

## **FABRICATION AND PHYSICAL PROPERTIES OF NiMn<sub>2</sub>O<sub>4</sub>, ZnO+0.02TiO<sub>2</sub> AND FeNbO<sub>4</sub> AS THERMOELECTRIC, HEATING ELEMENT AND NEGATIVE TEMPERATURE COEFFICIENT MATERIALS**

**Thongchai PANMATARITH\*, Preeyanan THEPNUAN, Rudeerat SUNTAKO  
and Chiraporn SRIPROM**

**Department of Physics, Faculty of Science, Prince of Songkla University,  
Hat Yai 90112, THAILAND**

### **ABSTRACT**

This present work was undertaken as a study to clarify the thermoelectric, heating and negative temperature coefficient effects of NiMn<sub>2</sub>O<sub>4</sub>, ZnO+0.02 TiO<sub>2</sub> and FeNbO<sub>4</sub>. The composition of the samples before firing were NiO+MnO<sub>2</sub>, ZnO+0.02TiO<sub>2</sub> and Fe<sub>2</sub>O<sub>3</sub>+Nb<sub>2</sub>O<sub>5</sub>. After firing, the first sample was a single phase material (NiMn<sub>2</sub>O<sub>4</sub>), the second sample was mixed two phase material (ZnO, ZnTiO<sub>4</sub>) and the third sample was a single phase material (FeNbO<sub>4</sub>). The prepared NiO+MnO<sub>2</sub> and FeNbO<sub>4</sub> resistances were 18.65 kΩ and 5.9 kΩ, respectively. The NiO+MnO<sub>2</sub> material shows the thermoelectric effect. The thermoelectric voltage versus temperature relation corresponds to the equation,  $V = 0.0028T^2 - 0.4342T + 14.304$  with  $R^2 = 0.9953$ . So, the sample was an n-type material. The computer interfacing circuit which has been made can exhibit the picture of the thermoelectric versus time relation. This sample was investigated for obtaining the type of the electrical carriers in ceramics. The temperature versus supplied electric power relation of the ZnO+0.02TiO<sub>2</sub> material corresponds to the equation,  $T = -1 \times 10^{-6}P^2 + 0.0335P - 10.531$ , with  $R^2 = 0.9892$ . This sample can show the electricity to heat conversion very well. The sample shows the heating effect. The picture of temperature versus time can be shown with the computer interfacing circuit system. This material was studied for the heating element for the electric furnace. The sample resistance versus the temperature relation of the Fe<sub>2</sub>O<sub>3</sub>+Nb<sub>2</sub>O<sub>5</sub> material corresponds to the equation,  $R = 5.4961e^{(-0.0173T)}$ , with  $R^2 = 0.9945$ . The negative temperature coefficient of the resistance ( $\alpha$ ) was  $-1.03 \text{ } \%/^{\circ}\text{C}$  from 25 to 100°C. and  $-0.84 \text{ } \%/^{\circ}\text{C}$  from 100 to 200°C. So, the sample shows the NTC effect. The sample was an n-type semiconducting ceramic. After the calibration by the measurement of the temperature (T) versus voltage drop (V) T, a comparison between the true temperature (T<sub>true</sub>) from commercial apparatus and the measure temperature (T<sub>measure</sub>) from the computer with the prepared sample for determining the accuracy from 24 to 200°C was made. So, the prepared sample can be used as temperature sensor with computer display. The testing system for temperature measurement and control is composed of a prepared sample as a temperature sensor, solid state relay for control and the computer as a display device. After the furnace was heated and then the furnace temperature was increased to a setting temperature, the computer will control the temperature at constant temperature at a given interval.

**Keywords :** Thermoelectric effect, Heating effect and NTC effect.

---

\* To whom correspondence should be addressed:

E-mail : [pthongch@ratree.psu.ac.th](mailto:pthongch@ratree.psu.ac.th) Tel: (661) 969 3081

## INTRODUCTION

Today, ceramic materials are used extensively. Each types of ceramics shows different properties and phenomena. The interesting effects are studied such as the thermoelectric effect, the heating effect and the negative coefficient effect (NTC effect). So, it is necessary to study the preparation techniques, testing techniques and instrument construction for beneficial purposes (Buchanan, 1991). The studying of the ceramics is used for learning and industrial work. The thermoelectric effect can be used to study the type of the carrier in the material and the thermal response device, the heating effect can be used as a heating element and the NTC effect can be used as a temperature sensor and temperature control.

The thermoelectric effect or Seebeck effect is a phenomenon about the electricity and heat. Thermoelectric generation is the heat to electrical energy conversion. The efficiency of

$$Z = \sigma^2 \alpha / k \dots \dots \dots (1)$$

the thermoelectric material (Z) is  $\alpha$  is the Seebeck coefficient,  $\sigma$  is the electric conductivity and k is the thermal conductivity. For good thermoelectric material, Z,  $\alpha$  and  $\sigma$  are high values. But k is a low value. The Z value of the material for a thermoelectric generator will be high. The high temperature thermoelectric materials were FeSi<sub>2</sub>, CrSi<sub>2</sub>, SiC and ZnO. Ravinder (1994) had prepared Mn-Zn ferrite with the formula of Mn<sub>x</sub>Zn<sub>1-x</sub>Fe<sub>2</sub>O<sub>4</sub> (x=0.2, 0.6, 0.8) which was fired at 1,200°C. Thermo-emf was measured at a different temperature and then the Seebeck coefficient was calculated. The Seebeck coefficient ( $\alpha$ ) was decreased as temperature (T) increasing and the Seebeck coefficient was also measured for different compositions.

The heating effect is the phenomenon that when electricity was applied the material was heated. But a heating element is material that can convert electrical to thermal energy very well. The heating effect was found in the heating element. An important property is the

sample temperature versus electrical supplied power relation (T vs P). The heating element was used as a furnace element. The heating elements are composed of the two groups of materials (Moulson and Herbert, 1990).

- 1) A lowly resistive element which was low voltage power supply.
- 2) A highly resistive element which uses high voltage power supply.

The NTC effect is the phenomenon that material resistance was decreased as temperature increased due to the effect of intrinsic characteristics in which the sample shows the NTC effect very well. The resistance of this material is not changed as temperature is not changed at room temperature. The NTC thermistor is the semiconductor with a negative temperature coefficient of resistance (NTCR,  $\alpha$ ). The electrical property of temperature sensor corresponds to where,  $\rho$  is the resistivity, R is

$$\rho = RA/L \dots \dots \dots (2)$$

resistance, A is the effective area and L is the thickness of the material. The resistivity ( $\rho$ ) versus temperature relation corresponds to

$$\rho(T) = \rho \alpha \exp(B/T) \dots \dots \dots (3)$$

where,  $\rho(T)$  is the resistivity at temperature  $\rho \alpha$  is a constant which is independent to temperature and B is the constant which is involved with the energy for electron conduction. (Moulson and Herbert, 1990). The NTC thermistor property can be used as a temperature sensor, radiation sensor, vacuum gauge, pressure gauge and temperature compensation sensor (Buchanan, 1991).

Jiti Nukuaw (1991) had suggested temperature control with a computer. Ravinder (1994) had prepared Mn-Zn ferrite, measured the dependence of the Seebeck coefficient on temperature. Das Gupta (1996) and reported the method of application MoSi<sub>2</sub>, SiC and graphite material to be heating elements. F.A.S Soliman (1993) had studied the thermistor which was prepared from the composition of NiO+Mn<sub>2</sub>O<sub>3</sub>. The resistance versus temperature relation was measured.

*Fabrication and Physical Properties of  $NiMn_2O_4$ ,  $ZnO + 0.02TiO_2$  and  $FeNbO_4$  as Thermoelectric, Heating Element and Negative Temperature Coefficient Materials*

In this current study, the samples with formulas  $NiO + MnO_2$ ,  $ZnO + 0.02TiO_2$  and  $Fe_2O_3 + Nb_2O_5$  were prepared with the standard ceramic technique. Sample phases were identified for studying the phase of the materials. The sample sizes were measured and electrode. Thermoelectric, heating and the NTC effect were tested and applied.

## EXPERIMENTAL PROCEDURE

### *Sample preparation, phase identification, size measurement and electroding.*

The composition of the samples having the chemical formula  $NiO + MnO_2$ ,  $ZnO + 0.02TiO_2$  and  $Fe_2O_3 + Nb_2O_5$  were selected for this investigation. Commercial-grade powders (99.5% purity) were calculated and weighed. The powders were mixed in mortar and mixer. The mixture of PVA (polyvinyl alcohol) and distilled water were added to the mixed powder as a binder. Then, the powders were pressed with 5 tons force into small discs with a hydraulic press (CARVER, 973110A). The small disc samples were fired in an electric furnace with a temperature controller (FCR-13A-R/M) and type-R thermocouple at  $\sim 500$  (1200),  $\sim 1150$  and  $\sim 1200^\circ C$  with  $\sim 5^\circ C/min$  for  $\sim 1$  h, respectively. After, the furnace switch was closed. The samples were cooled naturally in air medium. The as-fired samples were removed from it.

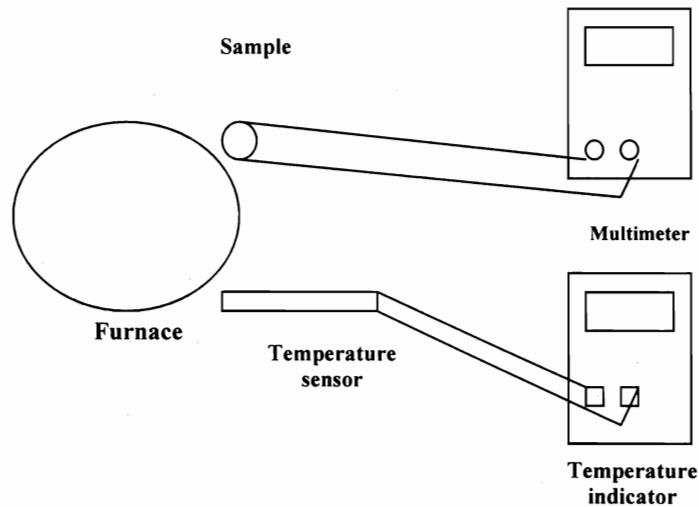
The phase of the samples were identified with XRD technique using a diffractometer (the Philips PW3710). X-rays were incident on the sample powder and moved through an interplanar spacing. The diffracted x-rays were detected with an x-ray detector. This detector will transform x-ray intensity into electric signals and then be

amplified. The amplified signal was transformed to be a square shape and sent to a computer. The computer will write the x-ray diffraction patterns. This pattern shows the phase of this sample. The intensity of the peak of the plane will appear at a different Bragg angle for lattice constant determination. The crystal structure of prepared materials will be different. Miller indices which are used for plane indication were studied. The XRD patterns were studied by Bragg law. This technique is an analysis of the XRD patterns between the prepared samples and the standard materials for determining the unknown materials.

The micrometer was used to determine the sample sizes. The prepared sample surfaces were polished with sand paper or polisher until the surface was smooth. The samples were electrode with silver paste (conductive epoxy CW2400) and electric wire by smearing on the surface and on the electric wire. This was fired at  $120^\circ C$  for 20 min and then cooled to room temperature.

### *Thermoelectric effect test of the $NiO + MnO_2$ material.*

Firstly, thermoelectric voltage versus temperature was measured. The experimental setup was shown in Figure 1. The sample and K-type thermocouple was held with the stand above the furnace at about 0.5 cm. The sample was heated slowly from 25 to  $55^\circ C$ . The thermoelectric voltage (V) was measured with a multimeter (Fluke 45). The sample temperature (T) was measured with a temperature apparatus (ADV M890C<sup>+</sup>). The V and T values were plotted.



**Figure 1** The resistance versus temperature measurement for the NiO+MnO<sub>2</sub> material.

Secondly, thermoelectric voltage versus time was displayed with the following step :

1) The computer interfacing circuit was designed and constructed for the thermoelectric test in Figure 2

2) The thermoelectric versus time of the sample was studied by attaching a computer through an ET-PC 8255 card to interfacing circuit for automatic data acquisition and analysis over the given temperature range. This interfacing circuit was tested with 5 V d.c. voltage by adjusting the VR 10 k $\Omega$  to apply the voltage to 26 pin (I<sub>o</sub>) of ADC0809. This IC will convert the analog voltage (AV) to digital voltage (DV). This DV was sent to 74LS244 for buffering. The output DV of this buffer was indicated with 8 LEDs. When adjusted the VR 10 k $\Omega$ , the LEDs will be bright and dark. If the voltage was low (0 V), the LED will be bright. If the voltage is high (5 V), the LED will be dark. The amplification circuit was constructed for amplifying the voltage from the thermoelectric sample. This circuit was tested by applying +9V to pin 7 and -9V to pin 4 and the ground electrode was connected to the ground of the circuit. Then, it was tested for amplification with a signal generator. The resistors of 160 k $\Omega$  and 1 M $\Omega$  were used for determining the gain. The reason for

amplification is because of the low voltage (mV) of the signal.

3) The method of thermoelectric versus time displaying with computer

- furnace preparation
- set the thermoelectric material at input of the Op amp
- connects Op amp with ADC 0809 and 74LS244
- connects 74LS244 with ET-PC 8255 Card
- set ET-PC 8255 Card into slot of computer
- open the computer to DOS
- goto C:\>cd TP and then Enter
- goto C:\TP>turbo and then Enter
- goto Turbo Pascal Program
- writes the program for thermoelectric (V) versus time (t) with computer
- press Ctrl+F9 to run program
- press Ctrl + Break to Exit program
- connect furnace electric line to electric plug. The sample surface was heated, it then generated the thermoelectric voltage.
- apply this voltage to input (pin 3) of Op Amp I for amplification and to input of Op Amp II for amplification again. The amplified voltage was sent to I<sub>o</sub> (pin 26) of ADC 0809. This voltage was called analog voltage (AV).

*Fabrication and Physical Properties of  $\text{NiMn}_2\text{O}_4$ ,  $\text{ZnO}+0.02\text{TiO}_2$  and  $\text{FeNbO}_4$   
as Thermoelectric, Heating Element and Negative Temperature Coefficient Materials*

- apply this AV to ADC 0809 (analog to digital converter) for converse analog voltage (AV) to digital voltage (DV). This digital voltage is 8 bits data ( $D_7 D_6 D_5 D_4 D_3 D_2 D_1 D_0$ ). This data was sent to 74LS244 as a buffer. Eight LEDs were used for this data indication and then sent to port A of ET- PC 8255 card. This DV was sent to the RAM.  $AV=(5/255)*DV$  was used for DV to AV conversion.

- lineto (x, y) instruction was used for showing the thermoelectric voltage versus time (V vs. T) and delay instruction was used to define the period of time.

- print the thermoelectric voltage (V) versus time (t) graph with the computer with the following step :

- goto C:\>cd dos
- goto C:\DOS>graphics.com
- goto C:\>cd TP and then Enter
- goto C:\TP>turbo and then Enter
- goto Turbo Pascal Program
- RUN the program to display V vs T

graph

- press Shift + Print Screen to print the V vs T graph

Program for thermoelectric voltage versus time displaying with computer

Program Thermoelectric\_Voltage\_vs\_Time\_Graph;  
uses crt, graph;

```
var grdrv, grmode, grerror   : integer;
    ch                       : char;
    DV                       : integer;
```

```
const PA      = $0304;
      Pcontrol = $0307;
```

procedure axis;

```
var p,q : integer;
    tex : string;
```

begin

```
  grdrv:=detect ; initgraph(grdrv,grmode,'c:\tp\bgi');
  setgraphmode(grmode);
  line(50,50,50,305) ; line(50,305,600,305);
  line(50,50,600,50) ; line(600,50,600,305);
  settextstyle(defaultfont , horizdir,0);
  for p := 50 to 600 do
  begin
```

```
    if p mod 32 = 0 then
```

```
  begin
```

```
    line(p+18, 295, p+18, 305);
```

```
    str(round(p/32-1),tex);
```

```
    outtextxy(p+18, 320, tex);
```

```
  end;
```

```
end;
```

```
settextstyle(defaultfont , horizdir,0);
```

```
for q:= 50 to 305 do
```

```
begin
```

```
  if q mod 51 = 0 then
```

```
  begin
```

```
    line(45,q,55,q);str(( (305-q)mod5)+1, tex);
```

```
    outtextxy(20,q,tex)
```

```
  end;
```

```
end;
```

```
procedure plot ;
```

```
var i, j, x, y, DV      : integer;
```

```
    AV                  : real;
```

```
begin
```

```
  outtextxy(190,10, THERMOELECTRIC  
VOLTAGE VS TIME GRAPH );
```

```
  outtextxy(190,18, ----- );
```

```
  outtextxy(50,30, 'Thermoelectric voltage (V)');
```

```
  outtextxy(540,340, 'Time (s)');
```

```
  outtextxy(48,303, '*');
```

```
begin
```

```
  DV := 0; AV:= 0;
```

```
  port[Pcontrol ]:= $90;
```

```
  for j:=0 to 550 do
```

```
  begin
```

```
    DV:=port[PA];
```

```
    AV:=(5/255)*DV;
```

```
    x:=j+50 ; y:=305-DV*5;
```

```
    lineto(x,y);
```

```
    delay(100)
```

```
  end;
```

```
end;
```

```
readln;
```

```
closegraph;
```

```
end;
```

```
begin (main)
```

```
  repeat
```

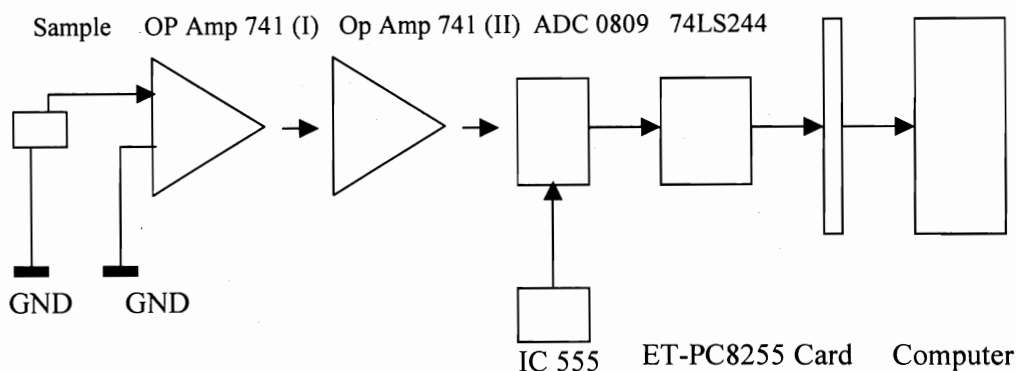
```
    axis;
```

```
    plot;
```

```
    ch:=readkey;
```

```
    until ord(ch) = 27;
```

```
end.
```

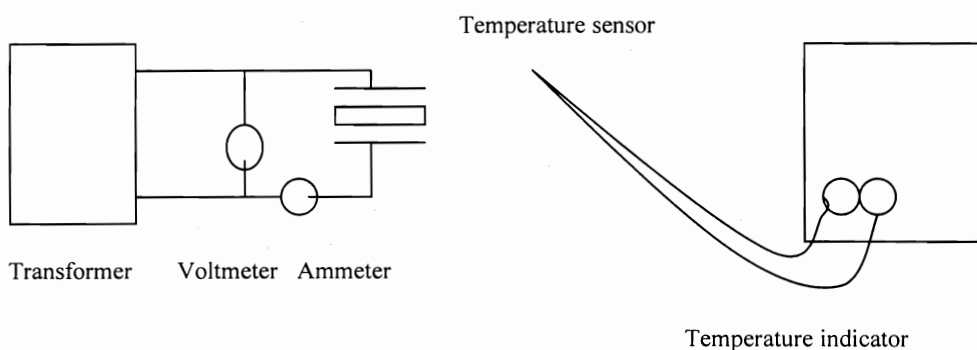


**Figure 2** Block diagram of thermoelectric study with computer for the NiO+MnO<sub>2</sub> material.

### Heating effect test of the ZnO+0.02TiO<sub>2</sub> material

Firstly, the electric current (I) from the transformer was flowed through the sample, then this sample received the electric voltage (V) and sample temperature (T) was increased as shown in Figure 3. The electric current was measured with a multimeter (Fluke 87), the

electric drop voltage was measured with multimeter (Fluke 45) and the sample temperature with temperature apparatus (AVD M890C<sup>+</sup>) with the type K thermocouple as a temperature sensor. The supplied electric power (P) was calculated from the equation,  $P = IV$ . The relation of sample temperature versus supplied electric power was plotted.



**Figure 3** Heating effect test for the ZnO+0.02TiO<sub>2</sub> material

Secondly, the computer interfacing circuit was designed and constructed for a heating test as shown in Figure 4 and followed the steps :

1) Computer interfacing for ceramic heating element temperature measurement was tested until useful.

2) Write the program for the computer displaying the temperature versus time graph and then tested.

### Program for computer displaying the temperature versus time

```

Program
Heating_Element_Temperature_vs_Time_Graph;
uses crt, graph;
var grdrv, grmode, gerror   : integer;
    ch                       : char;
    DV                       : integer;
    const PA                 = $0304;
        Pcontrol = $0307;
procedure axis;

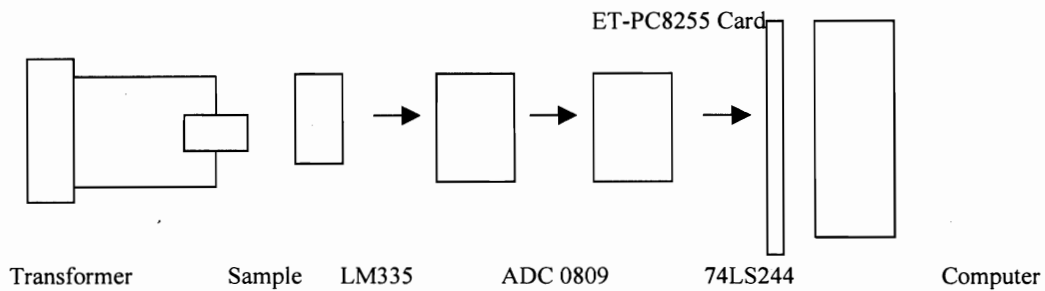
```

*Fabrication and Physical Properties of  $Ni_{0.9}Mn_{0.1}O_4$ ,  $Zn_{0.9}O_{0.1}TiO_2$  and  $Fenbo_4$   
as Thermoelectric, Heating Element and Negative Temperature Coefficient Materials*

```

var p,q : integer;
    tex : string;
begin
    grdrv:=detect ;
initgraph(grdrv,grmode,'c:\tp\bgi');
    setgraphmode(grmode);
    line(50,50,50,305) ; line(50,305,600,305);
    line(50,50,600,50) ; line(600,50,600,305);
    settextstyle(defaultfont , horizdir,0);
    for p := 50 to 600 do
    begin
        if p mod 110 = 0 then
            begin
                line(p+50, 295, p+50, 305);
str(round(p/110),tex);
                outtextxy(p+50, 320, tex);
            end;
        end;
        settextstyle(defaultfont , horizdir,0);
        for q:= 50 to 305 do
            if q mod 51 = 0 then
                begin
                    line(45,q,55,q) ; str((((305-q) mod
5)+1)*20, tex);
                    outtextxy(20,q,tex)
                end;
            end;
        procedure plot ;
        var i, j, x, y, DV : integer;
            AV, VT, T : real;
        begin
            outtextxy(235,10, TEMPERATURE VS
TIME OF HEATING ELEMENT GRAPH );
            outtextxy(235,18, ----- );
            outtextxy(50,30, Temperature (°C));
            outtextxy(540,340,Time (min));
            outtextxy(48,303,'*');
            begin
                DV := 0; AV:= 0;
                port [Pcontrol ]:=$90;
                for j:=0 to 550 do
                    begin
                        DV:=port[PA];
                        AV:=(5/255)*DV;
                        VT:=AV;
                        T:=(VT-2.73)/(0.01);
                        x:=j+50 ; y:=305-round((255/100)*T);
                        lineto(x,y);
                        delay(600)
                    end;
                end;
                readln;
                closegraph;
            end;
        begin (main)
            repeat
                axis;
                plot;
                ch:=readkey;
                until ord(ch) = 27;
            end.
        end.
    end;
    3) The electric current was applied
    from the transformer through the heating
    element sample for electricity to heat
    conversion.
    4) The sample temperature was
    measured with LM335 which was set on the
    surface of the sample. LM335 is the
    temperature sensor that is made from a
    semiconductor.
    5) The electric voltage from this
    sensor was sent to pin 26 (Io) of ADC0809 for
    analog voltage (AV) to digital voltage (DV)
    conversion. This 8 bits digital voltage which
    is composed of D7, D6, D5, D4, D2, D1, D0
    was sent to 74LS244 for buffering. This
    voltage was sent to the ET-PC8255 Card and
    through the port A of IC8255 and then to the
    RAM. The DV was displayed on the screen.
    The DV was convert to AV and displayed on
    the screen.
    6) The AV was convert to temperature
    (T).
    7) So, the computer can read the
    heating element temperature with the LM335
    temperature sensor.

```



**Figure 4** Block diagram for computer displaying the heating effect in the  $\text{ZnO}+0.02\text{TiO}_2$  material.

#### ***NTC effect test of the $\text{Fe}_2\text{O}_3+\text{Nb}_2\text{O}_5$ material***

Firstly, the resistance-temperature characteristics were measured over the temperature range  $25^\circ\text{C}$  to  $200^\circ\text{C}$  with the same experimental setup as Figure 1, but the direct voltage scale was changed to be the resistance scale. The measure resistance (R) versus temperature (T) was plotted. The negative temperature of the resistance ( $\infty$ ) of this material was calculated from this data.

Secondly, the computer interfacing circuit for the temperature measurement and control was designed and constructed as shown in Figure 5. The experimental steps were:

1) The sample was tested for temperature sensor function with this circuit. The electric voltage was applied to the circuit for testing. If the circuit can be operated, the LED can be bright or dark by as adjusting the VR10k $\Omega$ .

2) Write the OUTPUT Program with Turbo Pascal for sending the electric voltage out of port B. If the circuit operated completely, the LED can be bright and dark for 5 V and 0 V, alternately. The electrical voltage was moved from the computer with Port[PB]:=0 or Port[PB]:=255.

3) Write INPUT program with Turbo Pascal for computer reading the electric voltage from the interfacing circuit and then sent to port A. When the electric voltage dropped from VR10 k $\Omega$  was adjusted and applied to pin 26 ( $I_0$ ) of ADC0809 for analog voltage (AV) to digital voltage (DV)

conversion. The IC555 circuit will generate square shaped electric voltage and send to pin 10 of ADC0809 for operating. The 8 bit digital voltage (D7, D6, D5, D4, D2, D1, D0) was sent through 74LS244 for buffering. The 8 LED will indicate the DV at an output of 74LS244. The DV as sent through port A of IC8255 of the ET-PC8255 Card and the sent to RAM. The DV was sent to the computer with DV:=Port[PA]. The DV, AV and T were displayed on the screen with writeln(' ').

4) The computer was tested for reading temperature with the prepared sample as temperature sensor. The 5 V d.c. electric voltage was applied to the prepared sample in a series with a 100 k resistor  $\Omega$ . The voltage drop on the resistor was sent to pin 26 ( $I_0$ ) of ADC0809 for AV to DV conversion and sent through 74LS244 and ET-PC8255 Card into the RAM. The DV was displayed on the screen. The DV was converted to AV with AV:=(5/255)\*DV. The AV was displayed on the screen. The sample temperature was increased with the electric furnace. The true temperature (Ttrue) was read from the commercial temperature apparatus (Union 305) with type K thermocouple for the sensor and the electric voltage drop on the resistor (AV) was read on the computer screen for calibration from  $24^\circ\text{C}$  to  $100^\circ\text{C}$ . The Ttrue versus AV was plotted and displayed on this graph and showed the equation, Ttrue = f(AV) with EXCEL. The relation of Ttrue versus AV was written into the control and measure program.



*Fabrication and Physical Properties of  $\text{NiMn}_2\text{O}_4$ ,  $\text{ZnO}+0.02\text{TiO}_2$  and  $\text{FeNbO}_4$   
as Thermoelectric, Heating Element and Negative Temperature Coefficient Materials*

5) Then, this program was RUN and read the true temperature (Ttrue) and measured the temperature from the computer with the prepared sample as a sensor (Tmeasure). Both temperature were plotted for calibration. This finishes the calibration.

6) We obtain temperature apparatus that are displayed with a computer and the prepared sample as a sensor.

7) The prepared sample was tested for a temperature control device (Figure.3). The test program was written for controlling the relay drive circuit for supplying the electric voltage 5 V and 0 V for a relay switch opened and closed, alternately. The switch was opened and closed and will control solid-state relay operation for ON and OFF, alternately for heating the furnace at a given rate. The open and close of the relay switch will control the operation of the solid state relay for ON and OFF at a given rate. The computer will read temperature. When the temperature was increased to a set point, the electricity was not supplied to the electric furnace. The prepared sample can be used as a temperature measurement and control sensor with the computer as a control and display device.

***Program for computer temperature measurement and control with the prepare sample as a temperature sensor***

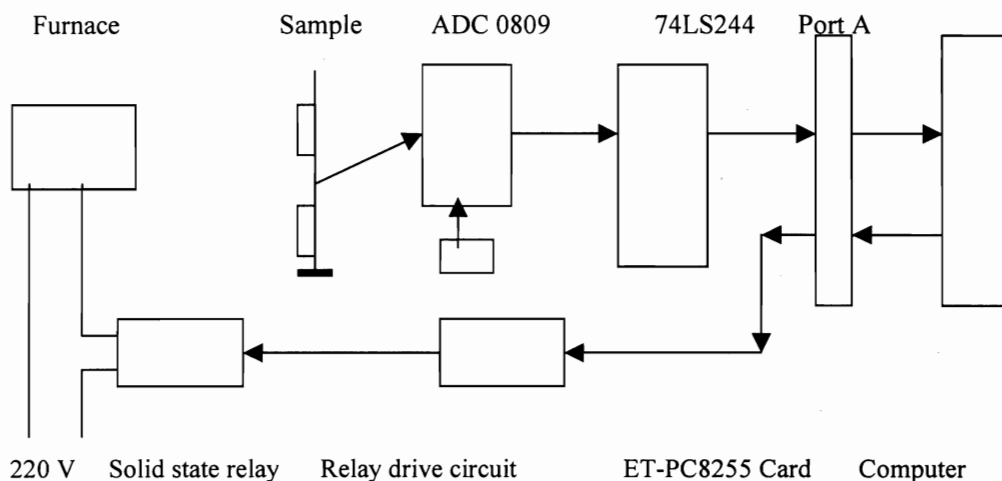
```
Program OVEN_Temperature_Controller;
uses crt;
var
  ch      : char;
  i, j, DV : integer;
  AV, VT, T, Ts : real;
const PA  = $0304;
      PB  = $0305;
      Pcontrol = $0307;
begin
  port[Pcontrol] := $90;
  Ts:=0;
  port[PB]:=255;
  delay(100);
  port[PB]:=0
  delay(100);
```

```
  clrscr;
  gotoxy(14,1); writeln('OVEN
TEMPERATURE
CONTROLLER (0-300 C) FOR
MATERIALS')
  gotoxy(14,2);writeln('-----')
  gotoxy(25,4); writeln('Setting Temperature
= ',Ts:3:0);
  gotoxy(50,4); writeln(' C ');
  gotoxy(47,4); readln(Ts);
  repeat
    gotoxy(33,12); writeln('OVEN START');
    port[PB]:=255;
    delay(2000);
    sound(900); delay(10); nosound;
    gotoxy(29,15); writeln('Reading
Temperature');
    DV := port[PA];
    gotoxy(35,17); writeln('DV = ',DV:3);
    AV := (5/255)*DV;
    VT:=AV;
    gotoxy(34,18); writeln('VT =
',VT:1:2,'V');
    T:=-4.6799*VT+75.099*VT;
    gotoxy(34,22); writeln('T = ',T:3:2);
    gotoxy(43,22); writeln(' C ');
    port[PB]:=0;
    delay(500);
    sound(9000); delay(10); nosound;
  until T > Ts;
  for i:= 1 to 1500 do
  begin
    repeat
      gotoxy(33,12); writeln('OVEN START');
      port[PB]:=0;
      sound(900); delay(10); nosound;
      gotoxy(29,15); writeln('Reading
Temperature');
      DV := port[PA];
      gotoxy(35,17); writeln('DV = ',DV:3);
      AV := (5/255)*DV;
      VT:=AV;
      gotoxy(34,18); writeln('VT =
',VT:1:2,'V');
      T:=-4.6799*VT+75.099*VT;
      gotoxy(34,22); writeln('T = ',T:3:2);
      gotoxy(43,22); writeln(' C ');
      delay(200);
```

```

until T < Ts;
repeat
  sound(5000); delay(90); nosound;
  port[PB]:=255;
  delay(1700);
until T > Ts-1;
end;
gotoxy(37,23); writeln('Relay OFF');
gotoxy(39,24); writeln('END');
delay(5000);
end.

```



**Figure 5** Block diagram for computer temperature measurement and control with the  $\text{Fe}_2\text{O}_3+\text{Nb}_2\text{O}_5$  sample as a temperature sensor.

## RESULTS AND DISCUSSION

### *Samples and XRD patterns*

The preparation of the samples was carried out using the standard ceramic technique. The composition of the samples before firing were  $\text{NiO}+\text{MnO}_2$ ,  $\text{ZnO}+0.02\text{TiO}_2$  and  $\text{Fe}_2\text{O}_3+\text{Nb}_2\text{O}_5$ . Characterization was carried out using the XRD technique. After firing, the first sample was a single phase material ( $\text{NiMn}_2\text{O}_4$ ), the second sample was the mixed phase material ( $\text{ZnO}$ ,  $\text{ZnTiO}_4$ ) and the third sample was a single phase material ( $\text{FeNbO}_4$ ). The as-fired  $\text{NiMn}_2\text{O}_4$ ,  $\text{ZnO}+0.02\text{TiO}_2$  and  $\text{FeNbO}_4$  was black, white and black color, respectively. The samples were identified as shown in Table 1.

The prepared  $\text{NiO}+\text{MnO}_2$  sample was a black color, and it had a smooth surface. The sample thickness and diameter was 4.53 mm

and 12.16 mm, respectively. The sample resistance was 18.65 k $\Omega$ .

The prepared  $\text{ZnO}+0.02\text{TiO}_2$  sample was white color and the surface was smooth. The sample thickness and diameter was 2.47 mm and 12.06 mm, respectively.

**Table 1** The composition formular and the formular from the XRD patterns of the prepared materials.

Composition	XRD patterns
$\text{NiO}+\text{MnO}_2$	$\text{NiMn}_2\text{O}_4$
$\text{ZnO}+0.02\text{TiO}_2$	$\text{ZnO}$ and $\text{ZnTiO}_4$
$\text{Fe}_2\text{O}_3+\text{Nb}_2\text{O}_5$	$\text{FeNbO}_4$

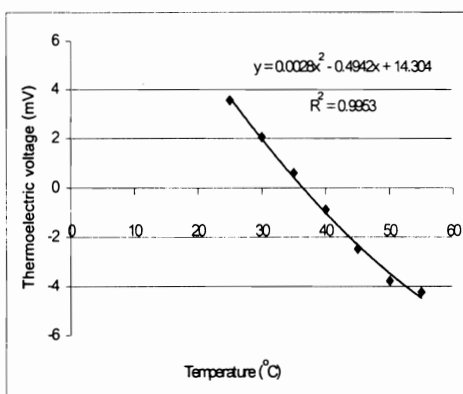
*Fabrication and Physical Properties of  $\text{NiMn}_2\text{O}_4$ ,  $\text{ZnO}+0.02\text{TiO}_2$  and  $\text{FeNbO}_4$  as Thermoelectric, Heating Element and Negative Temperature Coefficient Materials*

The prepared  $\text{FeNbO}_4$  sample was a gray-black and a smooth surface. The sample thickness and diameter was 4.88 mm and 11.73 mm, respectively. The sample resistance was 5.9 k $\Omega$ .

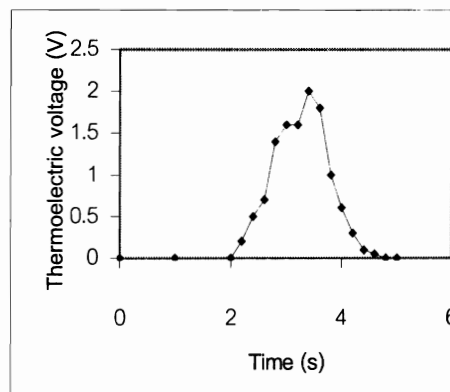
**Thermoelectric voltage versus temperature and thermoelectric voltage versus time of  $\text{NiO}+\text{MnO}_2$  material**

The thermoelectric voltage versus temperature of the sample was shown in Figure 6. The sample shows the thermoelectric effect. The thermoelectric voltage versus temperature relation corresponds to the equation,  $V = 0.0028T^2 - 0.4342T + 14.304$  with  $R^2 = 0.9953$ . The thermoelectric voltage was a negative value. So, the sample was n-type material. It can be suggested that the thermoelectric voltage generation is due to the motion of the electrical carriers from the hot to cold surface of the material. This thermoelectric effect was a direct conversion from thermal energy to electrical energy. This is a good material for electric generation without environmental pollution.

The computer interfacing circuit which has been made can exhibit the picture of the thermoelectric versus time relation as shown in Figure 7. This sample was investigated for obtaining the type of the electrical carriers in ceramics.



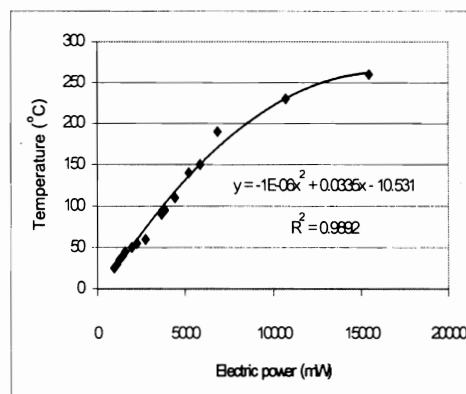
**Figure 6** The thermoelectric versus temperature for the  $\text{NiO}+\text{MnO}_2$ .



**Figure 7** Picture on the computer screen about sample the thermoelectric voltage versus time for the  $\text{NiO}+\text{MnO}_2$  sample.

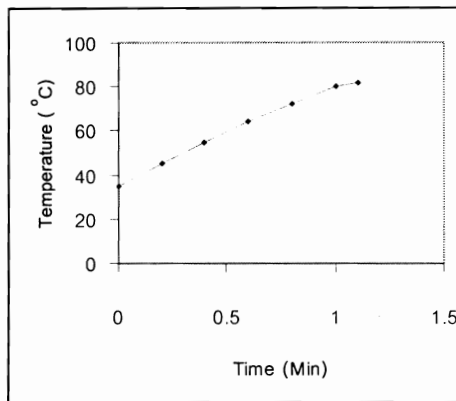
**Temperature versus supplied electric power and temperature versus time of the  $\text{ZnO}+0.02\text{TiO}_2$  material**

Temperature versus supplied electric power of the material was measured as shown in Figure 8. The supplied electric power was increased, and the sample temperature was increased. The sample shows heating effect as the increasing of supplied electric power. The temperature versus supplied electric power relation corresponds to the equation,  $T = -1 \times 10^{-6}P^2 + 0.0335P - 10.531$ , with  $R^2 = 0.9892$ . This sample can show the electricity to heat conversion



**Figure 8** Temperature versus supplied electric power for the  $\text{ZnO}+0.02\text{TiO}_2$  material.

the material from the computer screen was shown in Figure 9. The plot of temperature versus time can be shown with computer interfacing circuit system. This material was studied for the purpose of a heating element for the electric furnace.



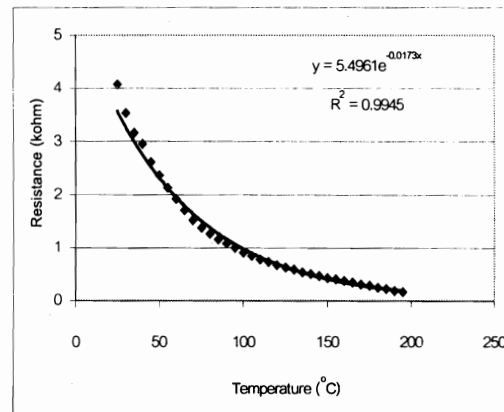
**Figure 9** Temperature versus time for the ZnO+0.02TiO<sub>2</sub> material from the computer screen.

***The resistance versus temperature, Temperature measurement and control of the Fe<sub>2</sub>O<sub>3</sub>+Nb<sub>2</sub>O<sub>5</sub> material.***

***Resistance versus temperature.***

The measured resistance versus temperature of the sample was shown in Figure 10. The temperature was increased, the resistance was decreased. The sample was observed to be sensitive to the heat. So, the sample shows the NTC effect. The sample resistance versus the temperature relation corresponds to the equation,  $R = 5.4961e^{-0.0173T}$ , with  $R^2 = 0.9945$ . The negative temperature coefficient of the resistance ( $\alpha$ ) was  $-1.03\%/^{\circ}\text{C}$  from 25 to  $100^{\circ}\text{C}$  and  $-0.84\%/^{\circ}\text{C}$  from 100 to  $200^{\circ}\text{C}$ . But from the text,  $\alpha = -1.0$  to  $-6.0\%/^{\circ}\text{C}$  (Buchanan, 1991). This NTC effect is believed to be associated with the energy band structure (Soliman, 1993). When the sample temperature is increased, the electrons are moved from valence band to conduction band and a hole is created in the valence band. The electron-hole pairs become

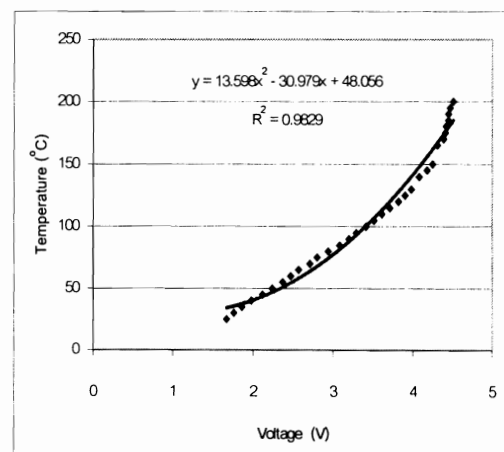
the moving electric current. The sample resistance will decrease. The sample was an n-type semiconducting ceramics. The sample can be used as a temperature measurement and control sensor because of the thermal response properties.



**Figure 10** The measured resistance versus temperature for the Fe<sub>2</sub>O<sub>3</sub>+Nb<sub>2</sub>O<sub>5</sub> sample.

***Temperature measurement and control***

The temperature (T) versus voltage drop (V, AV) from the calibration was shown in Figure 11. After the calibration by the measurement of the  $T=f(AV)$ , then this equation was put in the program and RUN the program was RUN.

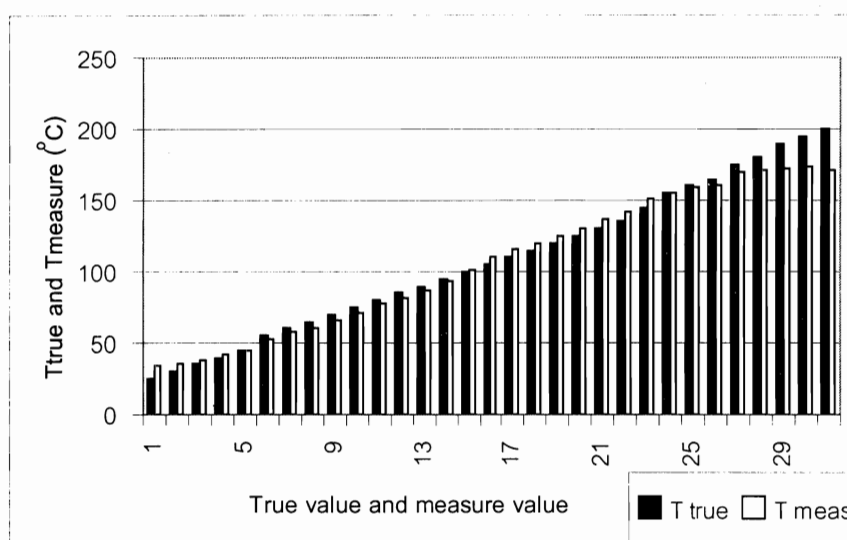


**Figure 11** The temperature versus drop voltage for the Fe<sub>2</sub>O<sub>3</sub>+Nb<sub>2</sub>O<sub>5</sub> material.

*Fabrication and Physical Properties of  $\text{NiMn}_2\text{O}_4$ ,  $\text{ZnO}+0.02\text{TiO}_2$  and  $\text{FeNbO}_4$  as Thermoelectric, Heating Element and Negative Temperature Coefficient Materials*

The comparison between the true temperature ( $T_{\text{true}}$ ) from commercial apparatus and the measured temperature ( $T_{\text{measure}}$ ) from the computer with the prepared sample for determining the accuracy) from 24 to 200°C is shown in Figure 12. So, the prepared sample can be used as a temperature sensor with a computer display. Finally, the sample can be used as a temperature sensor. A testing system for a temperature control measurement and control is composed of a prepared sample as a temperature sensor and the computer as a

reading device. After the program was run, the computer will control the electrical power supplying to the electric furnace with a 6 V relay and a solid-state relay for supplying and stop supplying the electric furnace, alternately. The furnace was heated. When the furnace temperature was increased to a setting temperature, the computer will control the temperature at a constant value at a given interval. The example of temperature control operation was done by setting the temperature to 180°C, reading controlling temperature with the computer was no exceeded.



**Figure 12** The comparison between  $T_{\text{true}}$  and  $T_{\text{measure}}$  for the  $\text{Fe}_2\text{O}_5+\text{Nb}_2\text{O}_5$  material.

## CONCLUSIONS

The thermoelectric effect, heating effect and NTC effect which are the electrical behavior of the ceramics, were investigated. It was demonstrated that the three effects can be used as carrier type inspection, heating element and NTC device, respectively. For the improvement of the instrumentation, the computer interfacing techniques were designed and constructed in this research laboratory room. It should be noted that the computer interfacing system and program could be used to design some experiments in the future. In addition, the studying of the physical

properties of the ceramics, such as the thermoelectric, heating and NTC effect has been developed and used for further research.

## ACKNOWLEDGEMENTS

The author gratefully acknowledge from Prince of Songkla University (PSU) and The National Research Council of Thailand (nrct), Thailand for financial supporting this research work.

**REFERENCES**

- Buchanan, R. C. 1991. *Ceramic Materials for Electronics*. 2<sup>nd</sup> ed. New York, Mercel Dekker Inc.
- Das Gupta, S. 1996. Heating elements and electrically conducting ceramics. *Eng. Mater.* **122** : 279-282.
- Moulson, A. J. and Herbert, J. M. 1990. *Electroceramics*. London, Champman & Hall.
- Nukuaw, Jiti. 1991. *IBM PC Computer interfacing circuit with external device for application*. Science, Engineering & Education CO. : 1-159.
- Ravinder, D. 1994. Electrical conductivity of Mn-Zn ferrites. *J. Appl. Phys.* **75 (10)** : 6118-6120.
- Soliman, F. A. S. 1993. Radiation effects on non-linear resistances. *J. Mater. Sci. : Mater. Elect.* **4** : 293-300.