,45
313	217248	5,34	4,08	1419	3,15	5,37
323	311142	5,49	3,92	2207	3,34	5,25
333	446505	5,65	3,77	3253	3,51	5,14
343	601911	5,78	3,65	5072	3,71	5,01
353	693461	5,84	3,57	7616	3,88	4,88
363	850701	5,93	3,49	10984	4,04	4,76
373	1139363	6,06	3,41	14833	4,17	4,66
383	1613673	6,21	3,34	20838	4,32	4,54
393	2401699	6,38	3,25	28602	4,46	4,42
403	4458684	6,65	3,11	36620	4,56	4,33
413	9542251	6,98	2,99	51147	4,71	4,21
423	34073036	7,53	2,85	75204	4,88	4,07
433	55324841	7,74	2,76	79651	4,90	4,09
443	82969851	7,92	2,66	69204	4,84	4,22
453	103753716	8,02	2,58	85219	4,93	4,16
463	123745223	8,09	2,51	118284	5,07	4,07
473	158989384	8,20	2,40	148467	5,17	3,99
483	180696391	8,26	2,34	198510	5,30	3,88
Measurement frequency 10 kHz						
293	82189	4,92	4,24	51	1,71	5,34
303	80797	4,91	4,17	593	2,77	5,31
313	97036	4,99	4,04	763	2,88	5,24
323	150225	5,18	3,87	1147	3,06	5,14
333	223618	5,35	3,74	1649	3,22	5,03
343	301792	5,48	3,63	2498	3,40	4,92
353	347406	5,54	3,55	3736	3,57	4,80
363	424773	5,63	3,47	5361	3,73	4,70
373	561792	5,75	3,39	7312	3,86	4,60
383	777214	5,89	3,32	10376	4,02	4,49
393	1128577	6,05	3,23	14534	4,16	4,37
403	2073461	6,32	3,09	18904	4,28	4,28
413	4425478	6,65	2,98	26702	4,43	4,16
423	14367728	7,16	2,84	39228	4,59	4,03
433	22727813	7,36	2,75	39566	4,60	4,07
443	34175796	7,53	2,65	33516	4,53	4,19
453	43345223	7,64	2,57	40940	4,61	4,14
463	52411040	7,72	2,50	57187	4,76	4,04
473	68501911	7,84	2,40	72250	4,86	3,96
483	78744798	7,90	2,34	97570	4,99	3,85

The results of this investigation given in the table 1 and figure 1 show that $\text{LaNa}_2\text{CoCuMnO}_6$ has the semiconductor conductivity in the temperature interval at 293–483 K and measurement frequencies of 1, 5, and 10 kHz, the band gap is 0.54 eV and it can be fitted to the narrow-band gap semiconductors.

This compound has the high values of the dielectric constant. Thus, $\text{LaNa}_2\text{CoCuMnO}_6$ at 293 K has the following values of the dielectric capacity (ϵ): $2.17 \cdot 10^6$ (1 kHz), $2.31 \cdot 10^5$ (5 kHz), $8.22 \cdot 10^4$ (10 kHz) and at 483 K — $< 8.49 \cdot 10^8$ (1 kHz), $1.81 \cdot 10^8$ (5 kHz) and $7.87 \cdot 10^7$ (10 kHz).

$\text{LaNa}_2\text{NiCuMnO}_6$ in interval of 293–483 K demonstrates the semiconductor conductivity, at 433–443 K — the metallic conductivity and at 453–483 K — the semiconductor conductivity. The band gap at 293–433 K is 0.48 eV, and at 443–483 K — 2.33 eV, and this compound can be fitted to narrow-band gap semiconductors.

$\text{LaNa}_2\text{NiCuMnO}_6$ at 293 K has the following values of the dielectric capacity — $4.97 \cdot 10^3$ (1 kHz), $9.2 \cdot 10^2$ (5 kHz), $5.1 \cdot 10$ (10 kHz) and at 483 K — $1.02 \cdot 10^6$ (1 kHz), $1.98 \cdot 10^5$ (5 kHz) and $9.76 \cdot 10^4$ (10 kHz).

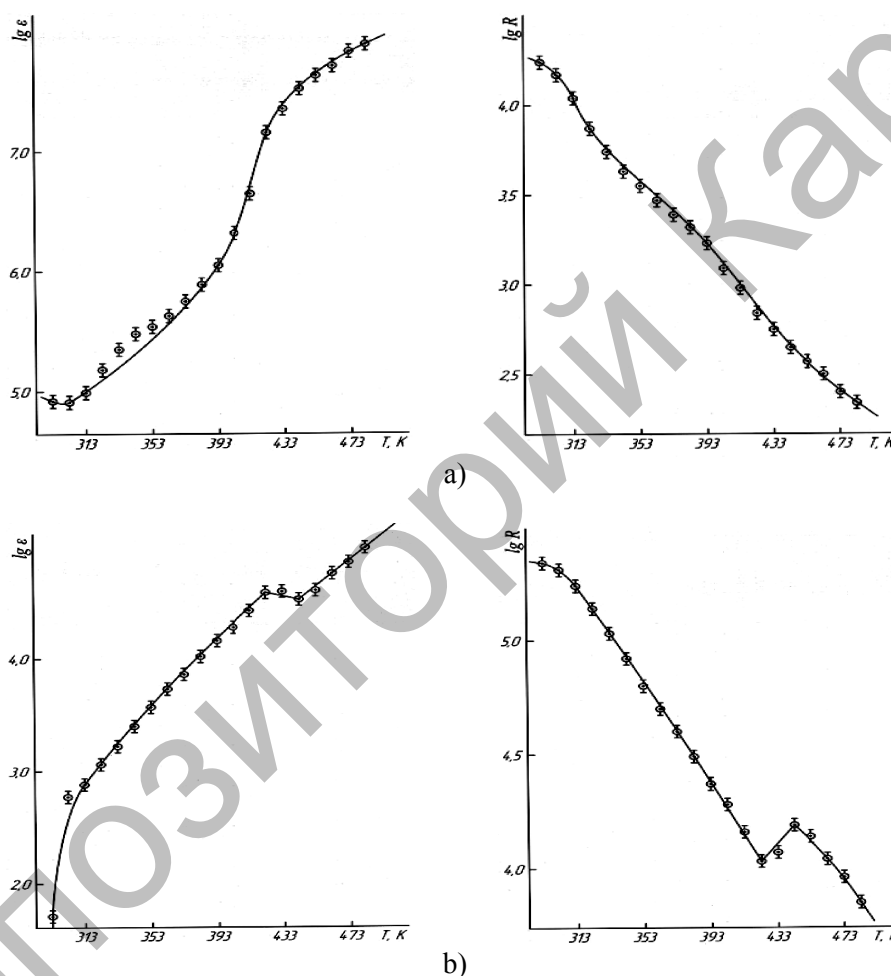


Figure 1. Dependence of dielectric constant (ϵ) and electrical resistance (R) of $\text{LaNa}_2\text{CoCuMnO}_6$ (a) and $\text{LaNa}_2\text{NiCuMnO}_6$ (b) on temperature at measurement frequency of 10 kHz

Conclusion

The temperature dependences of the dielectric constant and electrical resistance of the new nanodimensional $\text{LaNa}_2\text{CoCuMnO}_6$ and $\text{LaNa}_2\text{NiCuMnO}_6$ were first investigated in the interval of 293–483 K. It was found that $\text{LaNa}_2\text{CoCuMnO}_6$ is of interest for the semiconductor and microcapacitor technologies, having the high values of the dielectric constant at all measured frequencies of 1, 5, and 10 kHz.

$\text{LaNa}_2\text{NiCuMnO}_6$ compound is of interest for the semiconductor technology. Both compounds belong to narrow-band gap semiconductors.

Acknowledgments

The work was performed within the Contract No. 65 dated 23.02.2018 signed between Committee of science of the Ministry of Education and Science of the Republic of Kazakhstan and Abishev Chemical-Metallurgical Institute, the Grant Code of Individual Registration Number: AP05131317; AP05131333).

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**Наноөлшемді $\text{LaNa}_2\text{CoCuMnO}_6$ кобальт-купрат-манганитімен
 $\text{LaNa}_2\text{NiCuMnO}_6$ никелит-купрат-манганитінің
электрофизикалық сипаттамалары**

10 К арқылы 293–483 интервалында 1 кГц, 5 кГц және 10 кГц жиілікте жұмыс істейтін LCR-800 (Тайвань өндірісінің) сериялы кондырғысында $\text{LaNa}_2\text{CoCuMnO}_6$ лантан мен натрий кобальт-купрат-

манганиті және $\text{LaNa}_2\text{NiCuMnO}_6$ лантан мен натрий никелит-купрат-манганитінің электрсыымдылықтары, диэлектрлік өтімділіктері, электрқарсылықтарының температураға тәуелділіктері үздіксіз құрғақ ауада зерттелген. $\text{LaNa}_2\text{CoCuMnO}_6$ қосылысының 293–483 К аралықта жартылай өткізгіштік қасиет танытуы анықталды. Тыйым салу аймақ ені (ΔE) 0,54 эВ тең. Қосылыс 293 К $2,17 \cdot 10^6$ (1 кГц), $2,31 \cdot 10^5$ (5 кГц), $8,22 \cdot 10^4$ (10 кГц) және 483 К-де $8,49 \cdot 10^8$ (5 кГц), $7,87 \cdot 10^7$ (10 кГц) тең болатын диэлектрлік өтімділіктің алып мәніне ие. $\text{LaNa}_2\text{NiCuMnO}_6$ қосылысы 293–483 К аралықта ($\Delta E = 0,48$ эВ) жартылай өткізгіштік, 433–443 К-де — металдық және 453–483 К-де — ($\Delta E = 2,33$ эВ) қайта жартылай өткізгіштік қасиетке ие. Диэлектрлік өтімділік мәндері 293 К-де $4,97 \cdot 10^3$ (1 кГц), $9,2 \cdot 10^2$ (5 кГц), $5,1 \cdot 10^1$ (10 кГц) және 483 К-де $1,02 \cdot 10^6$ (1 кГц), $1,98 \cdot 10^5$ (5 кГц) және $9,76 \cdot 10^4$ (10 кГц) тең. Қосылыстарды жіңішке аймақты (қуыс аймақты) жартылай өткізгіштерге жатқызуға болады және олар жартылай өткізгіштер мен микроконденсаторлар технологиясы үшін қызығушылық тудырады.

Кілт сөздер: электрофизика, кобальт, никелит, купрат, манганит, лантан, натрий, диэлектрлік өтімділік, электрқарсылық.

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Электрофизические характеристики наноразмерных кобальто-купрато-манганита $\text{LaNa}_2\text{CoCuMnO}_6$ и никелито-купрато-манганита $\text{LaNa}_2\text{NiCuMnO}_6$

На серийном приборе LCR–800 (производство Тайвань) при рабочих частотах 1, 5 и 10 кГц в интервале 293–483 К через 10 К непрерывно в сухом воздухе исследованы температурные зависимости электроемкости, диэлектрической проницаемости, электросопротивления кобальто-купрато-манганита лантана и натрия $\text{LaNa}_2\text{CoCuMnO}_6$ и никелито-купрато-манганита лантана и натрия $\text{LaNa}_2\text{NiCuMnO}_6$. Установлено, что $\text{LaNa}_2\text{CoCuMnO}_6$ в интервале 293–483 К проявляет полупроводниковый характер проводимости. Ширина запрещенной зоны (ΔE) равна 0,54 эВ. Соединение обладает гигантскими значениями диэлектрической проницаемости, которые равны при 293 К $2,17 \cdot 10^6$ (1 кГц); $2,31 \cdot 10^5$ (5 кГц); $8,22 \cdot 10^4$ (10 кГц) и при 483 К $8,49 \cdot 10^8$ (5 кГц); $7,87 \cdot 10^7$ (10 кГц). $\text{LaNa}_2\text{NiCuMnO}_6$ в интервале 293–483 К также проявляет полупроводниковую ($\Delta E = 0,48$ эВ), при 433–443 К — металлическую и при 453–483 К — опять полупроводниковую проводимость ($\Delta E = 2,33$ эВ). Значения диэлектрической проницаемости равны при 293 К $4,97 \cdot 10^3$ (1 кГц); $9,2 \cdot 10^2$ (5 кГц); $5,1 \cdot 10^1$ (10 кГц) и при 483 К $1,02 \cdot 10^6$ (1 кГц); $1,98 \cdot 10^5$ (5 кГц) и $9,76 \cdot 10^4$ (10 кГц). Соединения можно отнести к узкозонным полупроводникам, которые представляют интерес для полупроводниковой и микроконденсаторной технологии.

Ключевые слова: электрофизика, кобальт, никелит, купрат, манганит, лантан, натрий, диэлектрическая проницаемость, электросопротивление.

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