CHAPTER IV

THE EXPERIMENTAL RESULTS AND DISCUSSION

In this chapter, the experimental results are analysed. The thermal conductivity property of epoxy adhesive was developed by the addition of thermal conductive filler as described in chapter III.

Types of thermal conductive additive influence on thermal conductivity of each epoxy adhesive are presented in section 4.1. The relations between the filler percentage and the thermal conductivity characteristic of composites are summarized in section 4.2. Section 4.3 demonstrates the effect of temperature variation. The Lewis & Nielsen semi-theoretical model and the Ratcliffe model are presented to explain to experimental results in section 4.4 and finally, section 4.5 reports the adhesion property of adhesive composites by shear strength test.

4.1 The effect of type of filler

To study the effect of the kind of fillers on the thermal conductivity of filled epoxy adhesive composites, the samples were prepared from two types of epoxy adhesives, epoxy adhesive A and epoxy adhesive B, and five types of fillers, aluminium metal powder, berylium metal powder, copper metal powder, berylium oxide and silicon carbide.

All fillers mentioned above have much higher thermal conductivity than plain epoxy adhesives as shown in Table 4.1

Table 4.1 Thermal conductivity of epoxy adhesives and fillers

SUBSTANCE	THERMAL CONDUCTIVITY (W/mK)
Epoxy adhesive A	0.13
Epoxy adhesive B	0.20
Aluminium metal powder	237*
Berylium metal powder	200*
Copper metal powder	401*
Berylium oxide	272*
Silicon carbide	490*

* the thermal conductivity values from [26]

The thermal conductivity analyser can give the thermal conductive value of epoxy adhesive specimens in W/mK unit. The measurements show different increased values of the thermal conductivity of each prepared epoxy sample.

The different values of the thermal conductivity of composites read from the analyser depend on different types of fillers as shown in Table 4.2 to 4.3 and Figure 4.1 to 4.2.

Figure 4.1 and 4.2 indicate the thermal conductivity values of both epoxy adhesive composites filled with different types of fillers. The range of thermal conductive characteristic of both epoxy composites is 0.12 to 0.64.

Table 4.2 Thermal conductivity of epoxy adhesive composites A

% Weight of fillers	Thermal conductivity (W/mK)				
	aluminium metal filled	berylium oxide filled	berylium metal filled	copper metal filled	silicon carbide filled
0	0.120	0.120	0.120	0.120	0.120
2	0.130	0.130	0.130	0.124	0.125
4	0.140	0.140	0.150	0.128	0.130
6	0.150	0.150	0.160	0.133	0.138
8	0.160	0.160	0.180	0.138	0.147
10	0.180	0.170	0.200	0.144	0.158
12	0.200	0.180	0.220	0.149	0.170
14	0.220	0.200	0.250	0.154	0.180
16	0.250	0.220	0.290	0.158	0.200
20	0.310	0.270	0.380	0.174	0.240

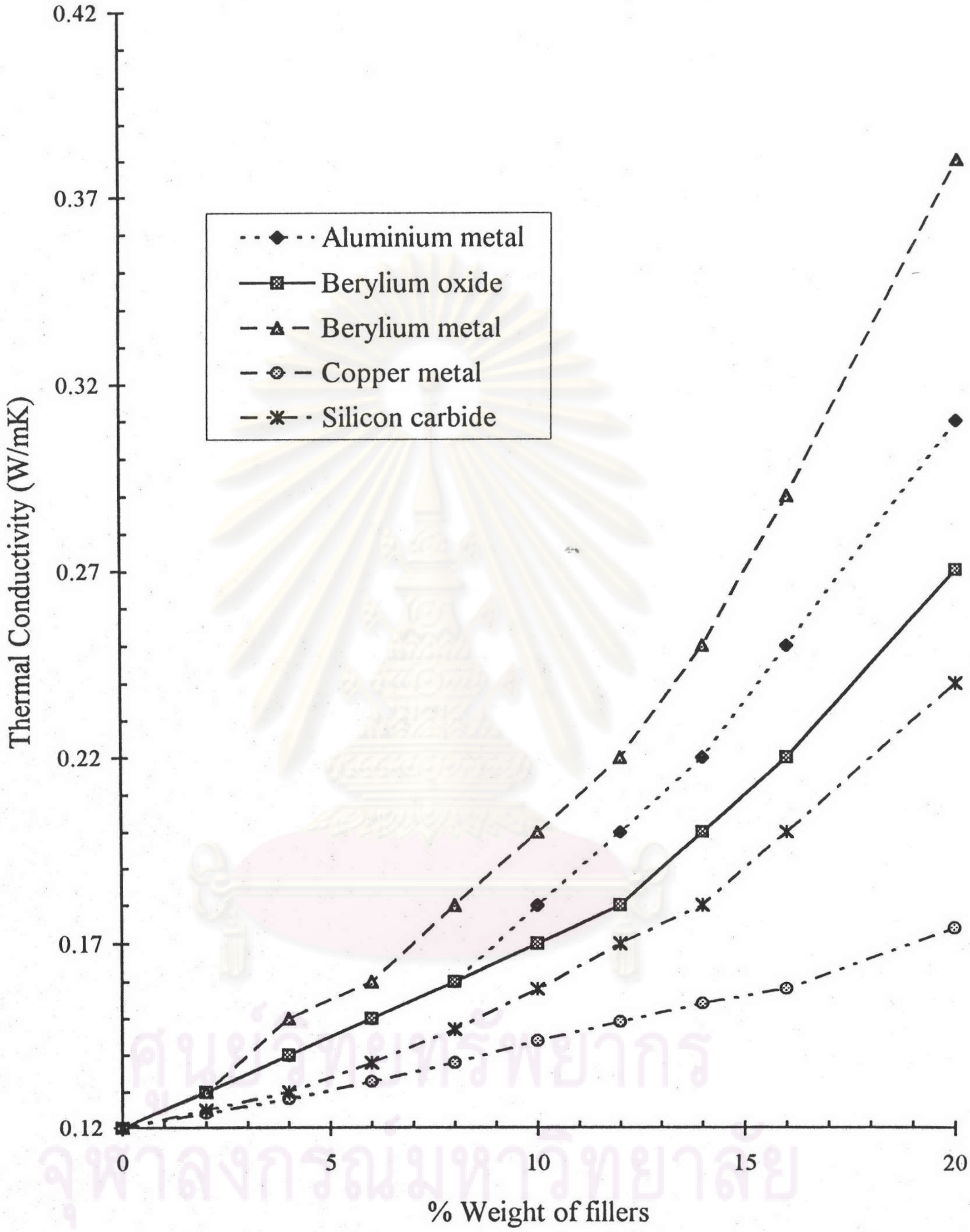

Figure 4.1 Thermal conductivity of epoxy adhesive composites A

Table 4.3 Thermal conductivity of epoxy adhesive composites B

% Weight of fillers	Thermal conductivity (W/mK)				
	aluminium metal filled	berylium oxide filled	berylium metal filled	copper metal filled	silicon carbide filled
0	0.200	0.200	0.200	0.198	0.200
2	0.210	0.210	0.220	0.204	0.210
4	0.230	0.220	0.250	0.211	0.220
6	0.250	0.240	0.270	0.217	0.230
8	0.270	0.260	0.300	0.223	0.250
10	0.300	0.280	0.330	0.229	0.270
12	0.330	0.300	0.380	0.237	0.290
14	0.370	0.330	0.430	0.246	0.320
16	0.410	0.360	0.490	0.258	0.350
20	0.500	0.430	0.640	0.282	0.420

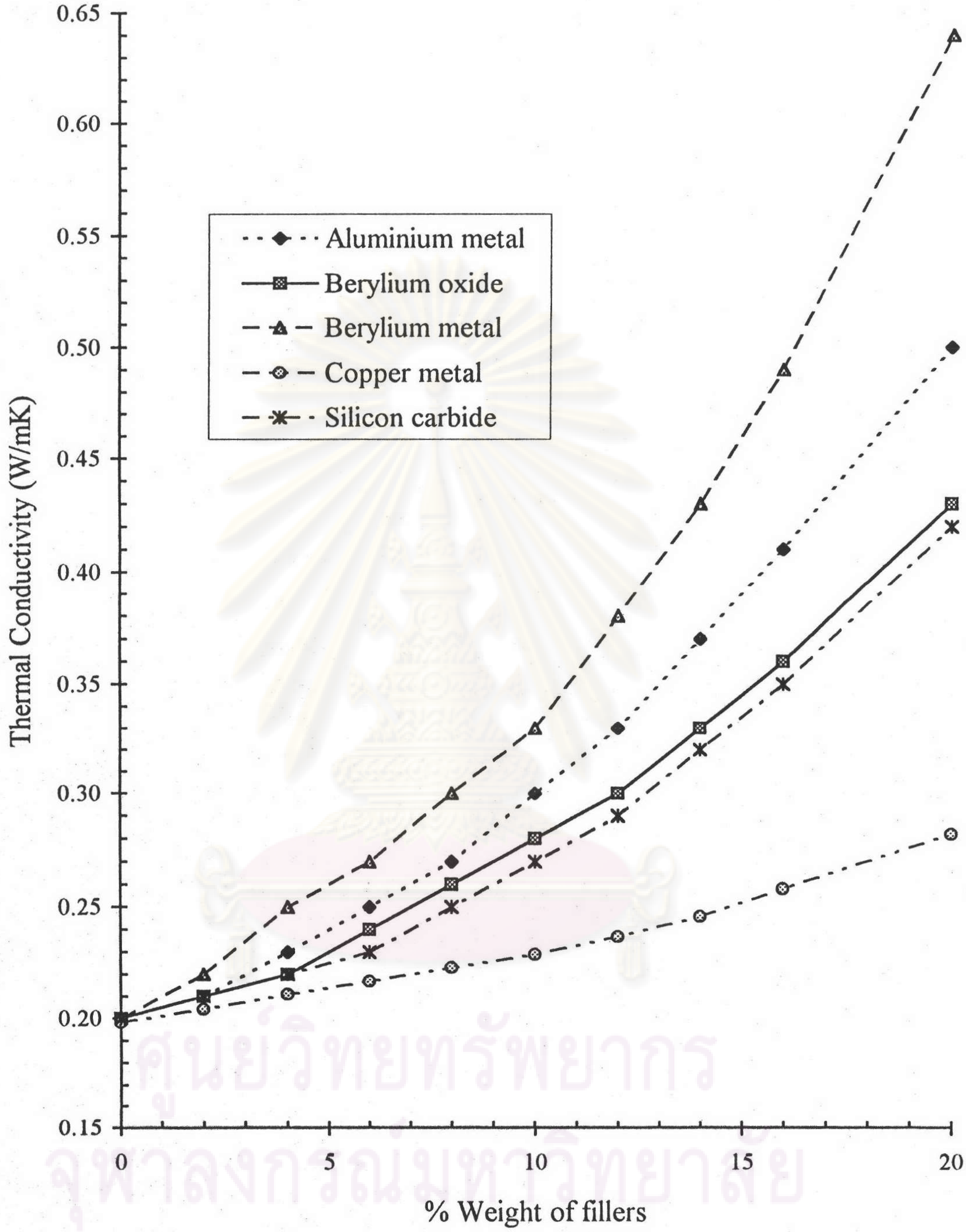

Figure 4.2 Thermal conductivity of epoxy adhesive composites B

4.2 The effect of percentage of fillers

This section reports the impact of the variation of filler percentage on the thermal conductivity value of epoxy composites. The results are illustrated in Figure 4.3 to 4.7.

The results show that the more filler percentage in epoxy matrix we added, the higher thermal conductivity value we measured. The increasing trend of thermal conductivity of different epoxy composites (both epoxy adhesive A and epoxy adheisve B) filled with the same filler showed the same increasing characteristic.

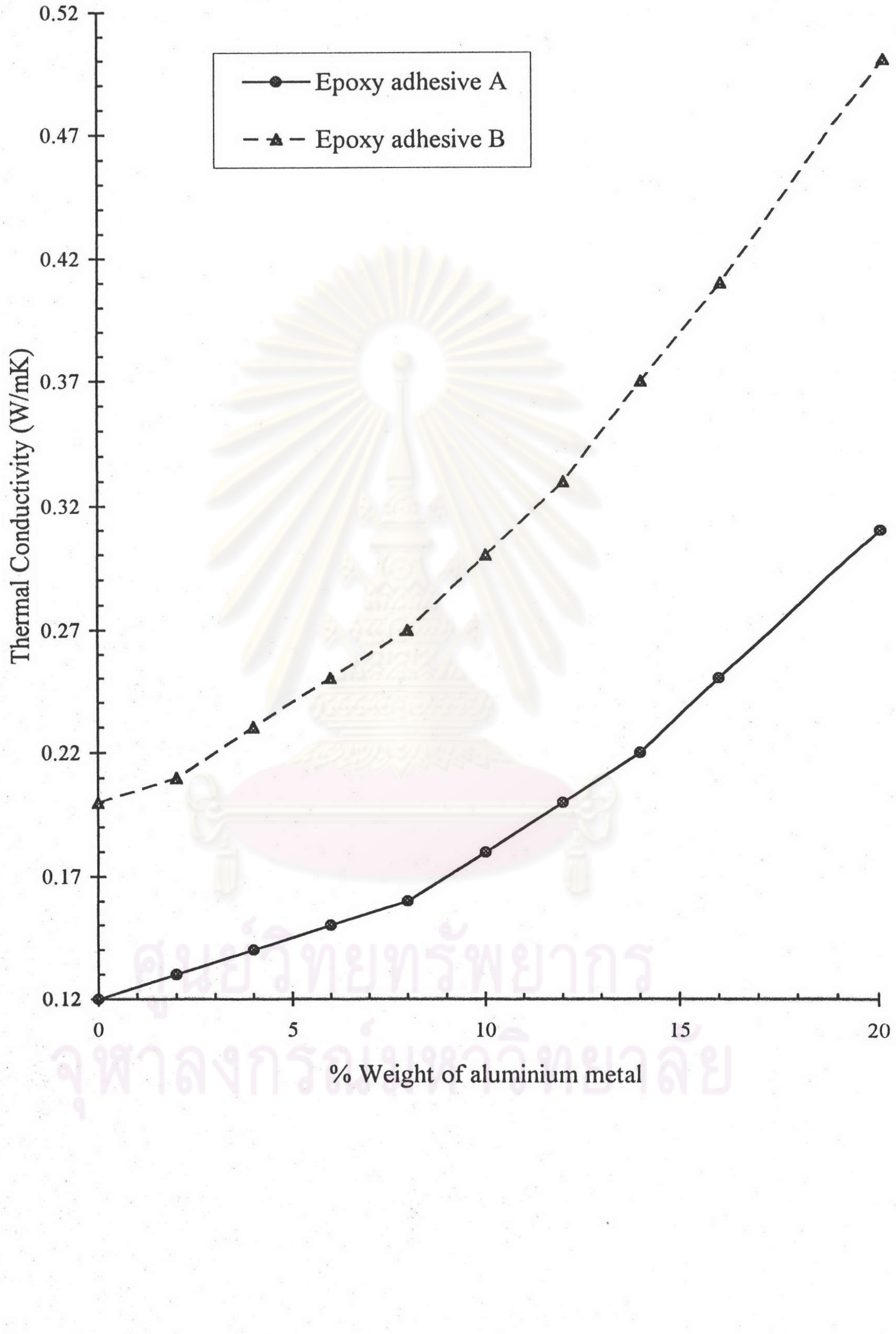

Figure 4.3 Thermal conductivity of aluminium metal filled epoxy adhesives

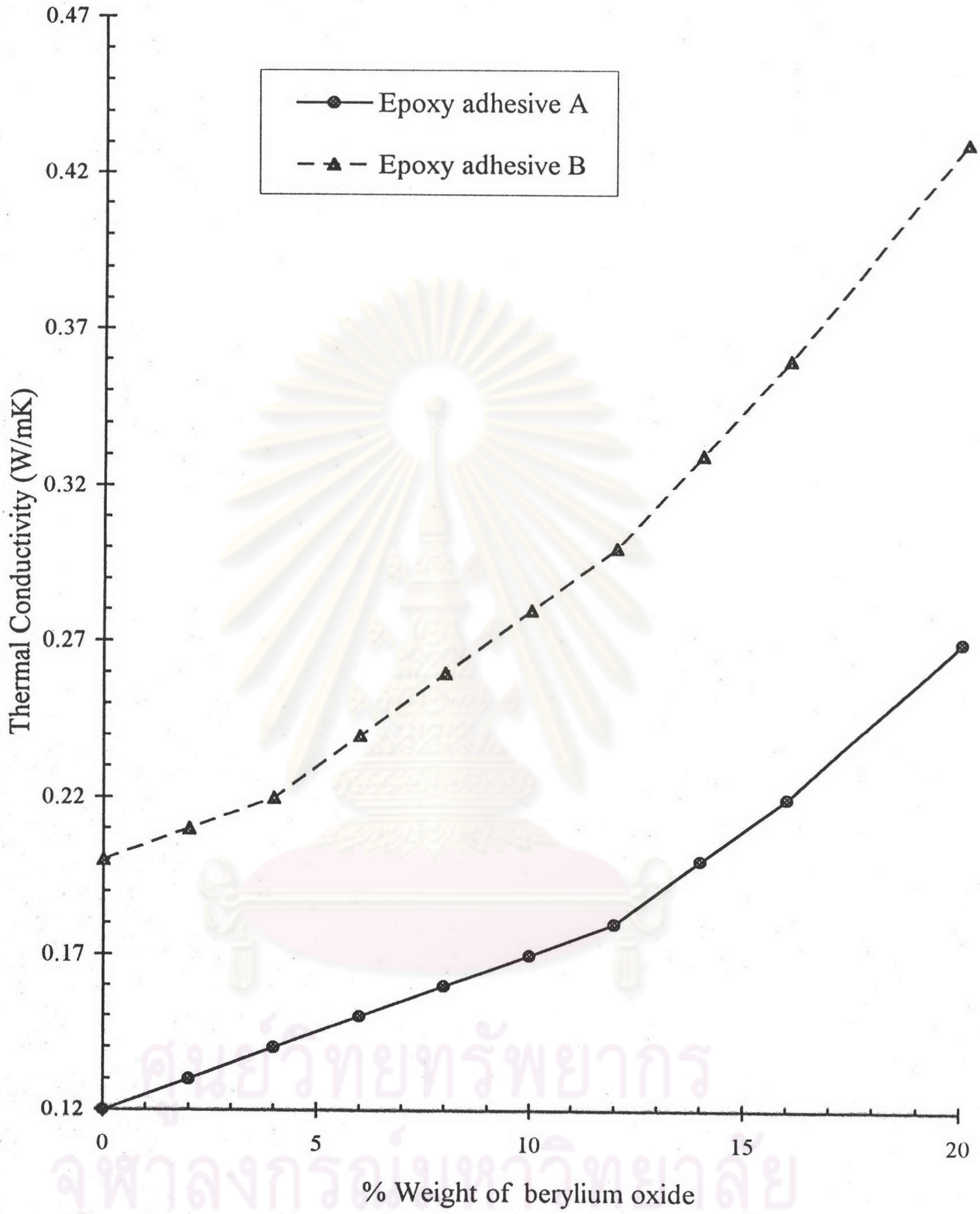

Figure 4.4 Thermal conductivity of berylium oxide filled epoxy adhesives

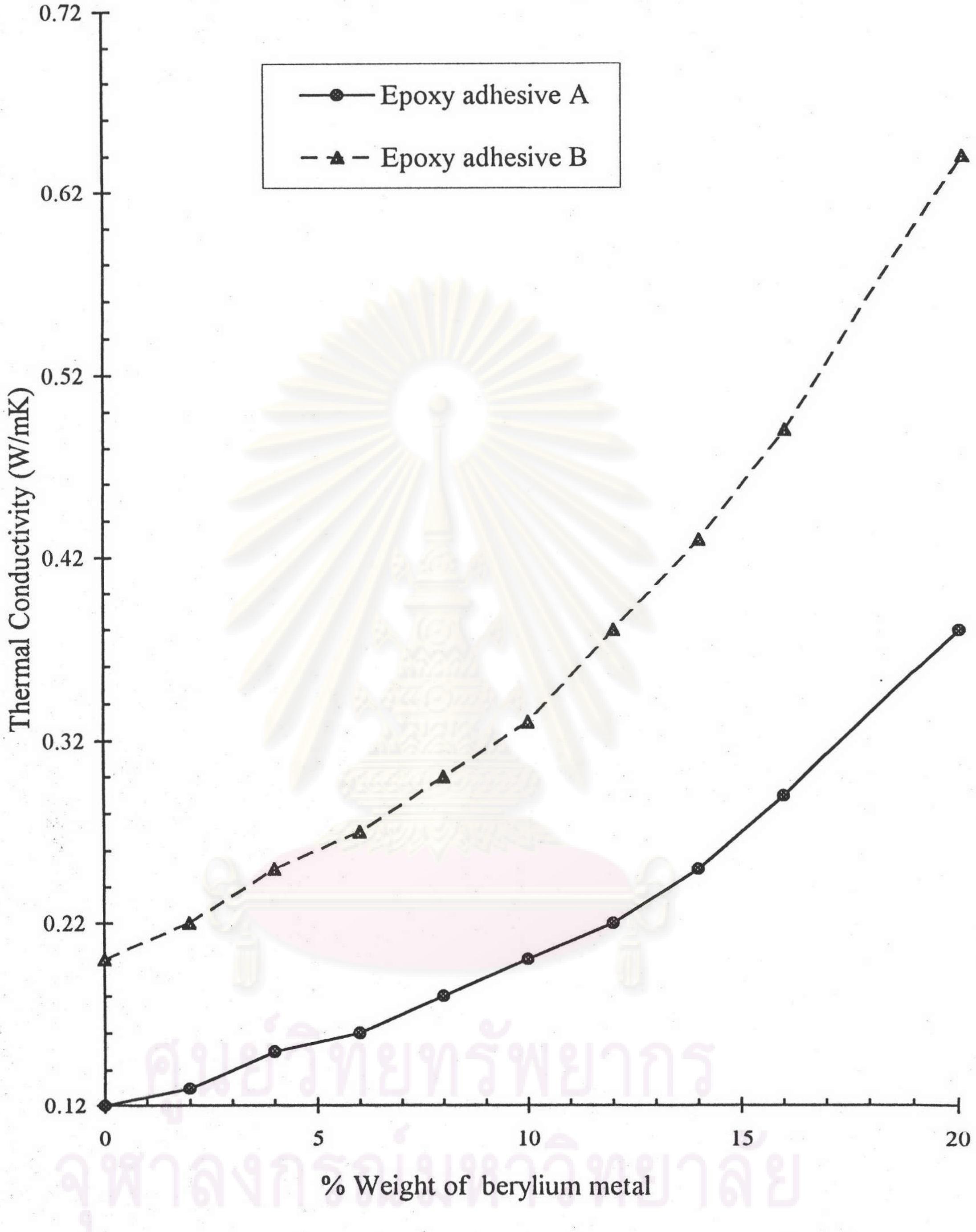

Figure 4.5 Thermal conductivity of berylium metal filled epoxy adhesives

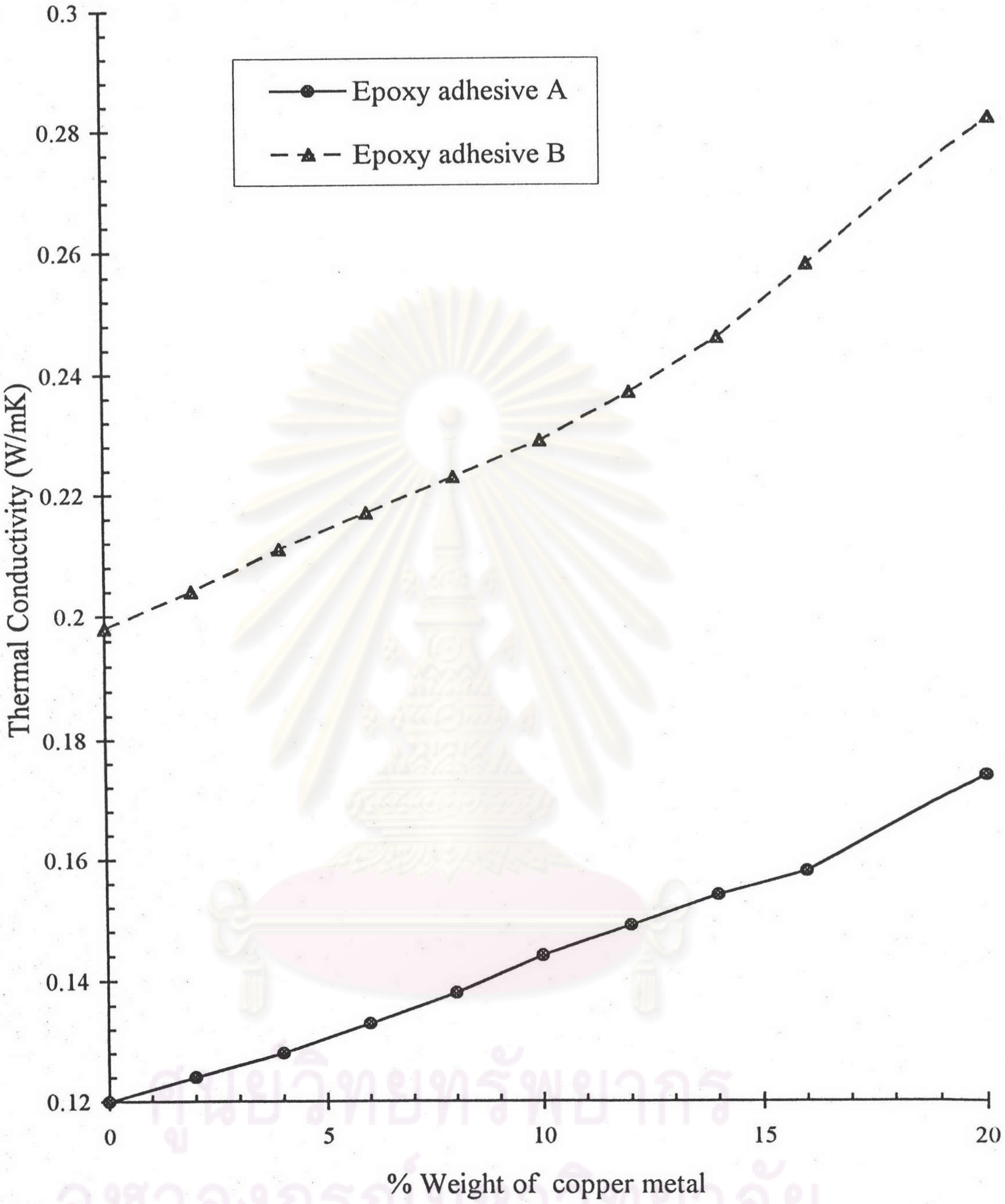

Figure 4.6 Thermal conductivity of copper metal filled epoxy adhesives

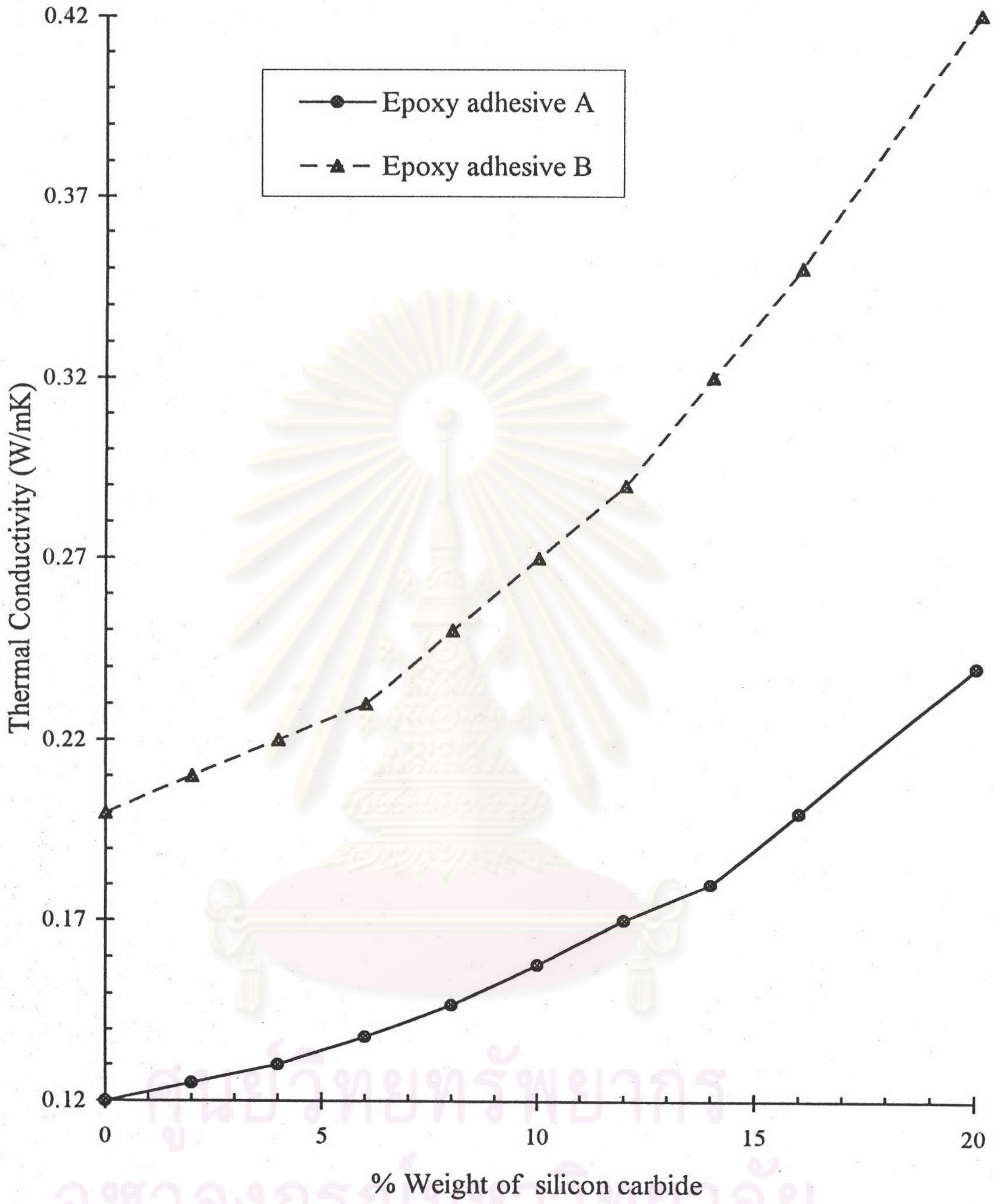

Figure 4.7 Thermal conductivity of silicon carbide filled epoxy adhesives

In order to determine a general equation to elaborate thermal conductivity characteristic of each epoxy composite prepared from the different kinds of fillers and different types of epoxy adhesives, use the data of Figure 4.3 to 4.7 to plot the relationship between the % weight of filler and the thermal conductivity of composites in semi-log scale as shown in Figure 4.8 to 4.12. The relationship can be performed in the general term of linear equation as :

$$\log k = A\,x + \log k_o$$

where k = thermal conductivity of epoxy adhesive composite

k_o = thermal conductivity of plain epoxy adhesive

x = % weight of filler that vary from 0 to 20 %

The different types of epoxy adhesive composites filled with the same filler perform the same slope of the increasing trend of thermal conductivity meaning that A value depends on type of filler. The values of A and $\log k_o$ of the studied composties are shown in Table 4.4. The interception $\log k_o$ of equation depends on the thermal conductivity of plain epoxy adhesive.

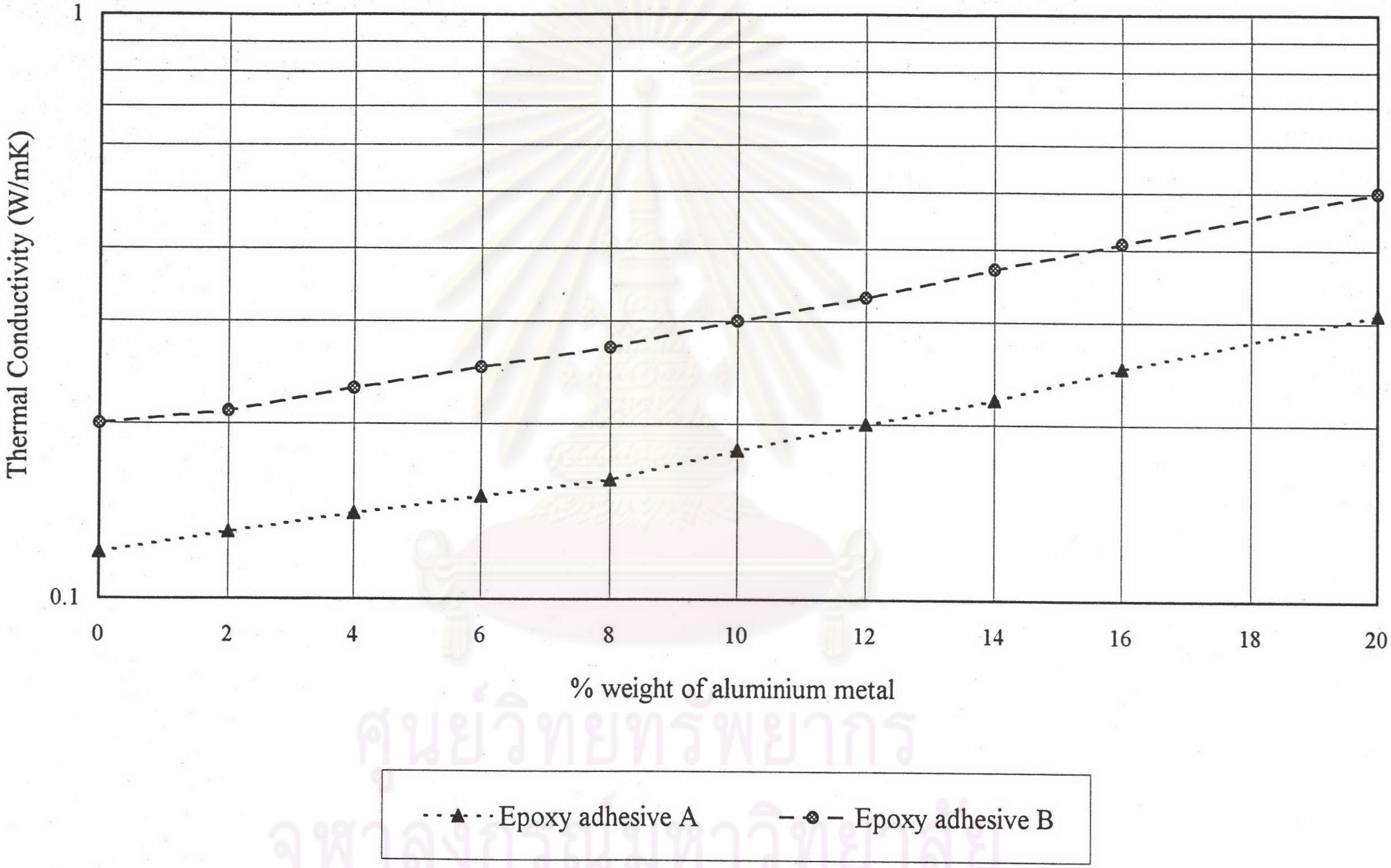

Figure 4.8 Relationship between thermal conductivity of composite and % weight of aluminium metal filler

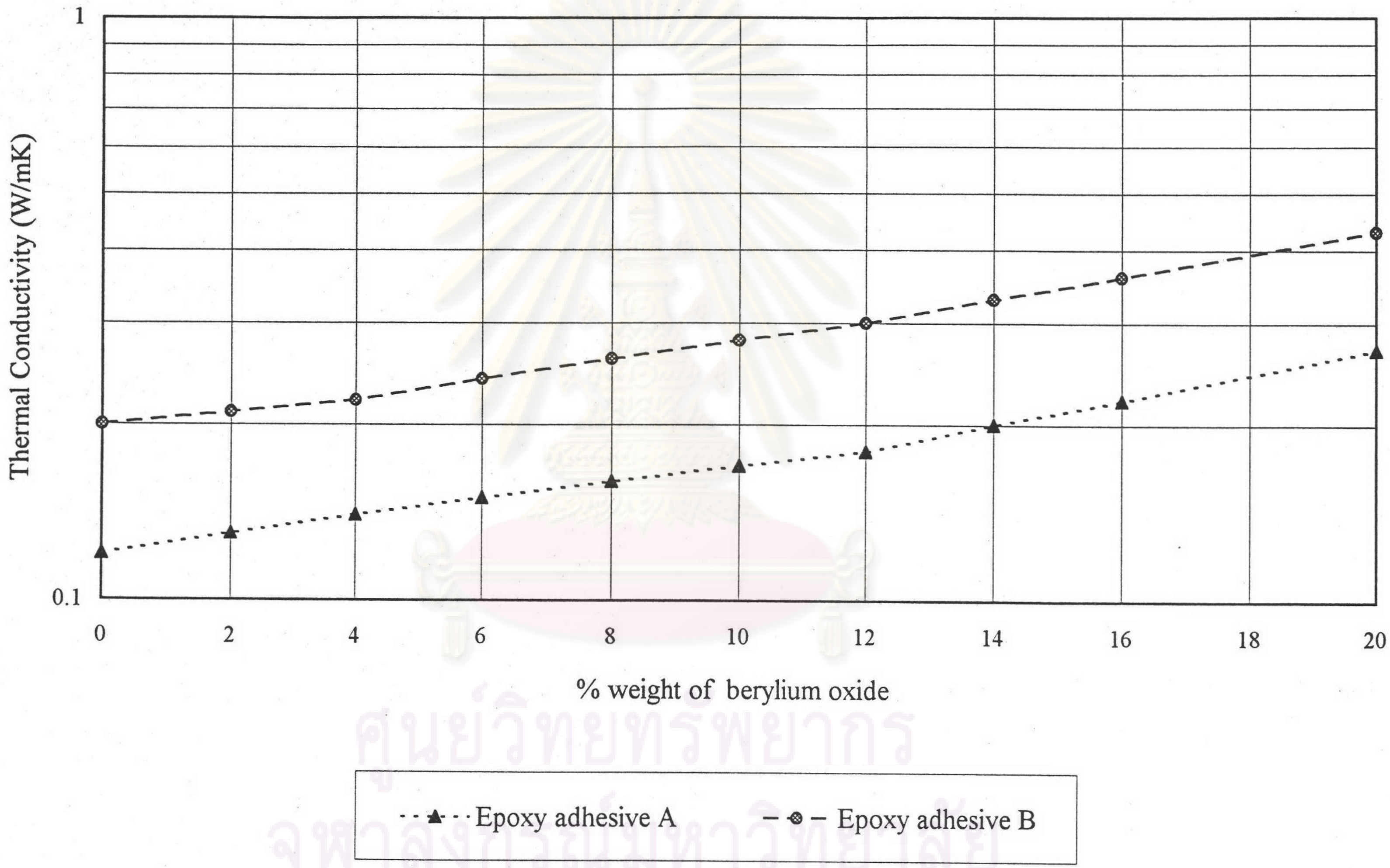

Figure 4.9 Relationship between thermal conductivity of composite and % weight of berylium oxide filler

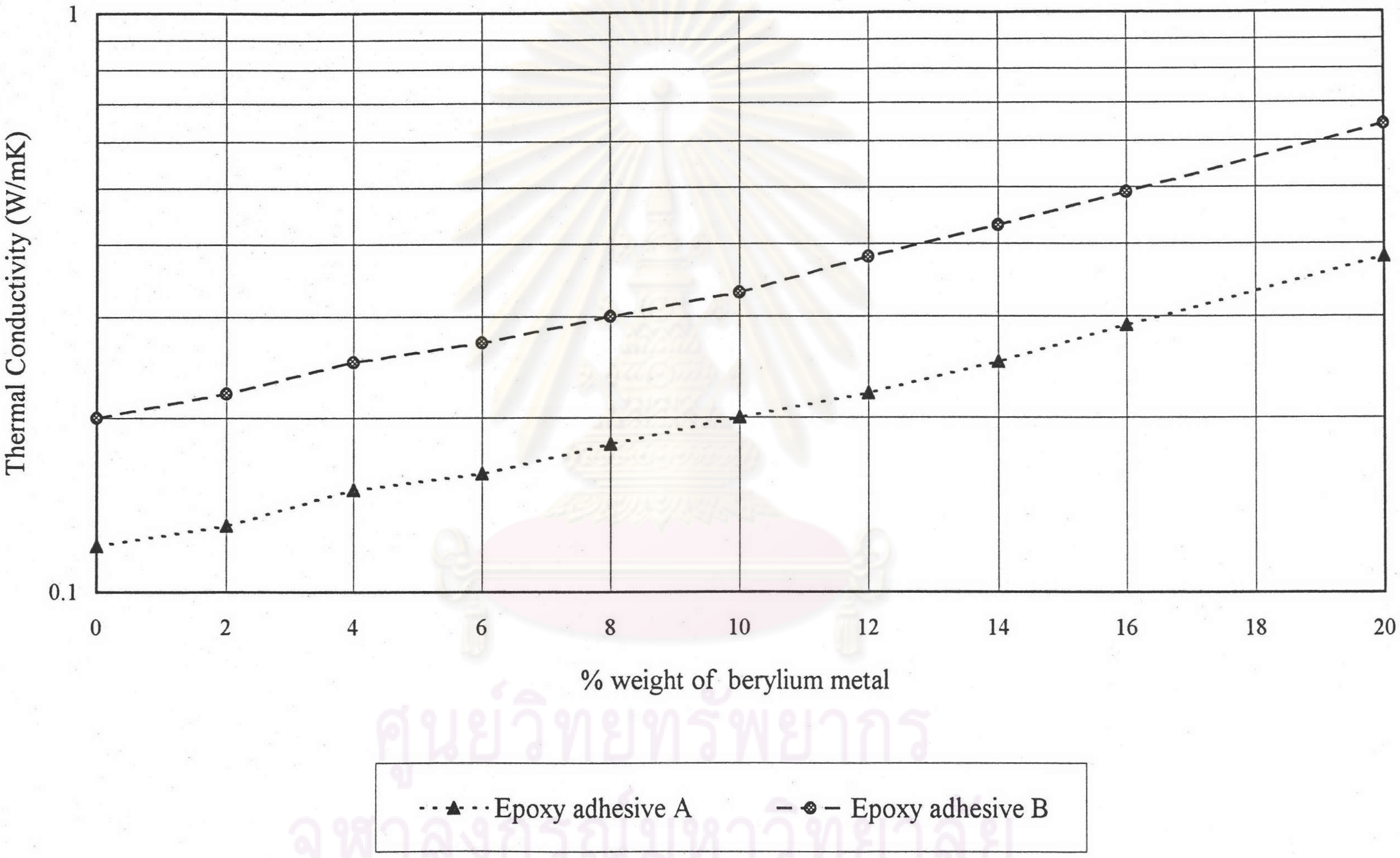

Figure 4.10 Relationship between thermal conductivity of composite and % weight of berylium metal filler

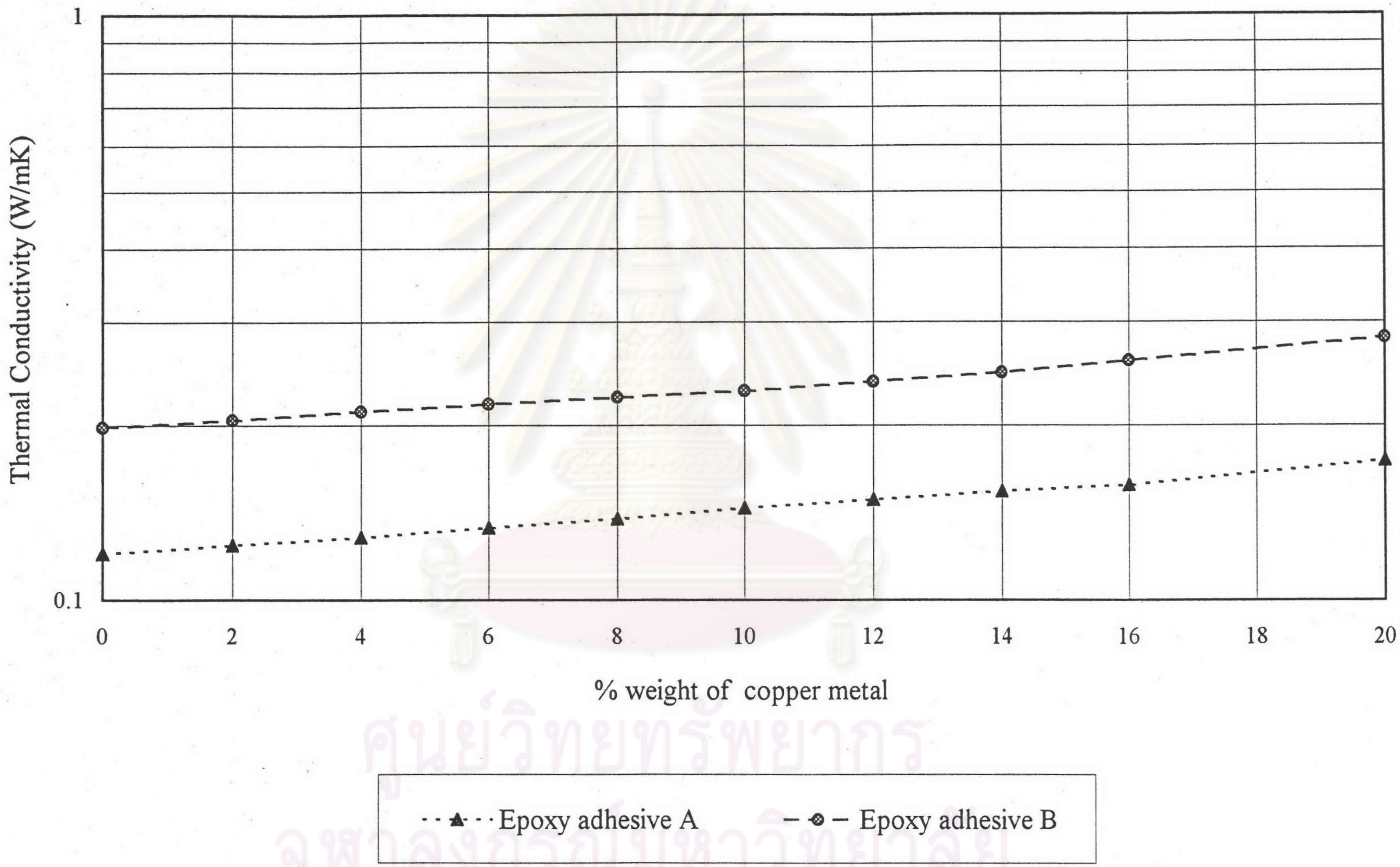

Figure 4.11 Relationship between thermal conductivity of composite and % weight of copper metal filler

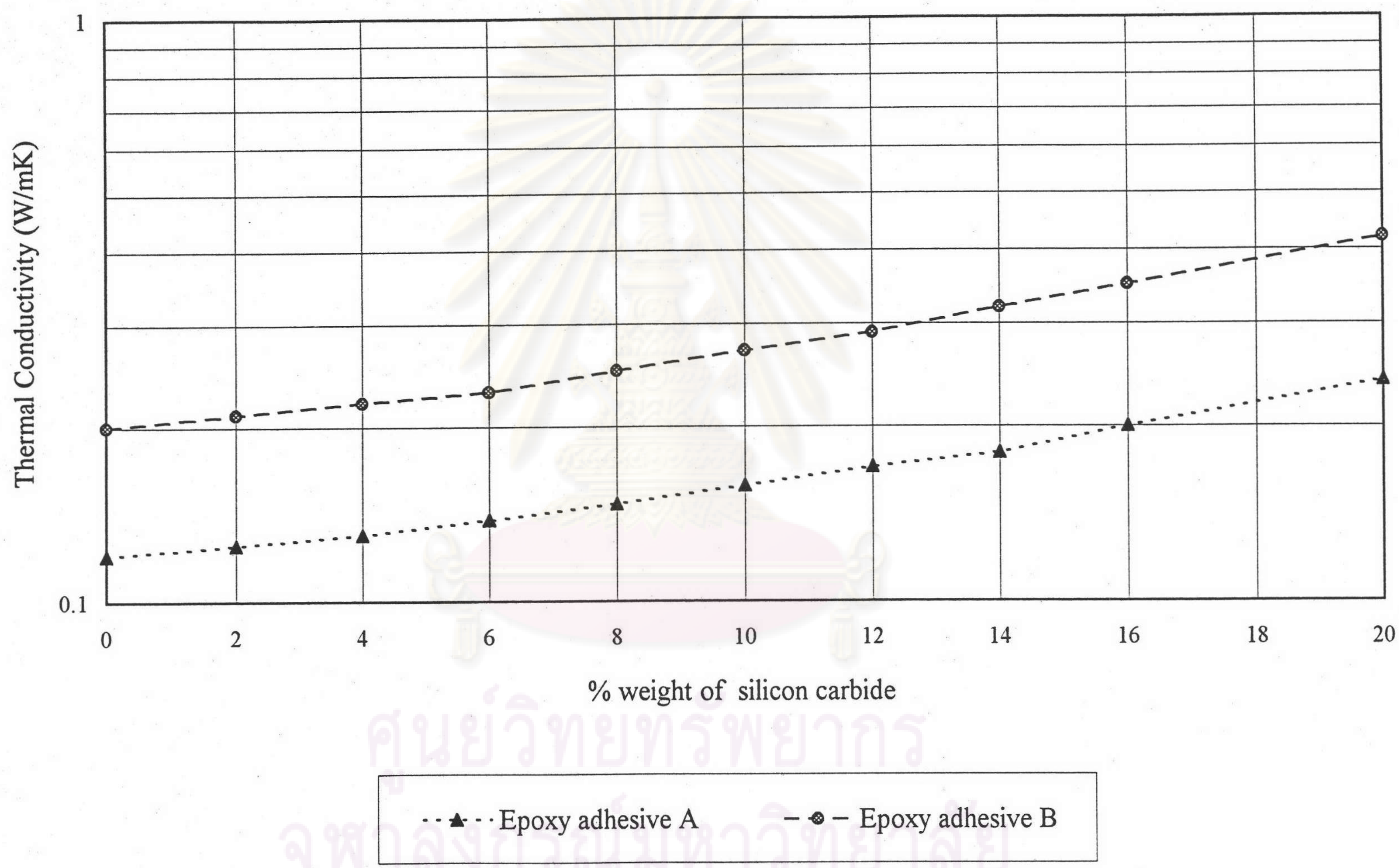

Figure 4.12 Relationship between thermal conductivity of composite and % weight of silicon carbide filler

Table 4.4 The values of A and log k_o for ten epoxy composites that prepared in this study

Epoxy Adhesives	Fillers	A	log k_o
Epoxy adhesive A	Aluminium metal	0.02	-0.92
	Berylium oxide	0.017	-0.92
	Berylium metal	0.025	-0.92
	Copper metal	0.008	-0.92
	Silicon carbide	0.0155	-0.92
Epoxy adhesive B	Aluminium metal	0.02	-0.7
	Berylium oxide	0.02	-0.7
	Berylium metal	0.025	-0.7
	Copper metal	0.008	-0.7
	Silicon carbide	0.0155	-0.7

4.3 The effect of temperature variation

The influence of temperature change on thermal conductivtiy of epoxy composite is clearly shown in Table 4.5 to 4.6 and Figure 4.13 to 4.14. This study varies temperature from 25°C to 150°C. The thermal conductivity of all epoxy adhesives have nearly constant value for the whole studied range.

Table 4.5 Thermal conductivity of epoxy adhesive composites A at various temperature (20 % weight of fillers)

Temperature (° C)	Thermal conductivity (W/mK)				
	aluminium metal filled	berylium oxide filled	berylium metal filled	copper metal filled	silicon carbide filled
25	0.300	0.270	0.380	0.176	0.230
50	0.310	0.270	0.380	0.174	0.240
100	0.310	0.260	0.380	0.175	0.230
150	0.310	0.270	0.370	0.174	0.240

Table 4.6 Thermal conductivity of epoxy adhesive composites B at various temperature (20 % weight of fillers)

Temperature (°C)	Thermal conductivity (W/mK)				
	aluminium metal filled	berylium oxide filled	berylium metal filled	copper metal filled	silicon carbide filled
25	0.510	0.440	0.640	0.280	0.420
50	0.500	0.430	0.640	0.280	0.420
100	0.500	0.430	0.630	0.282	0.410
150	0.510	0.430	0.630	0.281	0.400

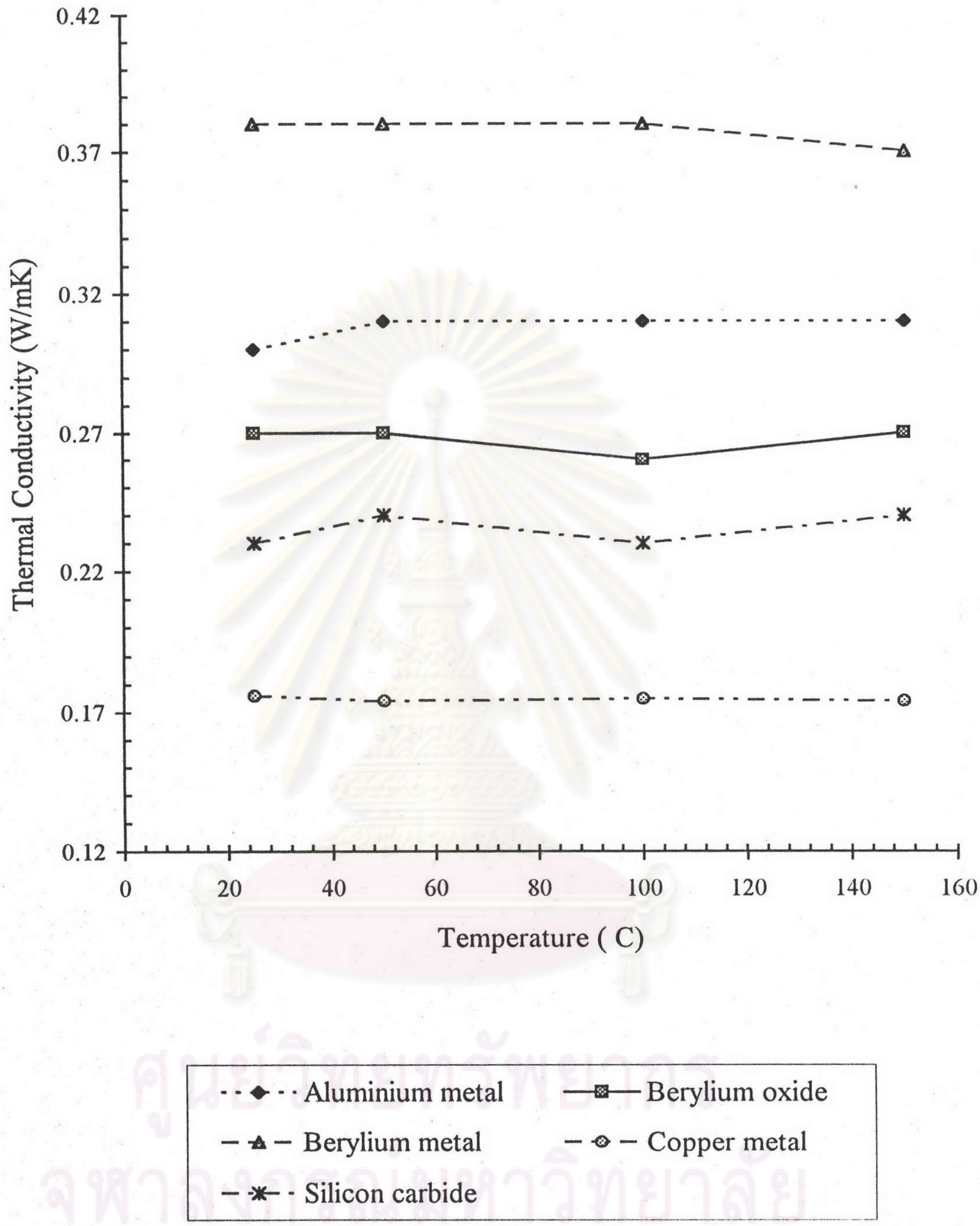

Figure 4.13 Thermal conductivity of 20% of fillers filled epoxy adhesive composites A at various temperature

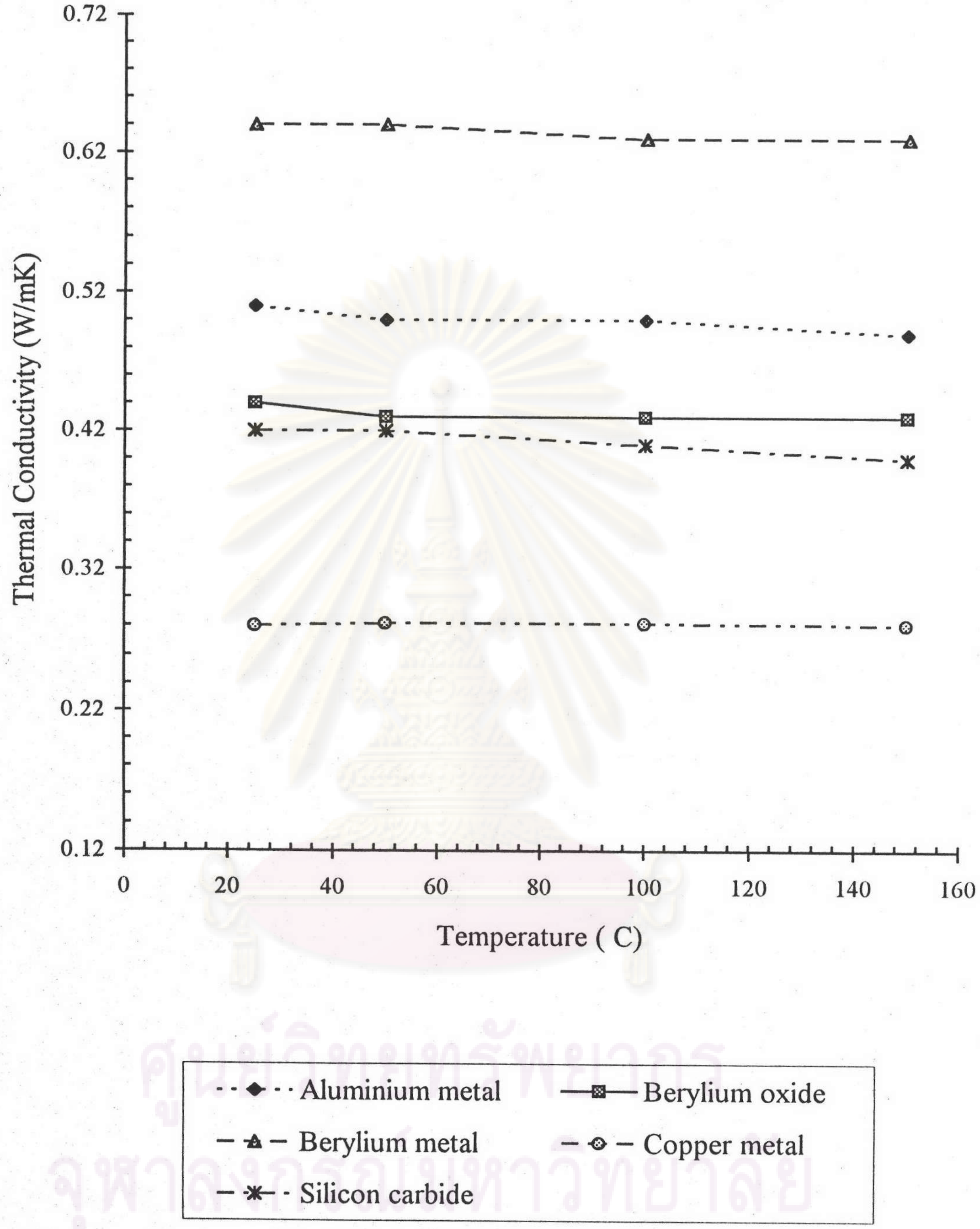

Figure 4.14 Thermal conductivity of 20% of fillers filled epoxy adhesive composites B at various temperature

4.4 The prediction of thermal conductivity

There are a lot of procedures to determine the thermal conductivity of epoxy composites by experimentation such as transient hot wire, transient hot strip and steady-state guarded hot plate method. There are also a lot of theoretical or semi-theoretical models which can be used to explain the thermal conductive characteristic of different types of composites. Some models are proper for solution, some appropriate for filaments, porus solid, suspension and foam.

In this thesis, the thermal conductivity was measured from the thermal conductivity analyser by steady-state guarded hot plate method and also use the Lewis & Nielsen semi-theoretical model and the Ratcliffe model to plot comparison curve with experimental results as shown in Table 4.7 to 4.16 and Figure 4.15 to 4.24.

Table 4.7 Thermal conductivity comparison between the models and the experimental data for the composite of epoxy adhesive A filled with aluminium metal

% Weight of fillers	% Volume fraction of fillers	Thermal conductivity (W/mK)			
		experiment 1	experiment 2	Lewis Model	Ratcliffe Model
0	0.0000	0.120	0.120	0.120	0.120
2	0.0086	0.130	0.130	0.123	0.128
4	0.0174	0.140	0.140	0.125	0.137
6	0.0264	0.150	0.150	0.128	0.147
8	0.0357	0.160	0.160	0.131	0.157
10	0.0452	0.180	0.170	0.134	0.169
12	0.0549	0.200	0.200	0.138	0.182
14	0.0648	0.220	0.210	0.141	0.196
16	0.0751	0.250	0.250	0.145	0.212
20	0.0962	0.310	0.300	0.153	0.249

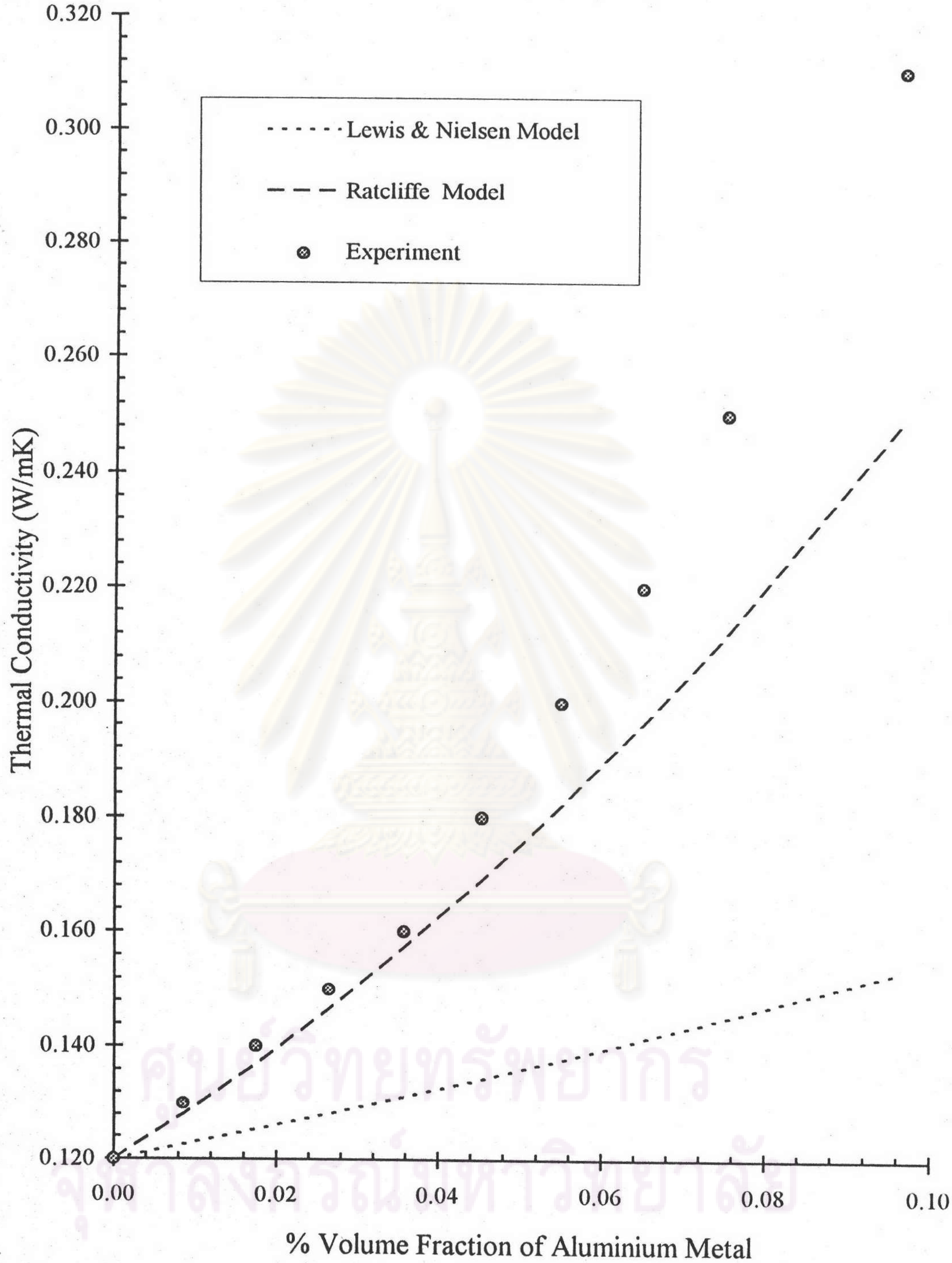

Figure 4.15 Comparison between the models and the experimental results for the composite of epoxy adhesive A filled with aluminium metal powder

Table 4.8 Thermal conductivity comparison between the models and the experimental data for the composite of epoxy adhesive B filled with aluminium metal

% Weight of fillers	% Volume fraction of fillers	Thermal conductivity (W/mK)			
		experiment 1	experiment 2	Lewis Model	Ratcliffe Model
0	0.0000	0.200	0.200	0.200	0.200
2	0.0094	0.210	0.210	0.205	0.214
4	0.0189	0.230	0.220	0.210	0.229
6	0.0287	0.250	0.240	0.215	0.245
8	0.0387	0.270	0.270	0.220	0.263
10	0.0489	0.300	0.300	0.226	0.283
12	0.0593	0.330	0.330	0.232	0.304
14	0.0701	0.370	0.380	0.239	0.328
16	0.0811	0.410	0.410	0.246	0.355
20	0.1037	0.500	0.500	0.260	0.417

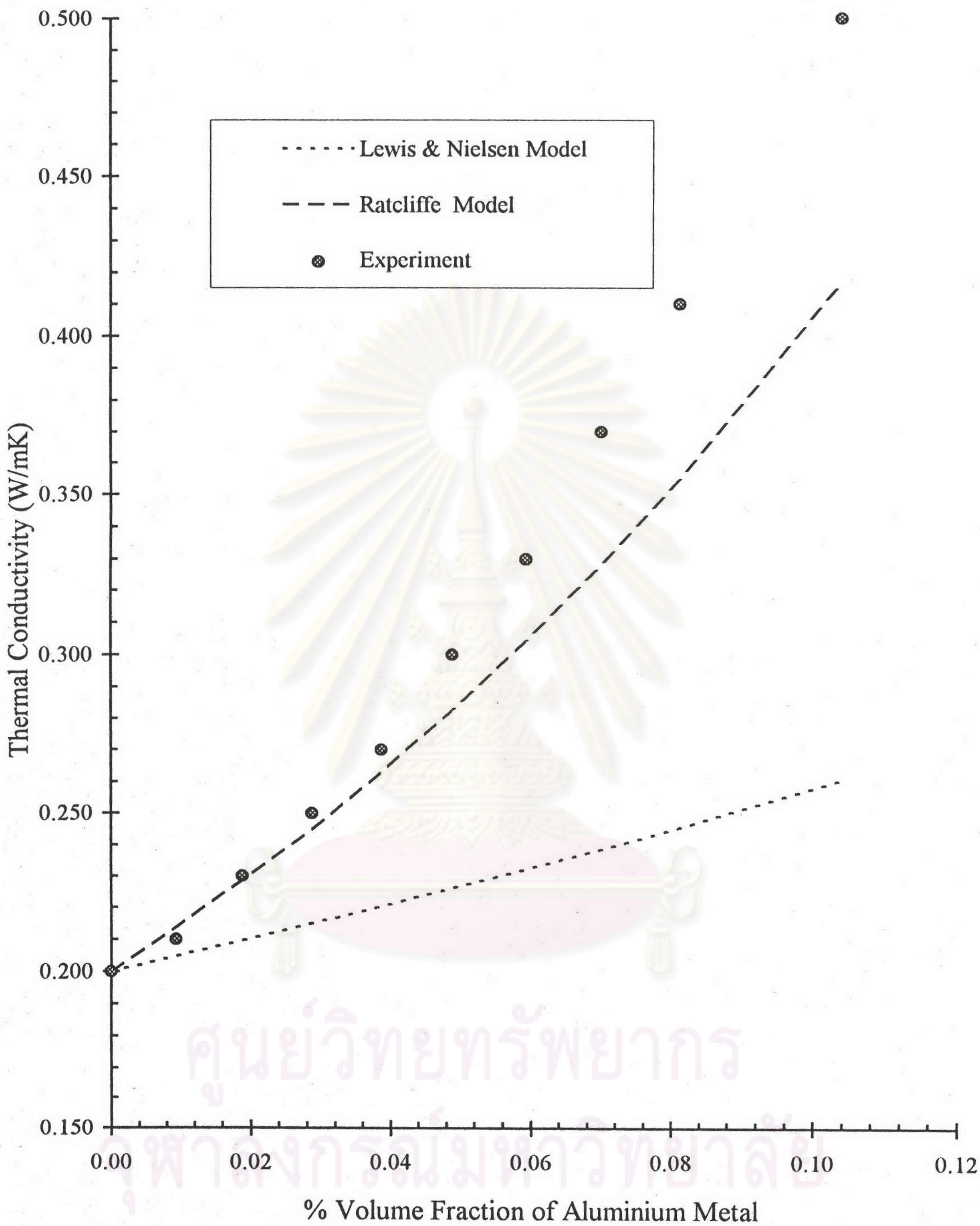

Figure 4.16 Comparison between the models and the experimental results for the composite of epoxy adhesive B filled with aluminium metal powder

Table 4.9 Thermal conductivity comparison between the models and the experimental data for the composite of epoxy adhesive A filled with berylium oxide

% Weight of fillers	% Volume fraction of fillers	Thermal conductivity (W/mK)			
		experiment 1	experiment 2	Lewis Model	Ratcliffe Model
0	0.0000	0.120	0.120	0.120	0.120
2	0.0078	0.130	0.130	0.122	0.127
4	0.0157	0.140	0.140	0.125	0.135
6	0.0239	0.150	0.150	0.127	0.144
8	0.0323	0.160	0.160	0.130	0.154
10	0.0409	0.170	0.170	0.133	0.165
12	0.0497	0.180	0.190	0.136	0.176
14	0.0587	0.200	0.200	0.139	0.189
16	0.0681	0.220	0.210	0.142	0.203
20	0.0875	0.270	0.270	0.150	0.236

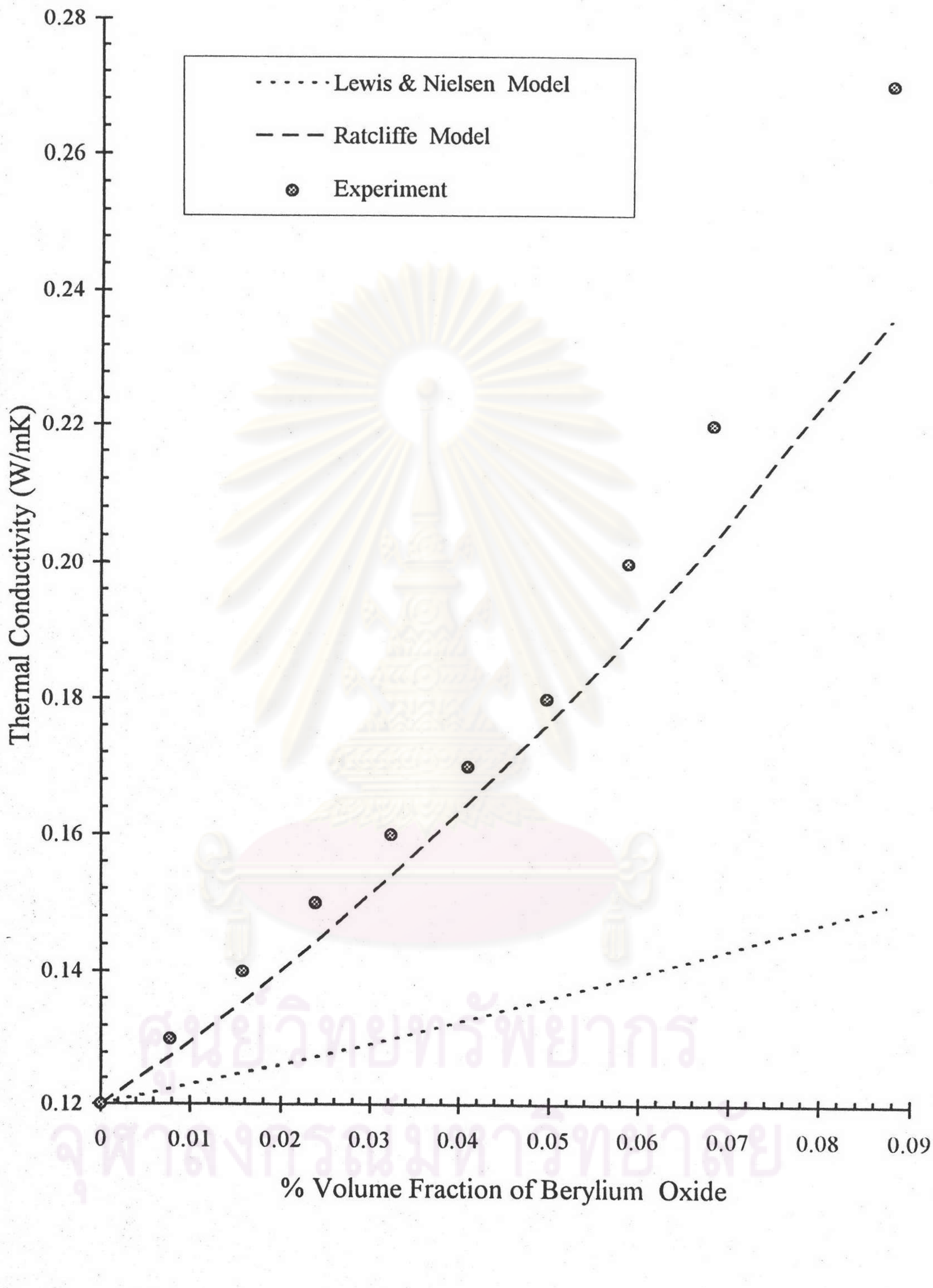

Figure 4.17 Comparison between the models and the experimental results for composite of epoxy adhesive A filled with berylium oxide powder

Table 4.10 Thermal conductivity comparison between the models and the experimental data for the composite of epoxy adhesive B filled with berylium oxide

% Weight of fillers	% Volume fraction of fillers	Thermal conductivity (W/mK)			
		experiment 1	experiment 2	Lewis Model	Ratcliffe Model
0	0.0000	0.200	0.200	0.200	0.200
2	0.0084	0.210	0.210	0.204	0.212
4	0.0171	0.220	0.220	0.209	0.226
6	0.0259	0.240	0.240	0.213	0.241
8	0.0352	0.260	0.270	0.218	0.257
10	0.0442	0.280	0.280	0.223	0.275
12	0.0538	0.300	0.300	0.229	0.295
14	0.0635	0.330	0.320	0.235	0.316
16	0.0735	0.360	0.350	0.241	0.340
20	0.0943	0.430	0.420	0.254	0.395

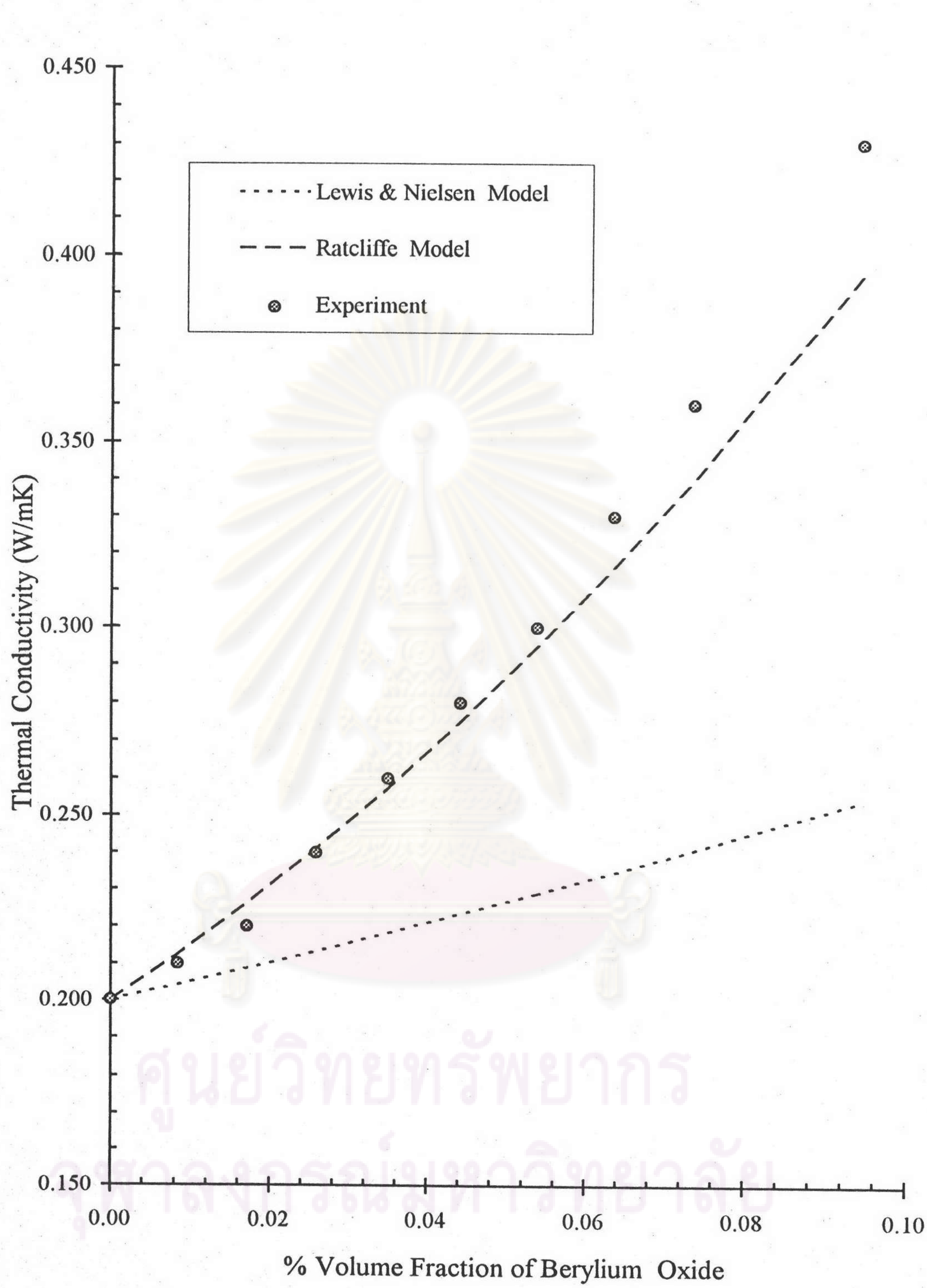

Figure 4.18 Comparison between the models and the experimental results for composites of epoxy adhesive B filled with berylium oxide powder

Table 4.11 Thermal conductivity comparison between the models and the experimental results for the composite of epoxy adhesive A filled with berrylium metal

% Weight of fillers	% Volume fraction of fillers	Thermal conductivity (W/mK)			
		experiment 1	experiment 2	Lewis Model	Ratcliffe Model
0	0.0000	0.120	0.120	0.120	0.120
2	0.0125	0.130	0.130	0.124	0.132
4	0.0252	0.150	0.140	0.128	0.145
6	0.0382	0.160	0.150	0.132	0.159
8	0.0513	0.180	0.180	0.137	0.176
10	0.0646	0.200	0.200	0.141	0.194
12	0.0781	0.220	0.220	0.146	0.214
14	0.0919	0.250	0.250	0.152	0.237
16	0.1059	0.290	0.280	0.157	0.263
20	0.1345	0.380	0.390	0.170	0.325

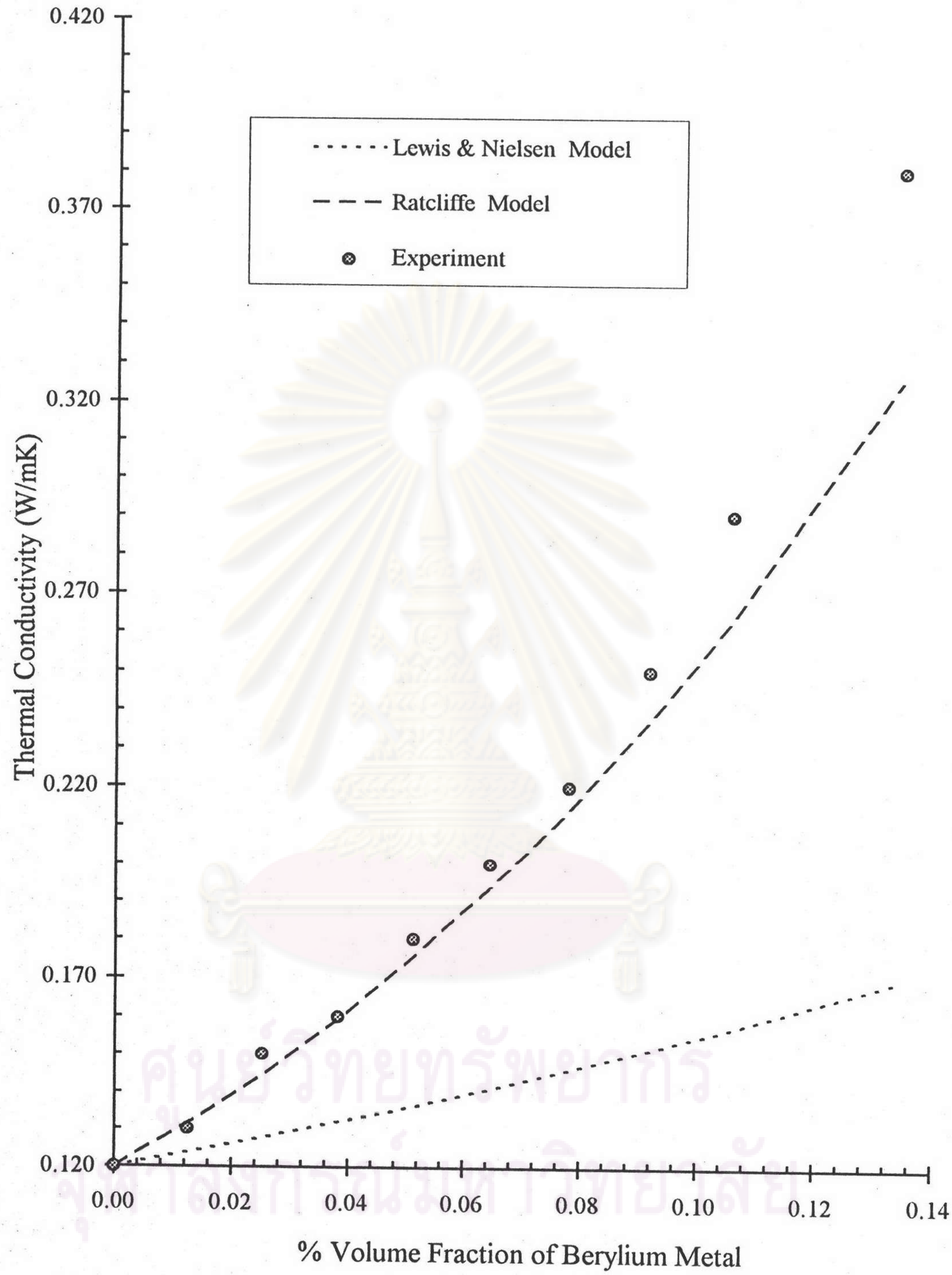

Figure 4.19 Comparison between the models and the experimental results for the composite of epoxy adhesive A filled with berylium metal powder

Table 4.12 Thermal conductivity comparison between the models and the experimental results for the composite of epoxy adhesive B filled with berylium metal

% Weight of fillers	% Volume fraction of fillers	Thermal conductivity (W/mK)			
		experiment 1	experiment 2	Lewis Model	Ratcliffe Model
0	0.0000	0.200	0.200	0.200	0.200
2	0.0136	0.220	0.220	0.207	0.220
4	0.0274	0.250	0.240	0.214	0.242
6	0.0413	0.270	0.270	0.222	0.266
8	0.0555	0.300	0.310	0.230	0.293
10	0.0698	0.330	0.330	0.239	0.324
12	0.0844	0.380	0.390	0.248	0.358
14	0.0991	0.430	0.450	0.257	0.397
16	0.1141	0.490	0.490	0.268	0.440
20	0.1445	0.640	0.630	0.290	0.543

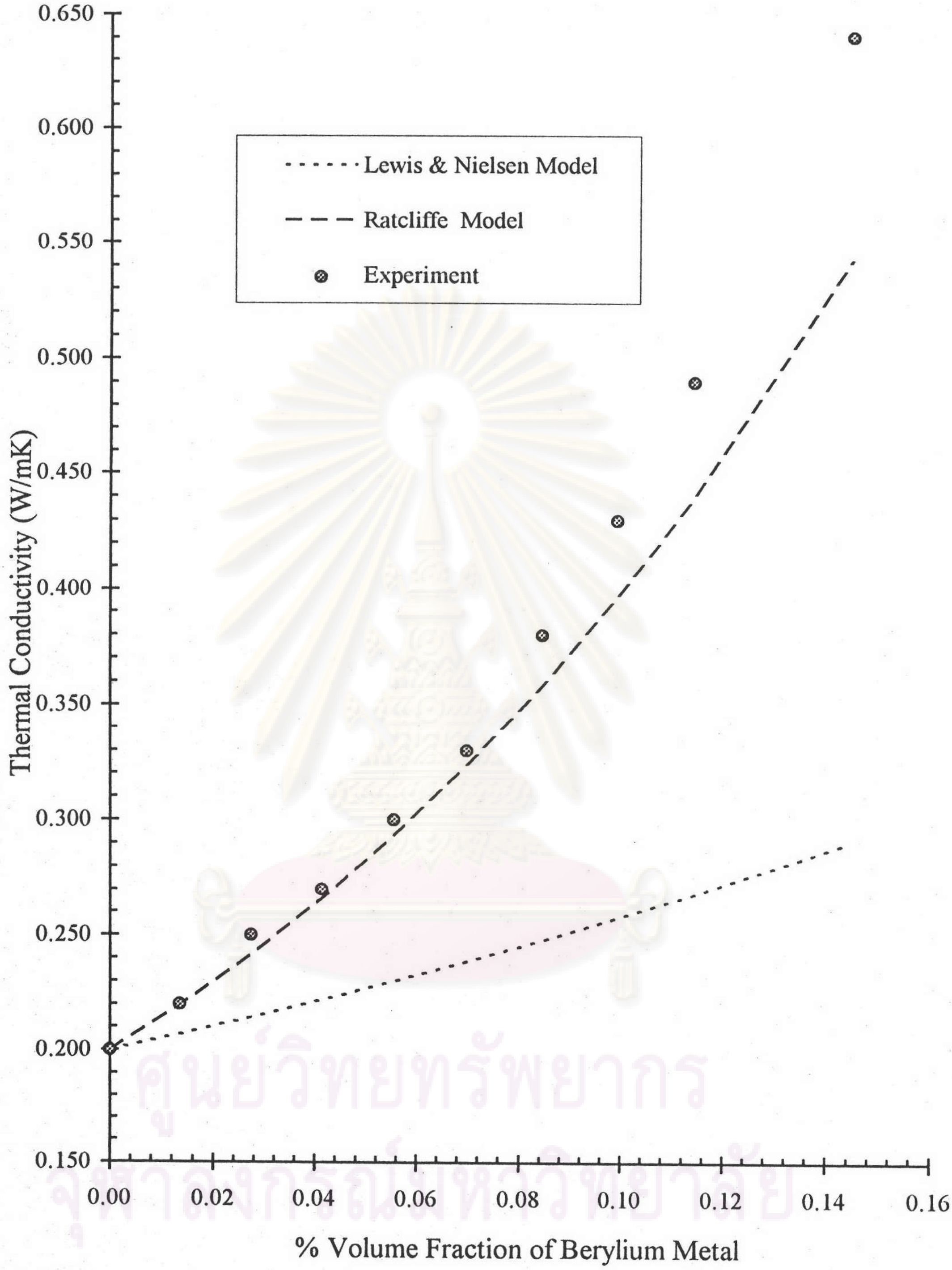

Figure 4.20 Comparison between the models and the experimental results for the composite of epoxy adhesive B filled with berylium metal powder

Table 4.13 Thermal conductivity comparison between the models and the experimental results for the composite of epoxy adhesive A filled with copper metal

% Weight of fillers	% Volume fraction of fillers	Thermal conductivity (W/mK)			
		experiment 1	experiment 2	Lewis Model	Ratcliffe Model
0	0.0000	0.120	0.120	0.120	0.120
2	0.0026	0.124	0.125	0.121	0.123
4	0.0053	0.128	0.130	0.122	0.125
6	0.0082	0.133	0.133	0.122	0.128
8	0.0111	0.138	0.138	0.123	0.131
10	0.0141	0.144	0.143	0.124	0.135
12	0.0173	0.149	0.150	0.125	0.138
14	0.0205	0.154	0.155	0.126	0.142
16	0.0239	0.158	0.159	0.127	0.146
20	0.0312	0.174	0.174	0.130	0.155

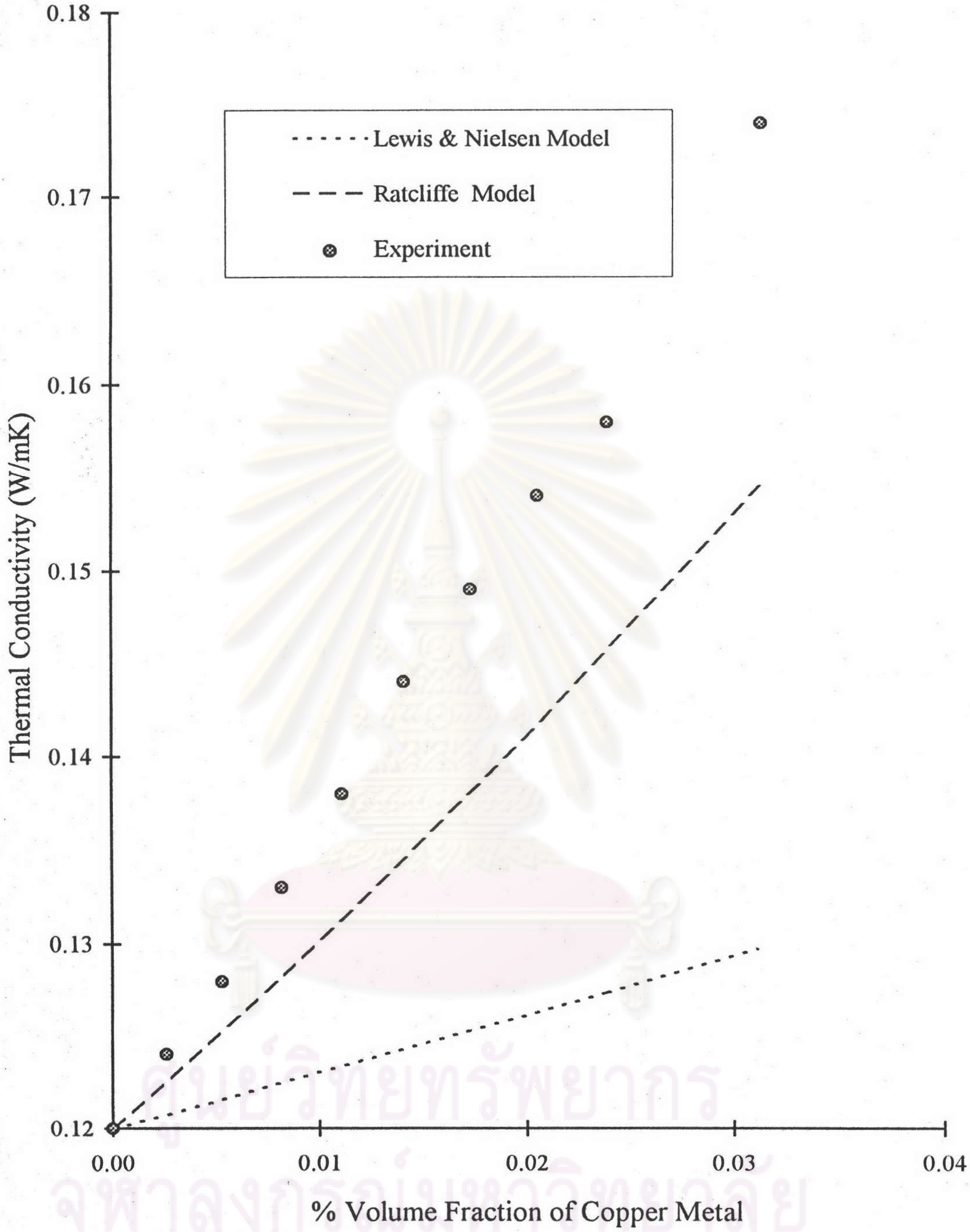

Figure 4.21 Comparison between the models and the experimental results for the composite of epoxy adhesive A filled with copper metal powder

Table 4.14 Thermal conductivity comparison between the models and the experimental results for the composite of epoxy adhesive B filled with copper metal

% Weight of fillers	% Volume fraction of fillers	Thermal conductivity (W/mK)			
		experiment 1	experiment 2	Lewis Model	Ratcliffe Model
0	0.0000	0.198	0.200	0.200	0.200
2	0.0028	0.204	0.204	0.201	0.204
4	0.0058	0.211	0.210	0.203	0.208
6	0.0089	0.217	0.217	0.204	0.213
8	0.0121	0.223	0.223	0.206	0.217
10	0.0153	0.229	0.229	0.208	0.222
12	0.0187	0.237	0.238	0.210	0.228
14	0.0223	0.246	0.245	0.211	0.233
16	0.0261	0.259	0.260	0.213	0.239
20	0.0338	0.282	0.282	0.218	0.253

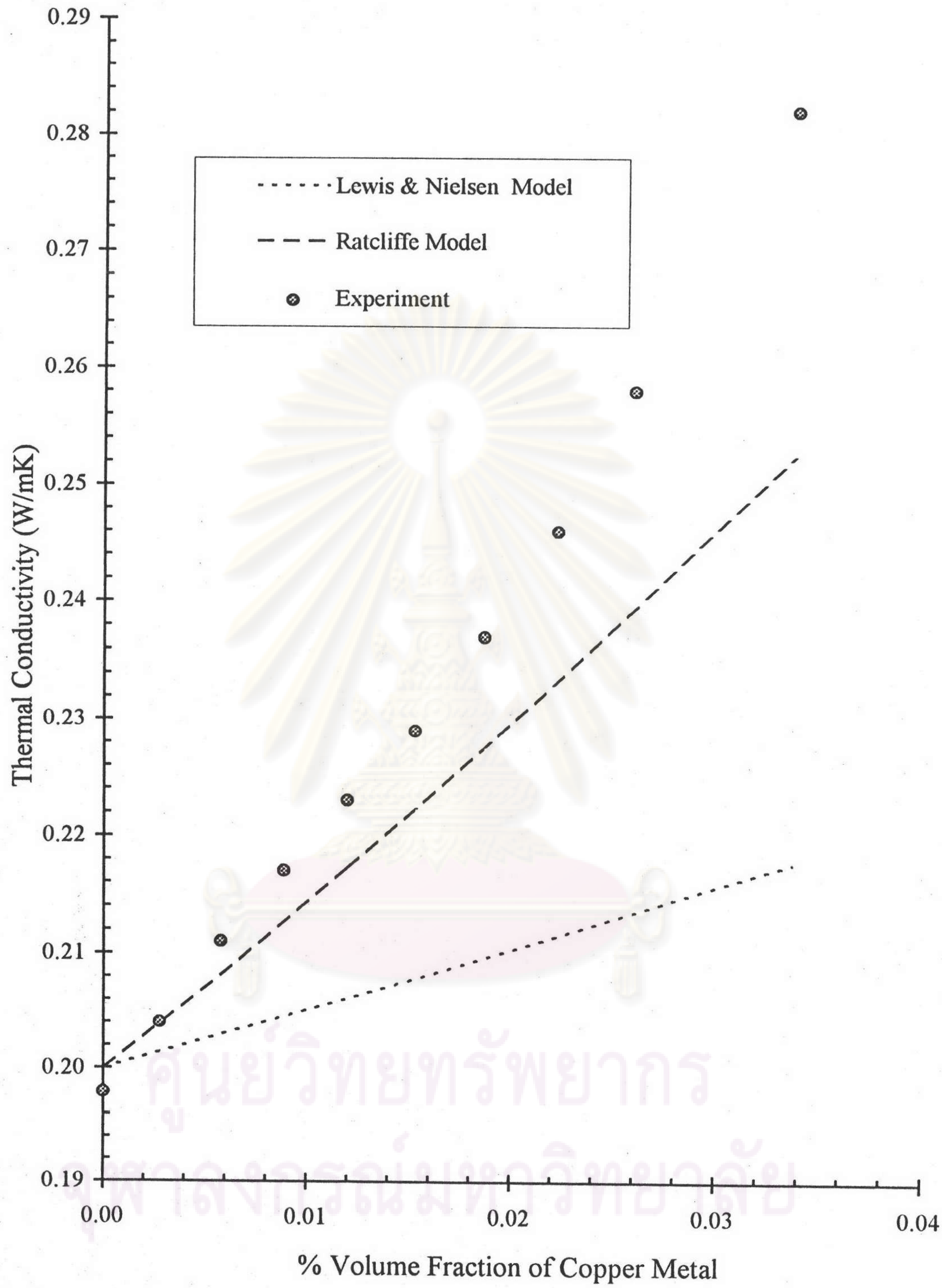

Figure 4.22 Comparison between the models and the experimental results for the composite of epoxy adhesive B filled with copper metal powder

Table 4.15 Thermal conductivity comparison between the models and the experimental results for the composite of epoxy adhesive A filled with silicon carbide

% Weight of fillers	% Volume fraction of fillers	Thermal conductivity (W/mK)			
		experiment 1	experiment 2	Lewis Model	Ratcliffe Model
0	0.0000	0.120	0.120	0.120	0.120
2	0.0074	0.125	0.125	0.122	0.127
4	0.0149	0.130	0.130	0.125	0.135
6	0.0227	0.138	0.137	0.127	0.144
8	0.0307	0.147	0.147	0.130	0.154
10	0.0389	0.158	0.159	0.132	0.165
12	0.0473	0.170	0.170	0.135	0.176
14	0.0559	0.180	0.180	0.138	0.189
16	0.0648	0.200	0.200	0.141	0.203
20	0.0834	0.240	0.240	0.148	0.236

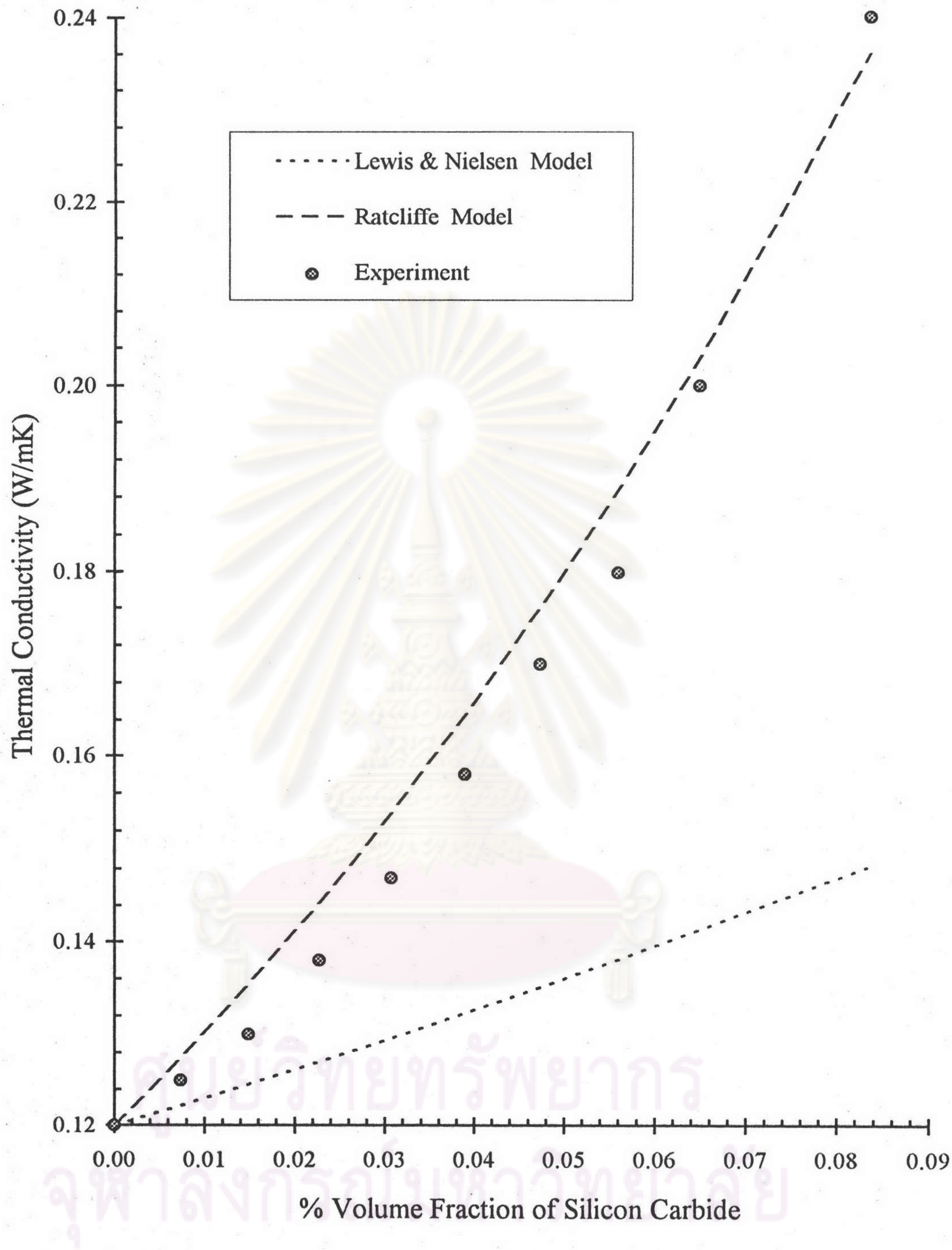

Figure 4.23 Comparison between the models and the experimental results for the composite of epoxy adhesive A filled with silicon carbide powder

Table 4.16 Thermal conductivity comparison between the models and the experimental results for the composite of epoxy adhesive B filled with silicon carbide

% Weight of fillers	% Volume fraction of fillers	Thermal conductivity (W/mK)			
		experiment 1	experiment 2	Lewis Model	Ratcliffe Model
0	0.0000	0.200	0.200	0.200	0.200
2	0.0081	0.210	0.210	0.204	0.213
4	0.0162	0.220	0.220	0.208	0.226
6	0.0246	0.230	0.230	0.213	0.241
8	0.0333	0.250	0.250	0.217	0.258
10	0.0421	0.270	0.270	0.222	0.275
12	0.0512	0.290	0.290	0.228	0.295
14	0.0605	0.320	0.320	0.233	0.317
16	0.0701	0.350	0.340	0.239	0.341
20	0.0901	0.420	0.420	0.251	0.396

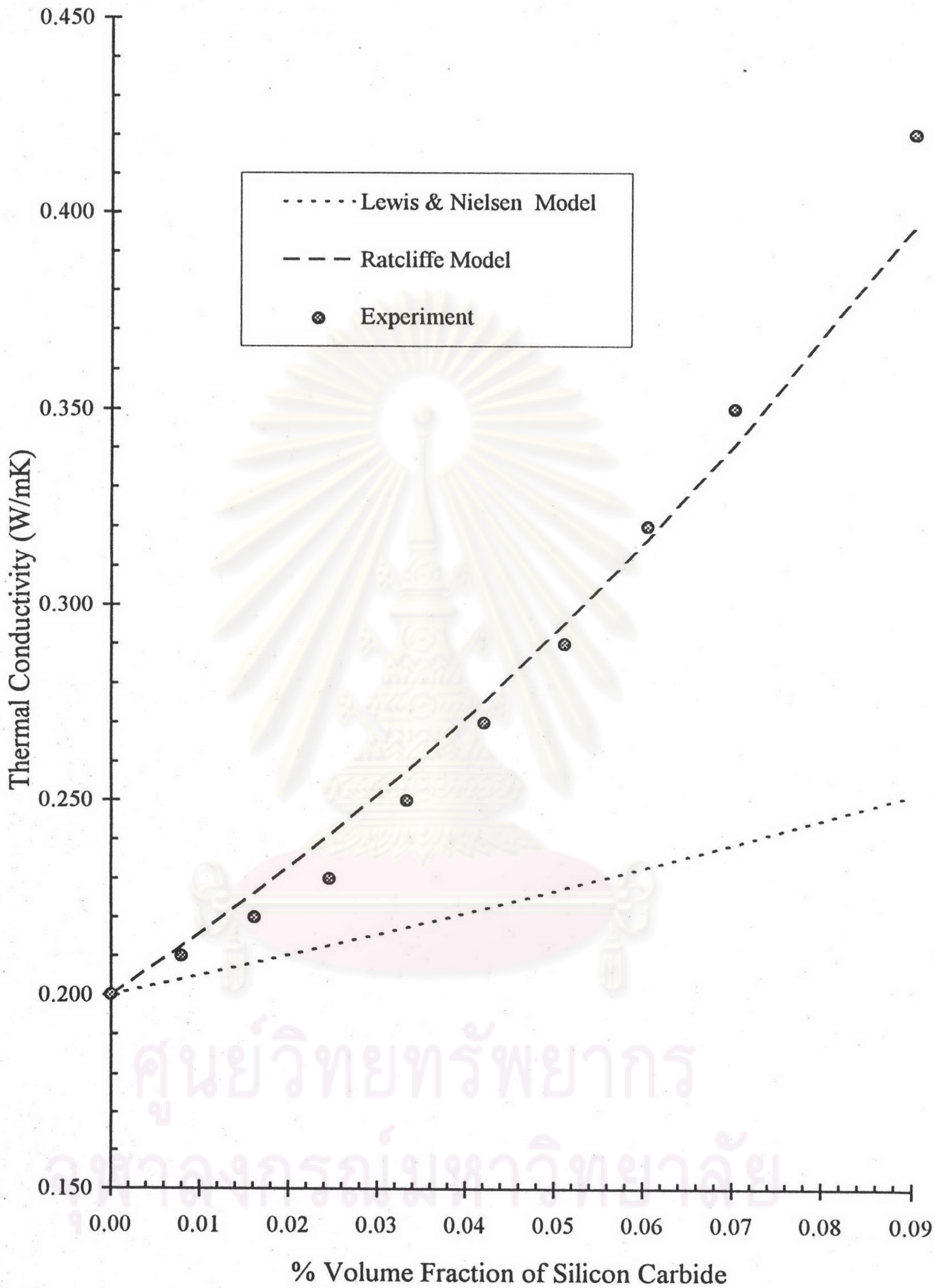

Figure 4.24 Comparison between the models and the experimental results for the composite of epoxy adhesive B filled with silicon carbide powder

In this study, the thermal conductivity of discrete phase or filler is two or three orders of magnitude larger than the thermal conductivity of the continuous phase or epoxy adhesive matrix. The thermal conductivity of matrix phase is 0.12 W/mK for epoxy adhesive A and 0.2 W/mK for epoxy adhesive B which is very different from the thermal conductivity of the fillers (in the range of 200-400 W/mK). These systems are the high contrast composites.

It is clear from Figure 4.15 to 4.24 that the Ratcliffe model is quite proper for explanation of these high contrast composites; therefore, we can use this model for the prediction of the thermal conductivity of epoxy adhesive composites.

From the Ratcliffe model, the thermal conductivity of these two phase systems depend on both the thermal conductivities of the matrix and the filler, and the weight percentage or volume fraction of the filler.

At high percent weight of filler in the composites, higher thermal conductivity than predicted from Ratcliffe model was measured; hewever, at low concentration of filler (the beginning of curve), the model and experimental result generally agree very well.

The Lewis and Nielsen semi-theoretical model shows the different curve from the experimental results. According to the theory, this model is proper to explain for solid composite system but not experimentally verified in this study.

4.5 Adhesion property

Because the objective of this study is to develop thermal conductivity of epoxy adhesive used in assembly process, the adhesion strength on assembly surface should be known. Adhesion property is an important factor that must be considered when increased thermal conductivity. The amount of filler particles increased in epoxy composites has direct impact on adhesion property.

To verify adhesion performance of adhesives in this study, shear strength testing procedure, ASTM : D1002-72, has been applied. This standard test method is for the determination of shear strength properties of adhesives by tension loading for metal-to-metal bonding. This method tests on a standard specimen and under specified conditions of preparation and test [27]. The adhesion performance of prepared epoxy adhesive composites was reported in Table 4.17 and Figure 5.25.

Table 4.17 Shear strength of epoxy adhesive composites

Epoxy adhesive	% weight filler	Shear strength (N/mm^2)				
		aluminium metal	berylium oxide	berylium metal	copper metal	silicon carbide
Epoxy adhesive A	0	27	27	27	27	27
	4	27	27	26	27	27
	8	27	26.5	25.5	27	27
	14	26.5	25.5	25	27	26.5
	20	25	24.5	23.5	26	25.5
Epoxy adhesive B	0	32	32	32	32	32
	4	32	32	32	32	32
	8	32	32	31	32	32
	14	31	31	31	32	31
	20	30	30	29	31	30.5

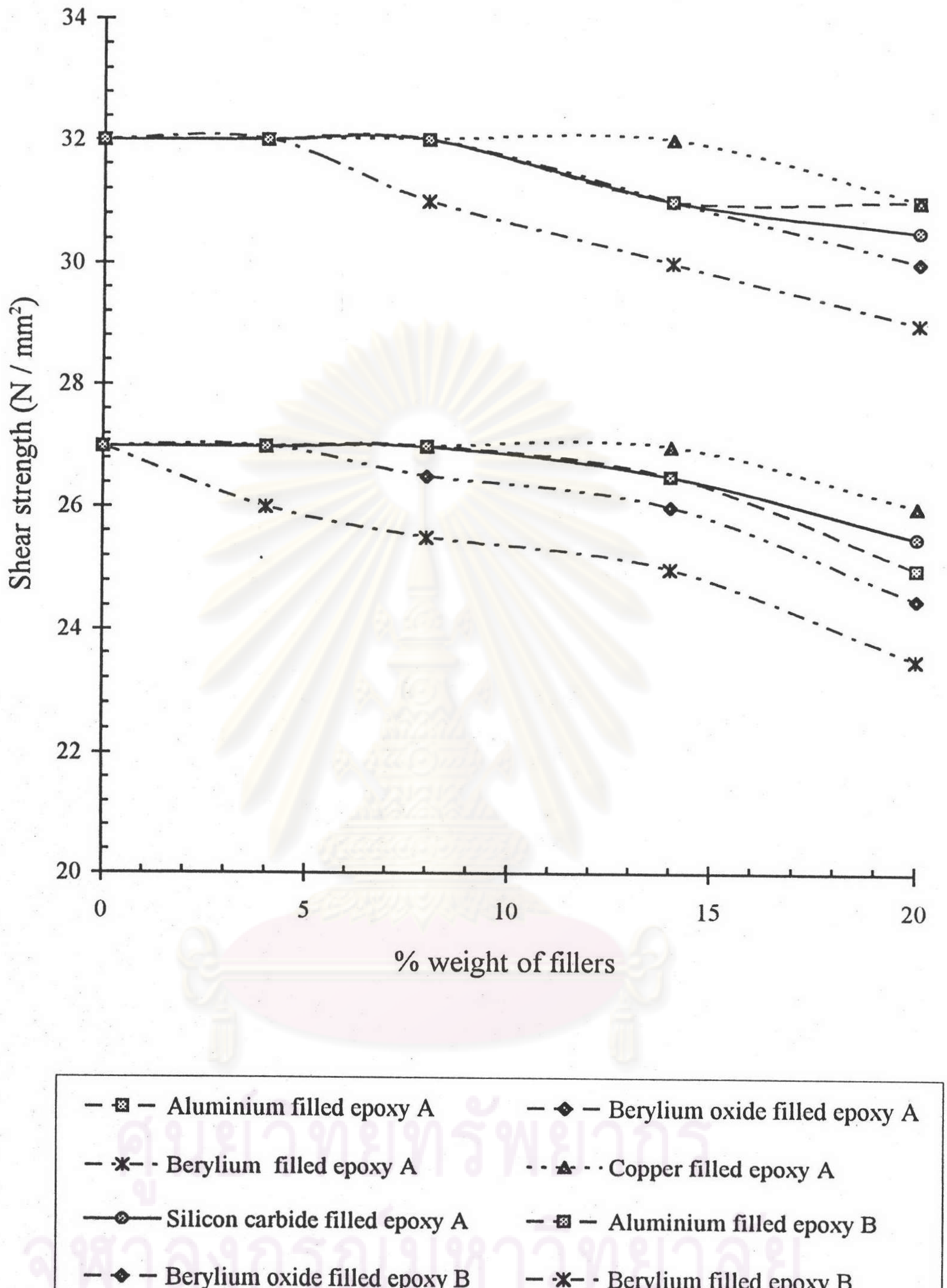

Figure 4.25 Shear strength of epoxy adhesive composites

The results show some reduction of adhesion strength. An adhesion performance decreases around 9 to 12 percent when the epoxy adhesive composites have 20 % weight of fillers. All prepared epoxy adhesive composites in this experiment still have enough adhesion property. This means that the maximum percentage, 20 percent weight of filler used to develop thermal conductivity of epoxy adhesive in this study still does not change the property of bonding.

The shear strength varies reverse to the increasing of filler percentage. In this study, the thermal conductivity values of prepared composites with 20 % weight of filler are high enough for heat dissipation of PCB assembly equipment, so it is not necessary to add more fillers. Since too many fillers added cause the adhesion strength to reduce and the material cost to increase. (see price of fillers in Appendix D)