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STUDY ON HUMIDITY SENSITIVE ELECTRICAL PROPERTIES OF BARIUM FERRITE

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Abstract

Barium ferrites, $BaFe_2O_4$, were prepared by solid state reaction method. The X-ray diffraction (XRD) method was used for qualitative phase analysis of the samples and to estimate the crystallite sizes of the samples. Scanning Electron Microscope (SEM) was used to examine the morphological features of the samples. The powders were made into circular shape pellets and their humidity sensitive electrical properties of the samples were studied in the relative humidity range of 50 RH% – 98 RH%. The sensitivities of the samples were compared with each other for humidity sensor application.

Keywords: Barium ferrites, XRD, SEM, humidity sensitive electrical properties, sensitivities

Introduction

Spinels type AB_2X_4 compounds exist in a large variety of combinations of cations (A,B) and several different anions (X) such as oxygen (O), sulphur (S), or selenium (Se). This leads to compounds with a wide range of physical properties, although they all have the same structure [Nowosielski, (2007); Pathan, (2010)]. The highly stable spinel structure allows different cations to be located on the same type of site, and owing to the site preference of the cations, many selective magnetic substitutions and various degrees of magnetic dilutions in the two sublattices may be obtained [Patil, (2013); Praveena, (2013)].

Barium ferrites are well known hard magnetic materials, which are based on iron oxide. They are also called as ferrite magnets and could not be easily replaced by any other magnets [Gul, (2007); Joshi, (2003)]. Cubic barium ferrite having the chemical formula of $BaFe_2O_4$ are widely used

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in magnetic recording media, microwave devices and electromagnetic shielding fields. Barium ferrite possesses relatively high Curie temperature and magnetic anisotropy field, as well as its excellent chemical stability and corrosion resistivity [Maria1, (2013); Rezlescu, (2002)]. In this work, Barium ferrites, $BaFe_2O_4$, were prepared by solid state reaction method. Structural and microstructural characteristics were investigated by XRD and SEM. Humidity sensitive electrical properties of the samples were also reported.

Materials and Method

Preparation of Barium Ferrites, $BaFe_2O_4$

For synthesis of Barium ferrite, $BaFe_2O_4$, the mixture of Barium Carbonate ($BaCO_3$) and Iron Oxide (Fe_2O_3) powder were used with stoichiometric composition. Analytical Reagent (AR) grade of the raw materials were used in this work. The powder sample was milled by using laboratory-made ball-milling machine for 5 h to be homogeneous and to obtain fine powders. The milling process was carried out in a rotatory mill, which generated vibrations of the balls and milled material inside the container. The weight ratio of balls to milled material was 5:1. After milling process the powders were heated at 900°C, 950°C and 1000°C for 1 h each in vacuum chamber by using thermal resistive heating coil. DELTA A Series DTA4896 and K-type thermocouple were used as the temperature controller and temperature sensor. The obtained as-prepared ferrites were reground in a rotatory mill for 3 h. Figure 1 shows the block diagram of the Barium ferrites samples preparation process. Photographs of the Barium ferrite, $BaFe_2O_4$, preparation are shown in Figures 2(a – m) respectively.

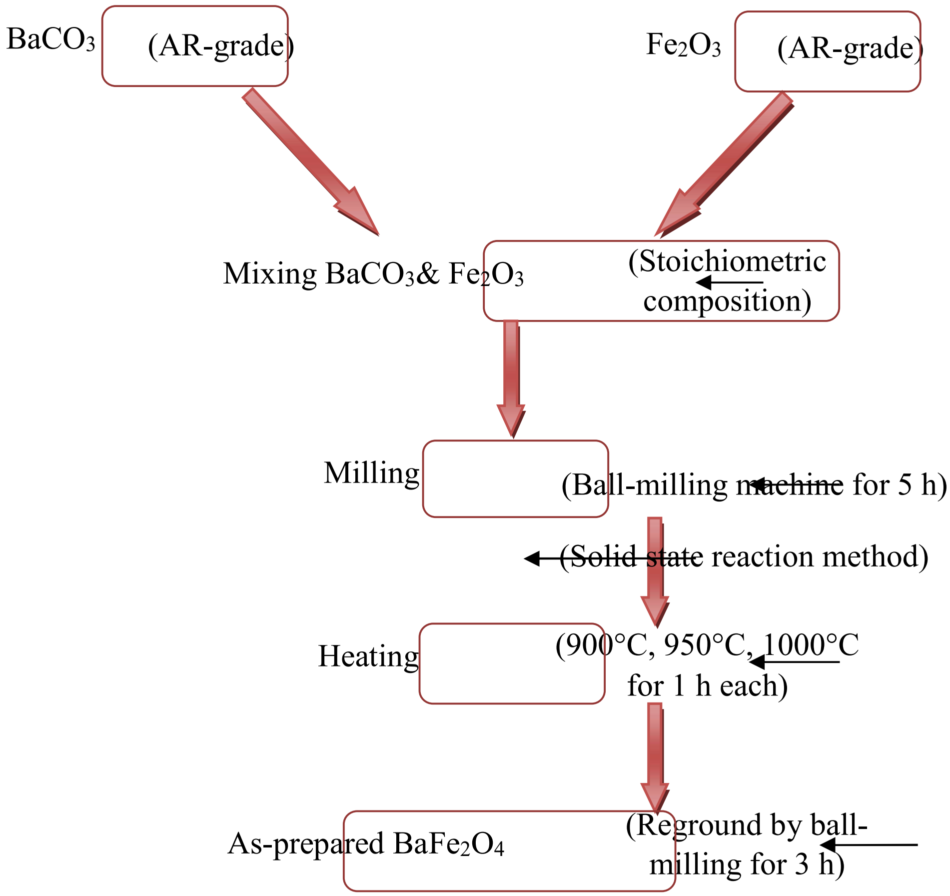

Figure1. Block diagram of the Barium ferrite sample preparation process

Figure 2.(a) Photograph of the weighed starting materials of Fe_2O_3 and $BaCO_3$

Figure2.(b) Photograph of the mixed weighed starting materials of Fe_2O_3 and $BaCO_3$

Figure 2.(c) Photograph of the uncoated stainless-steel ball

Figure 2.(d) Photograph of the plastic-coated stainless-steel ball

Figure 2.(e) Photograph of the laboratory-made ball-milling machine

Figure2.(f) Photograph of the precursor solid solution to prepare $BaFe_2O_4$

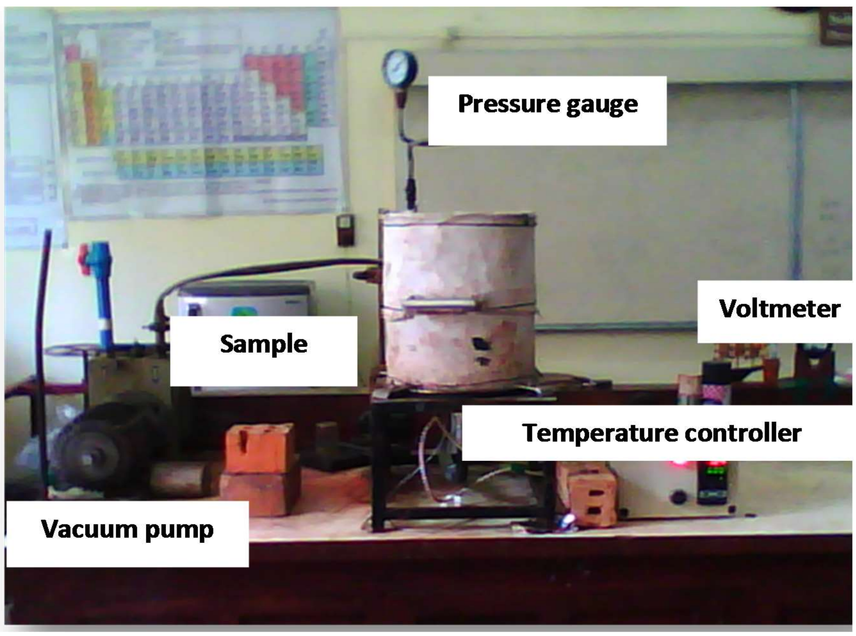

Figure 2.(g) Photograph of the experimental setup of sample preparation system

Figure2. Photographs of the (h) DELTA A Series Temperature Controller DTA4896 at 900°C and (i) as-prepared $BaFe_2O_4$ prepared at 900°C

Figure 2. Photographs of the (j) DELTA A Series Temperature Controller DTA4896 at 950°C and (k) as-prepared $BaFe_2O_4$ prepared at 950°C

Figure 2. Photographs of the (l) DELTA A Series Temperature Controller DTA4896 at 1000°C and (m) as-prepared $BaFe_2O_4$ prepared at 1000°C

XRD and SEM Measurements

The structural characteristics of the samples were investigated by RIGAKU MULTIFLEX X-ray Diffractometer using CuK_{α} radiation. The lattice parameters were calculated from XRD data using the following formula, $a = \frac{\lambda\sqrt{(h^2+k^2+l^2)}}{2\sin\theta}$ where 'a' is the lattice parameter (Å), 'λ' is the wavelength of the X-ray (Å), 'θ' is the Bragg's angle (º) and (h k l) is Miller indices. The average crystallite size of the particles was estimated from the XRD spectrum by applying Scherrer's formula, $D_{crystallite} = \frac{0.9\lambda}{B\cos\theta}$ where 'D' is the crystallitesize (nm), 'λ' is the wavelength of the X-ray (Å), 'B' is the full width at half maximum (rad) and 'θ' is the angle of diffraction (º).

Scanning electron microscopy (SEM) is a technique whereby a beam of energetically well-defined and highly focused electrons is scanned across a material (sample). Microstructural characteristics of the samples were investigated by using JEOL JSM-5610LV SEM with the accelerating voltage of 15 kV, the beam current of 50 mA and 5500 times of photo magnification.

Humidity-Sensitive Electrical Properties Measurement

Humidity sensitive electrical properties of the Barium ferrites, $BaFe_2O_4$ were prepared by conventional solid state reaction method at 900°C, 950°C and 1000°C for 1 h each were investigated in the relative humidity range of 50 RH% - 98 RH%.

Firstly, the as-prepared ferrites were made circular shape pellets by using SPECAC hydraulic press with the pressure 5 ton (~70 MPa). Then the pellet was polished by using filtered-paper to get the smoothing surface. Thickness and area of the each of the sample were as 2.95 mm 1.14×10^{-4} m^2. The sample was then fixed on glass plate and silver contacts were made over the sample to ensure good electrical contact.

In this measurement, XSW TDK 0302 Humidity Meter was used as the humidity sensing element. Humidity sensitive electrical resistance and voltage of the sample were observed by two probe method by using FLUKE

189 digital multimeter. The refrigerator (TOSHIBA) was used as the humidity generator. Photographs of the experimental setup of humidity sensitive electrical property measurement are shown in Figures3(a) and (b).

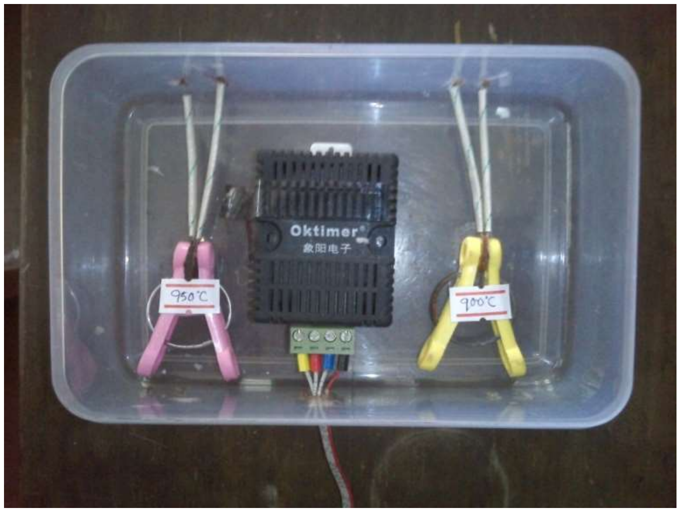

Figure 3.(a) Photograph of the sample and sensor placed in the same condition

Figure 3.(b) Photograph of the experimental setup of humidity sensitive electrical properties measurement

Results and Discussion

XRD Analysis

Powder X-ray diffraction patterns of Barium ferrites, $BaFe_2O_4$, with different preparation temperatures of 900°C, 950°C and 1000°C for 1 h each are shown in Figure4(a – c). The observed XRD lines were compared with JCPDS data library file ofCat. No. 46-0113> $BaFe_2O_4$, Barium Iron Oxide, to identify the qualitative phase analyses of powder samples. The observed XRD patterns were found to be agreed with JCPDS. The appearance of the diffraction peaks demonstrates the single-phase Barium ferrites, $BaFe_2O_4$, powders.

According to XRD patterns, $BaFe_2O_4$ belongs to cubic structure at room temperature. The lattice parameters were evaluated by using the equation $\frac{\sin^2\theta}{(h^2+k^2+l^2)}=\frac{\lambda^2}{4a^2}$. The lattice parameters of the sample are tabulated in Table 1. Comparison of the lattice parameters of the samples at different preparation temperatures are shown in Figure 5(a). The lattice parameters of the sample prepared at 950ºC was the longest and at 1000ºC was the shortest among the samples.

The crystallite sizes of each of the sample are estimated by using the Scherrer formula, $t=\frac{0.9\lambda}{B\cos\theta}$, where "*t*" is the crystallite size (nm), "λ' is wavelength (Å), "θ" is diffraction angle of the peak under consideration at FWHM (°) and "*B*" is observed FWHM (radians). In the present work, the obtained crystallite sizes are also presented in Table 5.1. Comparison of the crystallite sizes of the samples at different preparation temperatures are shown in Figure5(b). As shown in Figure 5(b), the crystallite sizes of the samples increased with increase in annealing temperature.

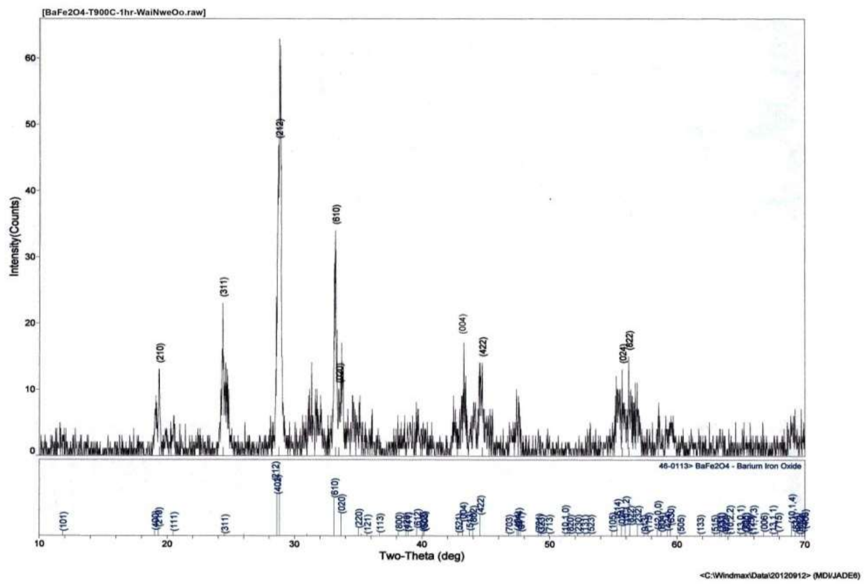

Figure 4.(a) XRD pattern of $BaFe_2O_4$ prepared at 900ºC

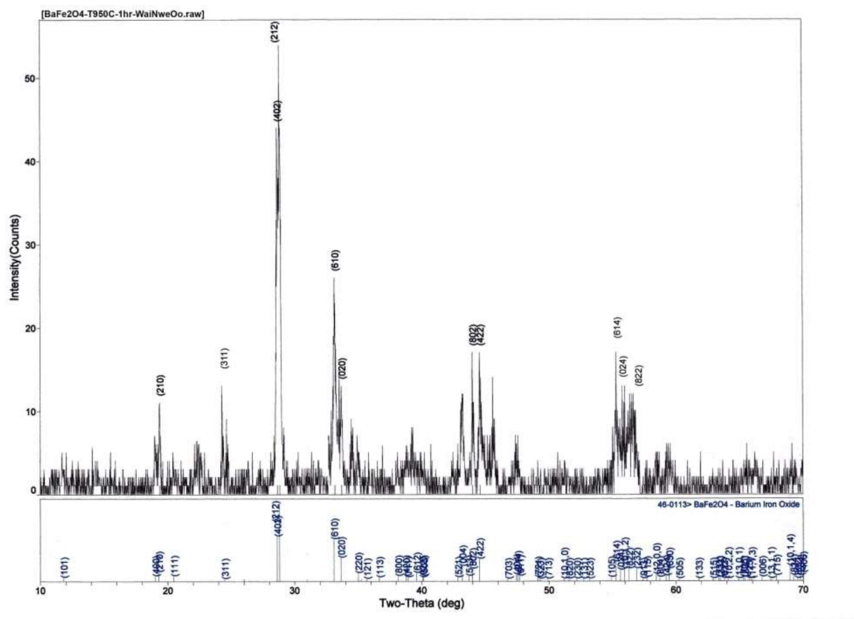

Figure 4.(b) XRD pattern of $BaFe_2O_4$ prepared at 950ºC

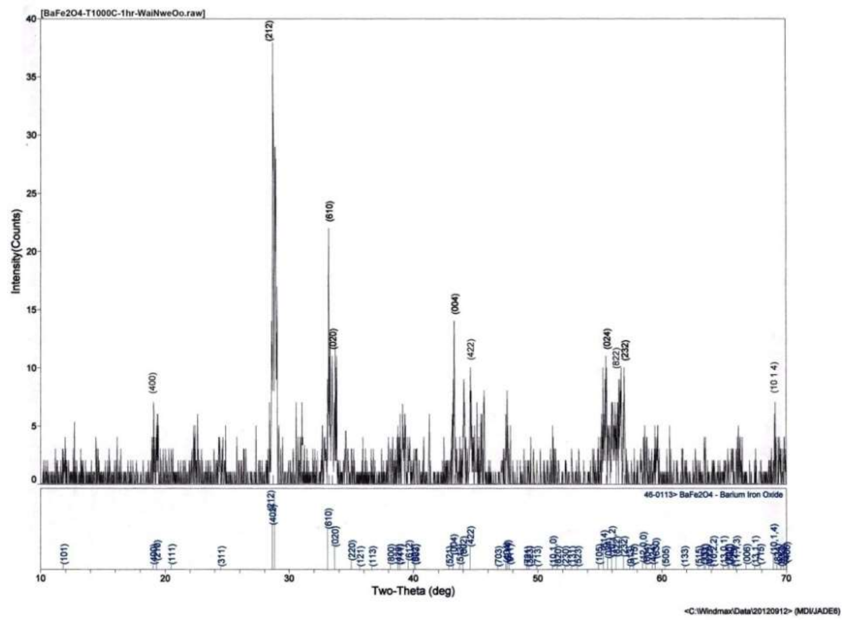

Figure 4.(c) XRD pattern of $BaFe_2O_4$ prepared at 1000ºC

Table 1. The lattice parameters and average crystallite sizes of $BaFe_2O_4$ at different preparation temperatures

Sr No	Preparation Temperature (ºC)	Lattice parameter (Å)	Crystallite size (nm)
1	900	10.5500	25.3315
2	950	12.0170	32.0367
3	1000	8.9103	53.9454

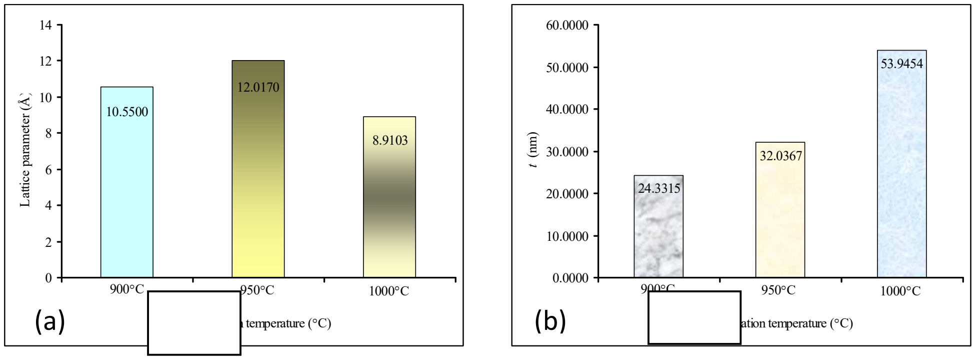

Figure 5. Comparisons of the (a)lattice parameters and (b) crystallite sizes of the $BaFe_2O_4$ with different preparation temperatures

SEMAnalysis

Material examination by SEM can yield the morphological information of the shape and size of the particles making up the object. Figures6(a – c) show the SEM images of Barium ferrite, $BaFe_2O_4$, powder of different preparation temperatures of 900ºC, 950ºC and 1000ºC.

As shown in figures, the grain shapes of the samples are uniform block and the obtained samples are found to be clear grain boundary. Some pores are also found in each of the image and these are increased with increase in preparation temperatures. The grain sizes of the samples are listed in Table 2. The largest pore area occurred in the $BaFe_2O_4$ sample prepared at the temperature of 950ºC but the smallest pore area occurred in the sample prepared at the temperature of 900ºC.

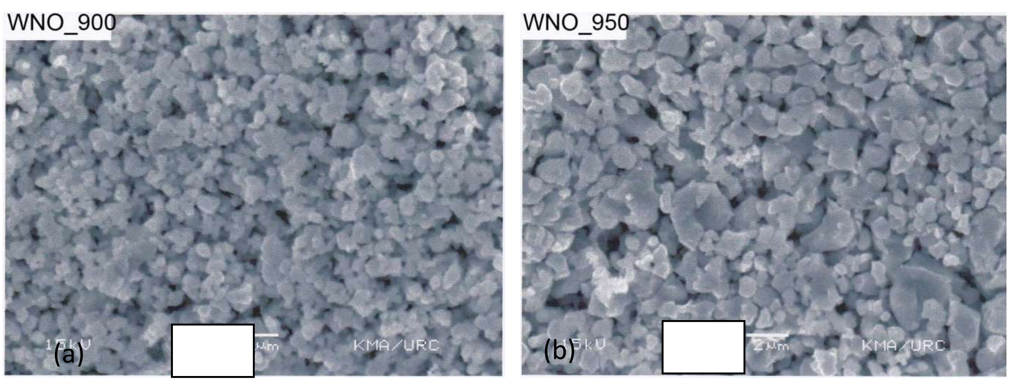

Figure 6.SEM micrographs of $BaFe_2O_4$prepared at(a) 900ºC and (b) 950ºC

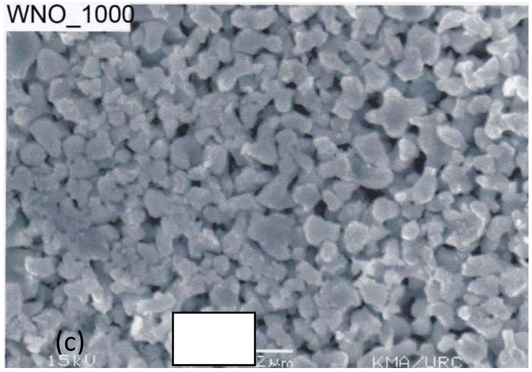

Figure 6.(c)SEM micrograph of $BaFe_2O_4$ prepared at1000ºC

Table 2. The grain sizes of the $BaFe_2O_4$ at different preparation temperatures

Sr No	Preparation Temperature (ºC)	Grain size (μm)
1	900	0.40 - 1.60
2	950	0.40 - 2.30
3	1000	0.40 – 2.20

Humidity Sensitive Electrical Properties Study

Humidity sensitive electrical resistances R_H versus relative humidity RH%, and dc voltage V_H and capacitance C_H versus relative humidity RH% of the investigated $BaFe_2O_4$prepared at three different temperatures of 900°C, 950°C and 1000°C for1 h each are shown in Figures7(a – c) and Figures 8 (a – c) respectively.

As shown in RH versus RH% graphs, the electrical resistance of the $BaFe_2O_4$sample decreased with increased in relative humidity and the obtained R_H versus RH% curves were fitted with linear type to examine the sensitivity of the sample. The slope of the R_H versus RH% graph can be taken as the sensitivity of the sample. The sensitivities of the samples are tabulated in Table 3. The sensitivity of the $BaFe_2O_4$sample prepared at 950°C is the most sensitive material among the candidate samples. The resistance changes in porous spinel type ferrites with increasing of the humidity level occur because of adsorption and capillary condensation of water [(2004). *Humidity Measuring Technology*.].

The sensitivity factor "S_f" of the sample can be evaluated by using the following relation,

$$S_f = R_{50\%}/R_{98\%}$$

where $R_{50\%}$ and $R_{98\%}$ are the electrical resistances of the $BaFe_2O_4$sample at the relative humidity 50 RH% (start point) and 98 RH% (end point) respectively. According to the above relation, for $BaFe_2O_4$ ferrite prepared at 900°C,

$$S_{f(900°C)} = R_{50\%}/R_{98\%} = 16.781\ M\Omega/12.00\ M\Omega = 1.3981$$

for $BaFe_2O_4$ferrite prepared at 950°C,

$$S_{f(950°C)} = R_{50\%}/R_{98\%} = 50.020\ M\Omega\ /17.840\ M\Omega = 2.8038$$

for $BaFe_2O_4$ ferrite prepared at 1000°C,

$$S_{f(1000°C)} = R_{50\%}/R_{98\%} = 24.945\ M\Omega\ /19.959\ M\Omega = 1.2498$$

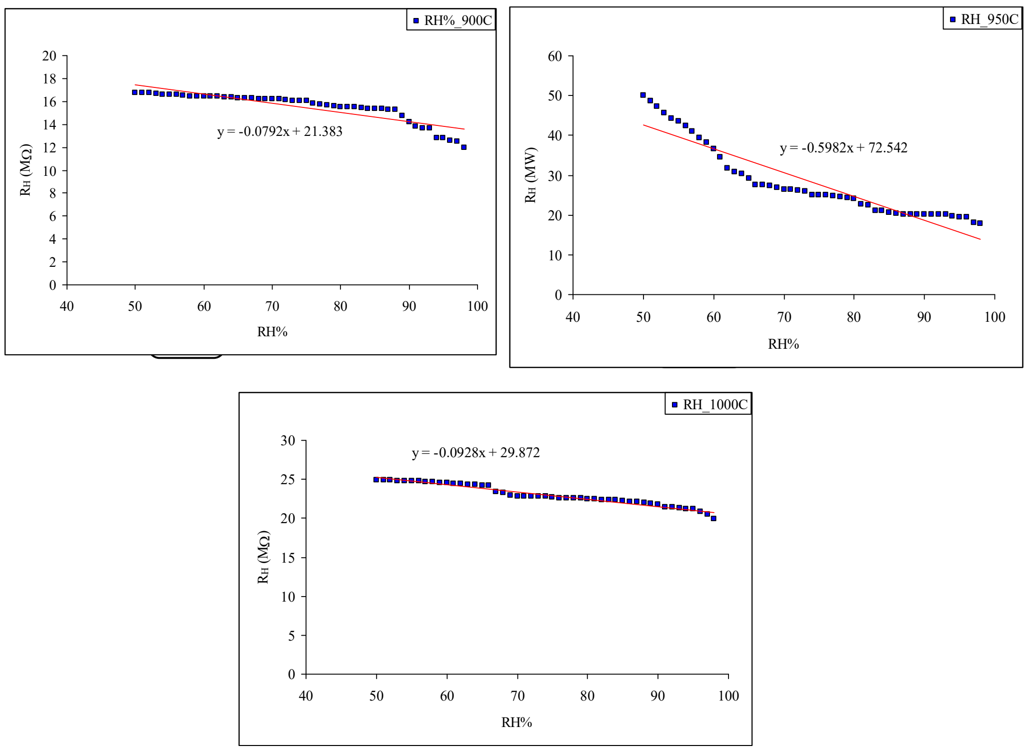

Figure 7. Humidity sensitive electrical resistances of $BaFe_2O_4$ prepared at (a) 900°C, (b) 950°C and (c) 1000°C for 1 h each

Table 3. Sensitivities and sensitivity factors of the $BaFe_2O_4$ prepared at three different preparation temperatures of 900°C, 950°C and 1000°C for 1 h each

Sr No	Sample Preparation temperature (°C)	Sensitivity (MΩ/RH%)	Sensitivity factor
1	900	0.0792	1.3981
2	950	0.5982	2.8038
3	1000	0.0928	1.2498

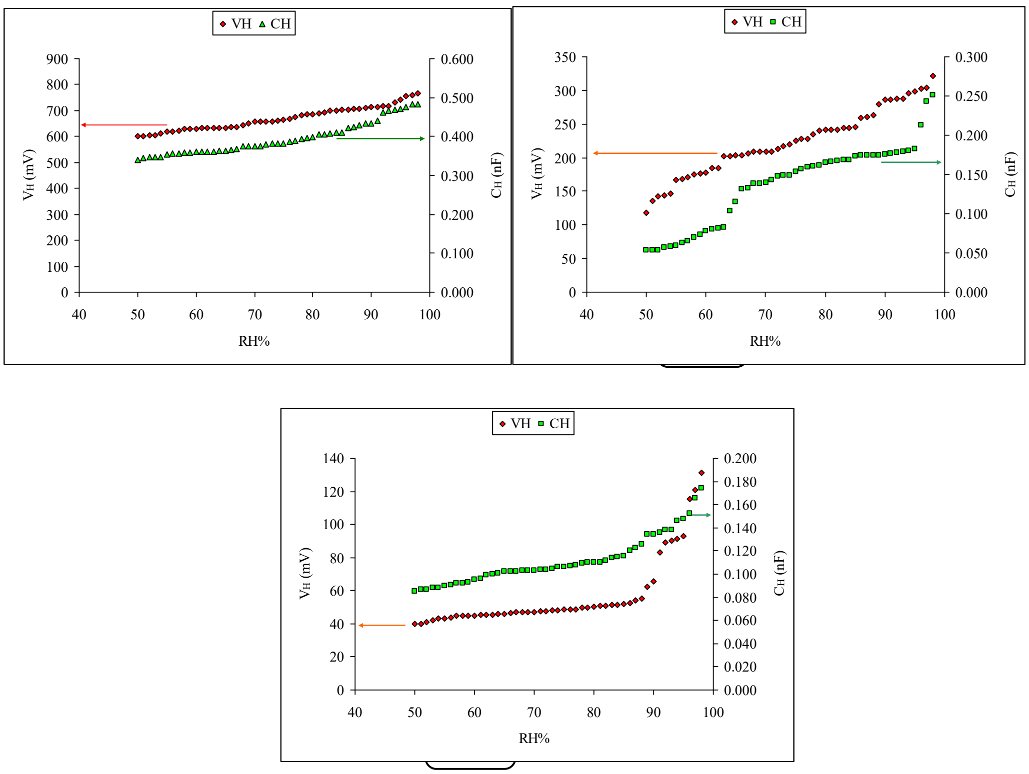

Figure 8. Humidity sensitive dc voltages and capacitances of $BaFe_2O_4$ prepared at (a) 900°C, (b) 950°C and (c) 1000°C for 1 h each

The obtained sensitivity factors are also presented in Table 3. As presented in Table 3, the sensitivity and sensitivity factor of the sample prepared at 950°C were found to be the largest one. It can be suggested that microstructure (porosity, grain size, structural defects) has a great role on the electrical resistivity. Smaller grains imply an increase of the grain boundary surface which normally account for high resistivity of a polycrystalline material. The larger the specific surface area and porosity of the specimens the more water vapors can be physically adsorbed, resulting in a larger decrease of the resistivity [(2011).*The Humidity/Moisture Handbook*.]. One can see that the sample prepared at 950°C is the most sensitive material to humidity change. As shown in Figures8(a – c), the dc voltage and capacitance of the samples increased with increase in relative humidity. Experimental data of the humidity sensitive electrical properties measurements are presented in Table 4(a – c).

Conclusion

Barium ferrites, $BaFe_2O_4$, were prepared at three different temperatures 900°C, 950°C and 1000°C for 1 h each by using solid state reaction method. The X-ray diffraction investigations of $BaCO_3$ and Fe_2O_3 annealed at 900°C, 950°C and 1000°C enabled the identification of $BaFe_2O_4$ phases. The estimated crystallite sizes were found to be increased with increase in annealing temperature and the smallest crystallite size was obtained for powders annealed at temperature of 900°C. The SEM images and distribution of powder particle size showed that the morphological features of $BaFe_2O_4$ powder particles. From the humidity sensitive electrical properties measurements, the electrical resistance decreased with increase in humidity and sensitivity of the sample also determined. The sensitivity factor was also evaluated. The sensitivity and sensitivity factor of the sample prepared at 950°C were found to be the largest one. The sensitivity and sensitivity factor were decreased with preparation temperature and thus they depend on preparation temperature. According to experimental results, Barium ferrite, $BaFe_2O_4$, samples can be used as the humidity sensors in which the sample prepared at 950°C is the most suitable than others.

Table 4.(a) Experimental data of humidity sensitive electrical properties measurement of $BaFe_2O_4$ prepared at 900°C for 1 h

RH%	**R_H(MΩ)**	**V_H(mV)**	**C_H(nF)**
50	16.781	600.000	0.341
51	16.744	600.900	0.344
52	16.738	603.300	0.346
53	16.704	605.800	0.346
54	16.617	612.300	0.346
55	16.616	619.300	0.353
56	16.592	619.300	0.356
57	16.510	623.300	0.356
58	16.482	627.600	0.358
59	16.482	628.000	0.358
60	16.481	630.300	0.360
61	16.439	631.200	0.361
62	16.428	631.400	0.361
63	16.397	633.400	0.362
64	16.343	633.400	0.363
65	16.300	634.200	0.364
66	16.296	635.800	0.366
67	16.287	636.700	0.369
68	16.236	643.600	0.374
69	16.202	650.500	0.375
70	16.198	655.700	0.376
71	16.196	656.900	0.376
72	16.123	657.300	0.379
73	16.064	658.400	0.381
74	16.045	661.000	0.382
75	16.038	663.000	0.383
76	15.837	669.500	0.387
77	15.760	673.300	0.390
78	15.690	682.300	0.394
79	15.591	684.900	0.396

RH%	R_H(MΩ)	V_H(mV)	C_H(nF)
80	15.525	686.800	0.398
81	15.514	688.000	0.405
82	15.485	691.900	0.406
83	15.420	698.100	0.408
84	15.395	698.500	0.409
85	15.372	702.700	0.411
86	15.338	702.900	0.422
87	15.302	705.400	0.424

Table 4.(a) (Continue) Experimental data of humidity sensitive electrical properties measurement of $BaFe_2O_4$ prepared at 900°C for 1 h

RH%	R_H (MΩ)	V_H (mV)	C_H (nF)
88	15.259	706.300	0.429
89	14.756	711.000	0.433
90	14.211	712.000	0.433
91	13.814	715.300	0.441
92	13.670	716.900	0.462
93	13.636	718.100	0.467
94	12.807	733.000	0.469
95	12.781	740.800	0.471
96	12.614	755.500	0.475
97	12.481	758.900	0.482
98	12.003	767.000	0.484

Table 4.(b) Experimental data of humidity sensitive electrical properties measurement of $BaFe_2O_4$ prepared at 950°C for 1 h

RH%	R_H(MΩ)	V_H(mV)	C_H(nF)
50	50.020	117.370	0.053
51	48.700	135.300	0.053
52	47.200	142.240	0.054
53	45.600	143.570	0.057
54	44.300	147.000	0.058
55	43.600	166.970	0.059
56	42.400	168.040	0.063
57	41.100	170.500	0.065
58	39.400	174.810	0.070
59	38.200	176.010	0.073
60	36.600	178.020	0.078
61	34.500	184.130	0.080
62	31.800	184.910	0.081
63	30.900	201.580	0.082
64	30.400	202.040	0.103
65	29.100	203.020	0.115
66	27.569	203.950	0.131
67	27.559	206.500	0.132
68	27.404	208.410	0.138
69	26.833	208.440	0.138
70	26.524	208.790	0.140
71	26.457	209.110	0.143
72	26.277	213.100	0.148
73	26.046	217.530	0.149
74	25.065	220.030	0.149
75	25.041	224.660	0.153
76	24.966	228.150	0.157
77	24.821	228.310	0.159
78	24.511	234.450	0.161
79	24.373	240.040	0.162

RH%	R_H(MΩ)	V_H(mV)	C_H(nF)
80	24.006	241.010	0.165
81	22.651	241.030	0.166
82	22.541	241.110	0.167
83	21.135	244.620	0.169
84	21.020	244.640	0.169
85	20.538	245.400	0.173
86	20.493	258.700	0.174
87	20.250	260.270	0.175

Table 4.(b) (Continue) Experimental data of humidity sensitive electrical properties measurement of $BaFe_2O_4$ prepared at 950°C for 1 h

RH%	R_H(MΩ)	V_H(mV)	C_H(nF)
88	20.249	263.410	0.175
89	20.173	279.890	0.175
90	20.151	285.840	0.176
91	20.147	286.720	0.177
92	20.090	287.460	0.178
93	20.074	287.740	0.179
94	19.791	296.130	0.180
95	19.446	298.430	0.183
96	19.359	302.010	0.213
97	18.180	304.380	0.243
98	17.840	321.760	0.251
99	17.583	323.570	0.277

Table 4.(c) Experimental data of humidity sensitive electrical properties measurement of $BaFe_2O_4$ prepared at 1000°C for 1 h

RH%	**R_H(MΩ)**	**V_H(mV)**	**C_H(nF)**
50	24.945	40.000	0.085
51	24.921	40.113	0.087
52	24.890	40.846	0.087
53	24.767	41.880	0.088
54	24.760	43.196	0.088
55	24.758	43.256	0.090
56	24.756	43.512	0.091
57	24.706	44.855	0.092
58	24.672	44.908	0.092
59	24.589	44.997	0.093
60	24.573	45.030	0.095
61	24.483	45.233	0.096
62	24.463	45.532	0.099
63	24.330	45.573	0.100
64	24.324	45.948	0.101
65	24.265	46.090	0.102
66	24.258	46.390	0.102
67	23.365	47.090	0.102
68	23.248	47.112	0.103
69	22.987	47.191	0.103
70	22.875	47.227	0.103
71	22.799	47.362	0.104
72	22.782	47.545	0.104
73	22.764	48.320	0.105
74	22.761	48.359	0.106
75	22.755	48.545	0.106
76	22.643	48.563	0.107
77	22.613	48.579	0.108
78	22.595	49.600	0.109
79	22.553	49.920	0.110

RH%	**R_H(MΩ)**	**V_H(mV)**	**C_H(nF)**
80	22.462	50.107	0.110
81	22.450	50.864	0.110
82	22.397	50.880	0.112
83	22.389	51.231	0.114
84	22.384	51.337	0.115
85	22.290	51.888	0.116
86	22.119	52.589	0.120
87	22.091	54.012	0.123

Table 4.(c) (Continue) Experimental data of humidity sensitive electrical properties measurement of $BaFe_2O_4$ prepared at 1000°C for 1 h

RH%	**R_H(MΩ)**	**V_H(mV)**	**C_H(nF)**
88	22.038	55.200	0.126
89	21.940	62.080	0.134
90	21.721	65.500	0.134
91	21.438	83.000	0.136
92	21.371	88.900	0.138
93	21.305	90.300	0.138
94	21.181	91.400	0.146
95	21.150	93.200	0.148
96	20.848	115.200	0.152
97	20.550	120.900	0.166
98	19.959	131.200	0.174

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