Transient Characteristics of Thermal Conduction in Dispersed Composites[1]

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ABSTRACT

The effective thermal conductivity of dispersed composites with hot melt adhesive matrix measured using the steady-state method is compared with the apparent thermal conductivity which is calculated with the average heat capacity and the thermal diffusivity measured by the laser flash method. All the experimental results are compared with those calculated by some existing models and FEM. The criterion for the homogeneity of dispersed composites under transient conditions or the limitation of the concept of effective thermal diffusivity as a transient-state characteristic constant is discussed for various percentage of dispersed phase, dispersed particle size, and ratio of the thermal conductivity values of dispersed and continue phase.

KEY WORDS: criterion of homogeneity; dispersed composites; FEM; laser flash method; thermal conductivity; thermal diffusivity; steady comparison method.

1. INTRODUCTION

For heterogeneous materials, an effective thermal conductivity can be defined by the extended Fourier law, with the average value of the temperature gradient over a region large in comparison with the size of the heterogeneity. Considerable progress has been made in relating the effective steady-state thermal conductivity to those of individual components. Under transient state, however, there is no simple models to relate the heterogeneous thermal diffusivity to that of the individual components as in steady-state case. In fact, as pointed by Kerrisk [1, 2], the meaning of the thermal diffusivity as a characteristic property of a heterogeneous material is not clear since the thermal conduction equation in which the thermal diffusivity appears as a transient-state characteristic constant applies only to homogeneous materials. Then an effective thermal diffusivity for transient-state thermal problem should be defined under the condition that the heterogeneous material can be considered as a homogeneous material. Experimentally, the criterion of homogeneity was studied in an early paper of Lee and Taylor [3], but an obvious difference between the results by laser flash method and steady-state model was not observed under 30 volume% of dispersed particles. And no further work for high volume percentage has been found in the literature.

The purpose of the present work is to investigate the criterion for the homogeneity of dispersed composites under transient conditions experimentally. The effective thermal conductivity measured using the steady-state method is compared with the apparent thermal conductivity which is calculated with the average heat capacity and the thermal diffusivity measured by the laser flash method for a wide region of volume percentage. Furthermore, the experimental results are compared with those calculated by some

a transient-state characteristic constant of a dispersed composites is discussed for various volume percentages of dispersed phase, dispersed particle size, and ratio of the thermal conductivity values of dispersed and continue phase.

2. EXPERIMENTS

Experimentally, the transient characteristic of heat conduction in dispersed composites can be investigated by comparing the values of thermal conductivity from the steady and unsteady state methods.

2.1. Measuring methods

In the present work, the widely used steady comparison method is employed as the steady state method to measure the effective thermal conductivity λ_e. For the unsteady state method, the famous laser flash method is employed to measure the thermal diffusivity a, then the apparent thermal conductivity λ_a can be obtained by the following,

$$\lambda_a = (c\rho)_m a \qquad (1)$$

for two-phase dispersed composites,

$$(c\rho)_m = c_d \rho_d V_d + c_c \rho_c (1 - V_d) \qquad (2)$$

where c, ρ, and V are density, heat capacity, and volume percentage, the subscripts d and c represent the values of dispersed and continue phase, respectively.

2.2. Specimens

Two different kinds of two-phase dispersed composites are prepared for the measurement. First, Aluminium spheres dispersed in hot melt adhesive and second, Alumina spheres dispersed in hot melt adhesive. The hot melt adhesive is composed of

mass), and Antioxidant (0.498% by mass). The thermophysical properties of the dispersed and continue phases are given in Table 1.

The preparation procedure of the specimens can be simply described as follows: The hot melt adhesive in a stainless container is heated to 150; determined amounts of dispersed particles are added into the molten adhesive; the uniformly mixed particle-hot melt adhesive mix is filled into a mold and degassed in a vacuum oven at 120-160 and 1 torr, then cooled to room temperature. The uniform distribution of the dispersed particles has been observed at the profile of the composites by microscope. The specimens for the steady comparison method (with diameter of 30 mm and thickness of 15 mm) and laser flash (with diameter of 10 mm and thickness of 1.5 mm) are listed in Table 2. The two kinds of specimens are made from the same particle-hot melt adhesive mixture.

2.3. General description of the experimental results

The results of the thermal conductivity of the two kinds of composites measured by the steady comparison method and the laser flash method are shown by a representative selection of curves in the figures. Fig.1 shows the variation of the thermal conductivity with volume percentage of three kinds of the dispersed particles for steady and laser flash method. Fig.2 shows the dependence of thermal conductivity on the size of dispersed particles for the two kinds of measuring methods. By some selected comparisons of the experimental results, the effects of volume percentage, particle size, and conductivity ratio on the conductivities of the composites are found and will be described as follows.

(1) Effect of volume percentage

In Fig.1, it is easy to find that these three groups of results have a common feature,

conductivity ratio. That is, for a small volume percentage, as in homogenous materials thermal conductivities from steady and laser flash method have the same values. With increasing of volume percentage, the difference between steady and unsteady method appears, i.e. the materials begin to show there heterogeneity or transient-state feature. And the bigger the volume percentage is, the more obvious the difference is.

(2) Effect of particle size

The effect of the particle size is demonstrated by comparison of (a) and (b) of Fig.1. For the composites with 35 μm (median diameter with standard deviation of 25 μm) dispersed particles, the difference of thermal conductivity between the steady and the unsteady method are still undistinguished until the volume percentage reaches about 25%. For the one with 75 μm dispersed particles, however, the difference appears at about 15%. The a change in dispersed particle size may result in the criteria shift of the range in which the composites can be treated as homogeneous materials. The bigger the size of dispersed particle is, the more narrow the range of homogeneity is. Fig.2 shows the conductivity change with particle size for a fixed volume percentage of 20%. For each of particle size, the results from steady and unsteady method are same. But the conductivities at the side of big particle size are obviously bigger than that at the side of small particle size. And this phenomenon can be also found for other volume percentages over 20% by comparing (a) and (b) of Fig.1.

(3) Effect of conductivity ratio

Comparing (a) and (c) of Fig.1 demonstrates the effect of the ratio of thermal conductivity of dispersed particles to that of continue phase. For Aluminium-hot melt composites (Fig.1(a)), the thermal conductivity ratio is $l_p/l_m = 1364$; while for

Alumina-hot melt composites, $l_d / l_c = 205$. Because the thermal conductivity of Alumina is smaller than that of Aluminium, all the results in (c) are smaller than the corresponding ones in (a). A obvious difference in the heterogeneity criteria for these two figures is not observed. Comparing with the other two parameters, the effect of conductivity ratio is small.

3. COMPARISON WITH CALCULATIONS AND DISCUSSION

3.1. Comparison with some well-used models

Fig.3 and 4 show comparisons of the predictions of four well-used models (the pararell model [4], the series model [4], the Maxwell model [5], and the Bruggman model [6]) with the experimental values obtained from measured data for Aluminium-hot melt adhesive and Alumina-hot melt adhesive composites respectively. In Fig.3, for the specimens with 35 μm dispersed particles, the agreement of the experiment and the four models is quit good for volume percentage up to 20%, while at higher volume percentage the experimental results are approximately agree with Bruggman model and larger than the other models. For the specimens with 75 μm particles, at the higher volume percentage the results from laser flash method are much larger than all of the four models. In Fig.4, at quite wide region of volume percentage the agreement of experimental results measured by steady state method and the models is quite good, while at higher volume percentage the results of laser flash method are little larger than the models.

All of the pararell, series, and Maxwell model are given under the assumption that the particles are in dilute dispersions, then for a high volume percentage of dispersed particles they could become invalid. The Bruggman model is derived on the basis of an

it can be available for a wide range of volume percentage of dispersed particles. For big particle sizes or the results from unsteady state method at higher volume percentage, Bruggman model becomes quite inaccurate.

3.2. Comparison with numerical calculation

3.2.1. A simplified model of dispersed composites and calculation method

The dispersed composites is considered to be described by a simply three-dimensional model as shown in Fig.5. The sphere dispersed particles are arranged in a cubic regular array. The whole composites is formed by piling up the unit cells, in which the dispersed particles are located at the center of the cubes. The thermal conductivities of the composites are determined by numerical analysis using the finite element method (FEM) under the conditions of steady and unsteady state.

3.2.2. Transient temperature response

Fig.6 demonstrates the temperature responses at the center ($X = 0, Y = 0$)of rear surface of a single cell and a piles of N cells heated by a pulse heat flux at the front surface. With the increase of number N, the temperature responses becomes slower and slower, finally they approach to a same curve. It is means that increasing the number of unit cells in the calculation may have the specimen homogenized.

For the situation in which N is big enough that the composites can be treated as homogeneous materials, the transient thermal diffusion may be described by an effective property, effective thermal diffusivity, which can be measured by laser flash method. The relationship between effective thermal diffusivity a_e and effective conductivity l_e is expressed as Eq.(2) with a_e and l_e instead of a and l_a respectively.

effect is considered for the situation that Aluminium particles are dispersed in hot melt adhesive (as shown in Fig.7). Here the criterion of homogeneity is given as the calculated thermal conductivities approach a fixed value. The figure shows that, when N>3 for 10% and N>6 for 40%, the composites may be treated as homogeneous materials.

3.2.3. Comparison of steady and unsteady method

The changes of apparent thermal conductivity with volume percentage calculated under steady are compared with those under unsteady condition for Aluminium-hot melt adhesive composites. The calculations are performed with enough unit cell numbers that the composites are treated as homogeneous materials. The differences in results between steady state method and laser flash method for all volume percentages are in the range of calculation errors. This means that the transient effect observed in experiments does not occur in calculation.

3.2.4. Comparison with experimental results

Fig.8 shows comparison of the calculated results with the experimental ones for 75 μm Aluminium-hot melt adhesive dispersed composites. In the figure, the calculated results for cell number N from 1 to 8 are demonstrated, for small N the results of unsteady state are quite different from each other and from those of steady state, while for big N all the results approach the same. When N=2, the curve obtained from the calculation is similar to that obtained from the experiment. For other values of N, big differences between calculation and experiment are observed. However, the real specimens (with thickness of 1.5 mm, 75 μm dispersed particles) may contain at least 10 unit cells in the direction of thickness. The obvious transient effect observed in experiments means that, for real composites, the macroscopic heterogeneity is important

4. CONCLUDING REMARKS

The criterion for the homogeneity of dispersed composites under transient conditions or the limitation of the concept of effective thermal diffusivity as a transient-state characteristic constant is investigated experimentally and theoretically. The differences between steady and unsteady method, experiment and calculation are discussed for various percentage of dispersed phase, dispersed particle size, and ratio of the thermal conductivity values of dispersed and continue phase. The obvious transient effect observed in the experiments at higher volume percentage can not predicted by the simplified calculation model with dispersed particles in regular array. The complexity of actual dispersion systems is certainly one reason. The thermal bridge caused by percolation effect may be another reason especially for high volume percentages.

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FIGURES CAPTIONS

Fig.1. Comparisons of thermal conductivities measured by steady and laser flash method.

(a) 35 μm Al (b) 75 μm Al (c) 35 μm Al_2O_3

Fig.2. Thermal conductivity of Al-hot melt adhesive dispersed composites with different size of particles at the same volume percentage (20%).

Fig.3. Comparison of measured results with models (Al).

Fig.4. Comparison of measured results with models (Al_2O_3).

Fig.5. Three-dimensional unit cell model of dispersed composites.

Fig.6. Temperature responses of Al-hot melt adhesive composites for various numbers of unit cells.

Fig.7. Effect of unit cell number on thermal conductivity.

Fig.8. Comparison of experimental and numerically calculated results.

Table 1. Properties of dispersed and continue phase

	Thermal conductivity ($Wm^{-1}K^{-1}$)	Heat capacity ($kJKg^{-1}K^{-1}$)	Density (kgm^{-3})
Hot melt adhesive	0.176	1.58	963
Aluminium	240	0.905	2700
Alumina	36	0.779	3880

Table 2. List of specimens

	Dispersed particle									
	Aluminium								Alumina	
Diameter (μm)	6		35		75		100		35	
Measuring method	S.C.	L.F.	S.C.	L.F.	S.C.	L.F.	S.C.	L.F.	S.C.	L.F.
Volume percentage (%)	20	20	0 5 10 15 20 30 40 50	0 5 10 15 20 30 40 50	0 5 10 20 30 45 50	0 5 10 20 30 45 50	20	20	0 5 10 20 30 40 50	0 5 10 20 30 40 50

S.C.: Steady Comparison Method; L.F.: Laser Flash Method.

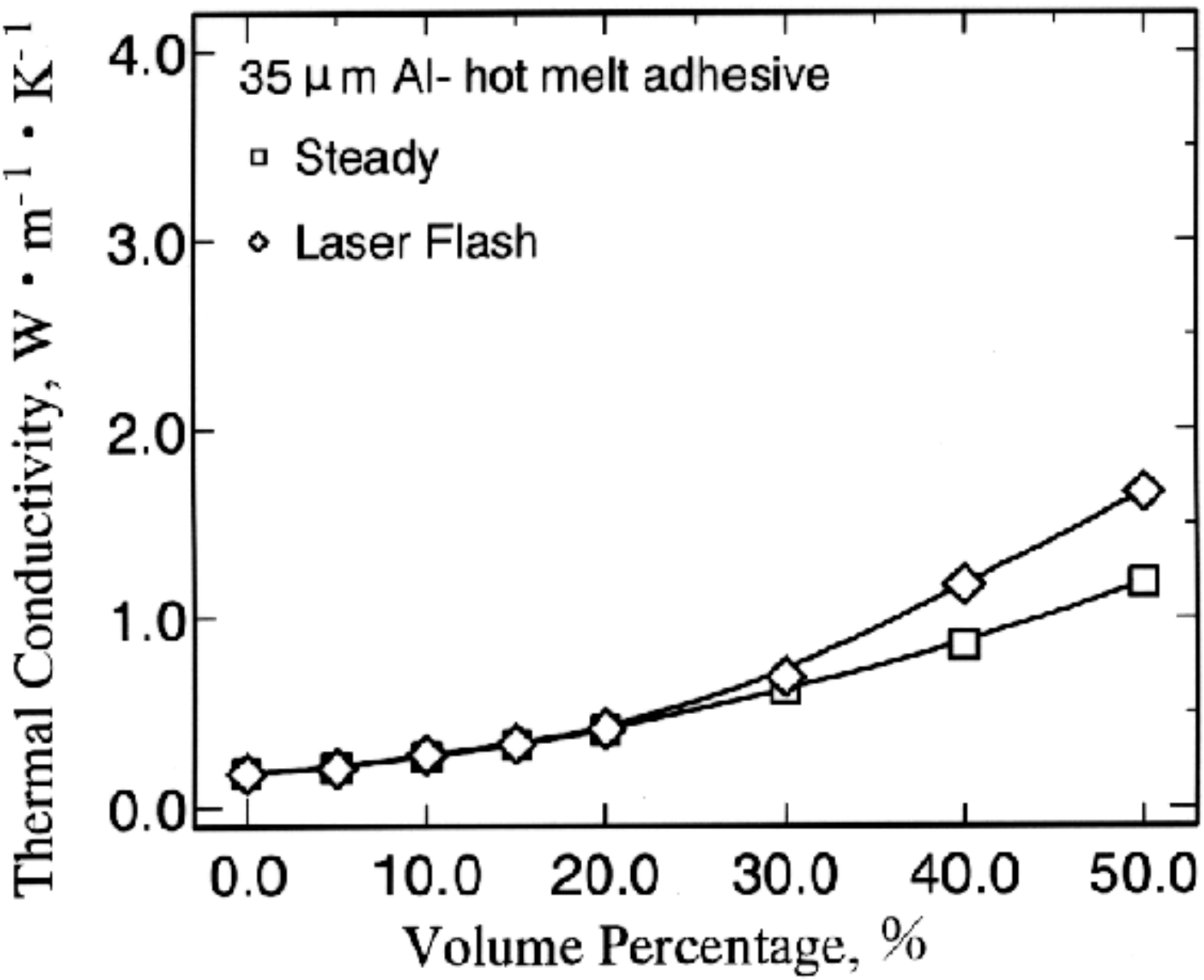
35 μm Al- hot melt adhesive
Steady
Laser Flash
Thermal Conductivity, W • m-1 • K-1
4.0
3.0
2.0
1.0
0.0
0.0
10.0
20.0
30.0
40.0
50.0
Volume Percentage, %

Figure 1(a)

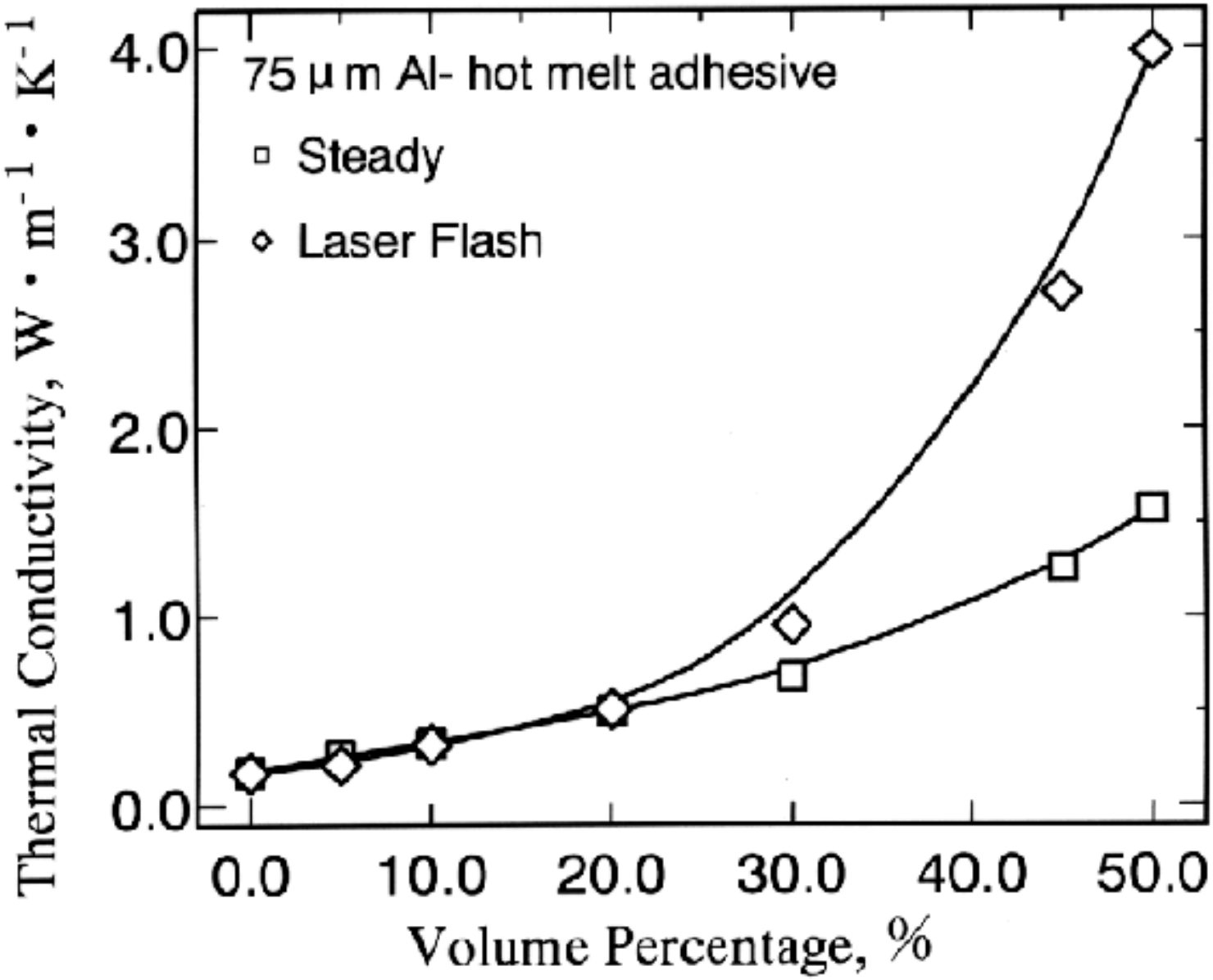
75 μ m Al- hot melt adhesive
Steady
Laser Flash
Thermal Conductivity, W • m-1 • K-1
4.0
3.0
2.0
1.0
0.0
0.0
10.0
20.0
30.0
40.0
50.0
Volume Percentage, %

Figure 1(b)

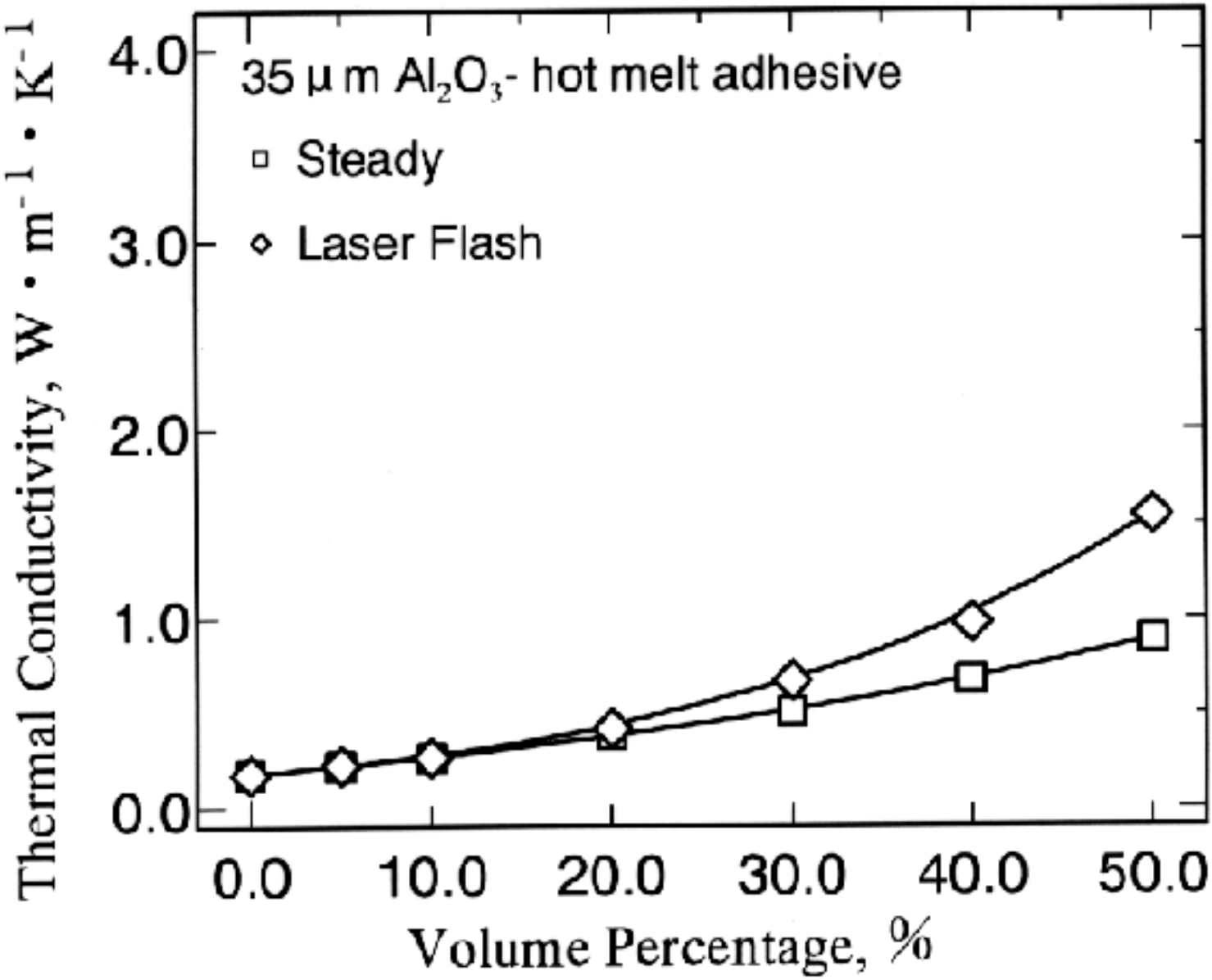
35 μ m Al_2O_3- hot melt adhesive
Steady
Laser Flash
Thermal Conductivity, W • m^{-1} • K^{-1}
4.0
3.0
2.0
1.0
0.0
0.0
10.0
20.0
30.0
40.0
50.0
Volume Percentage, %

Figure 1(c)

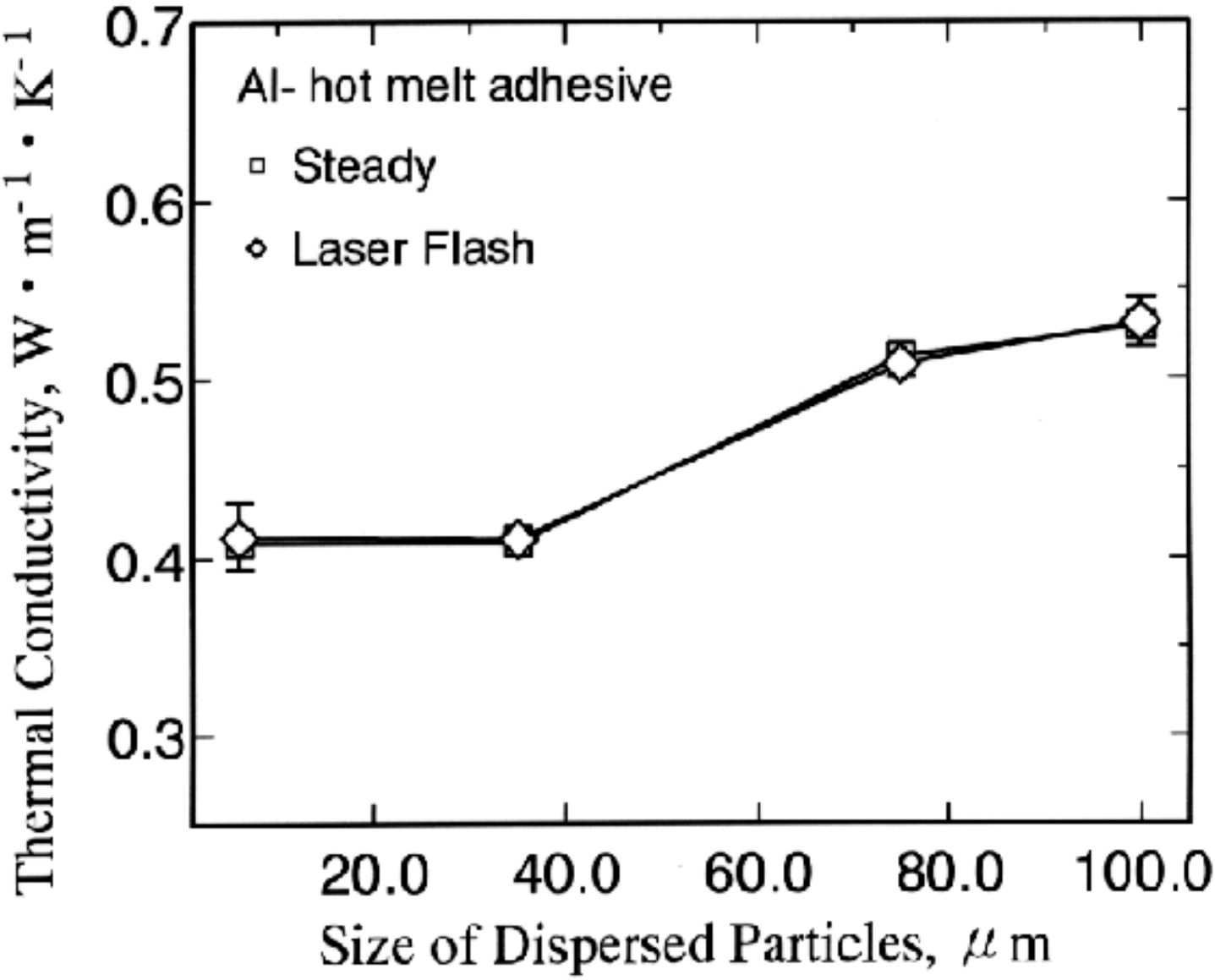
Al- hot melt adhesive
Steady
Laser Flash
0.7
0.6
0.5
0.4
0.3
Thermal Conductivity, W • m-1 • K-1
20.0
40.0
60.0
80.0
100.0
Size of Dispersed Particles, μ m

Figure 2

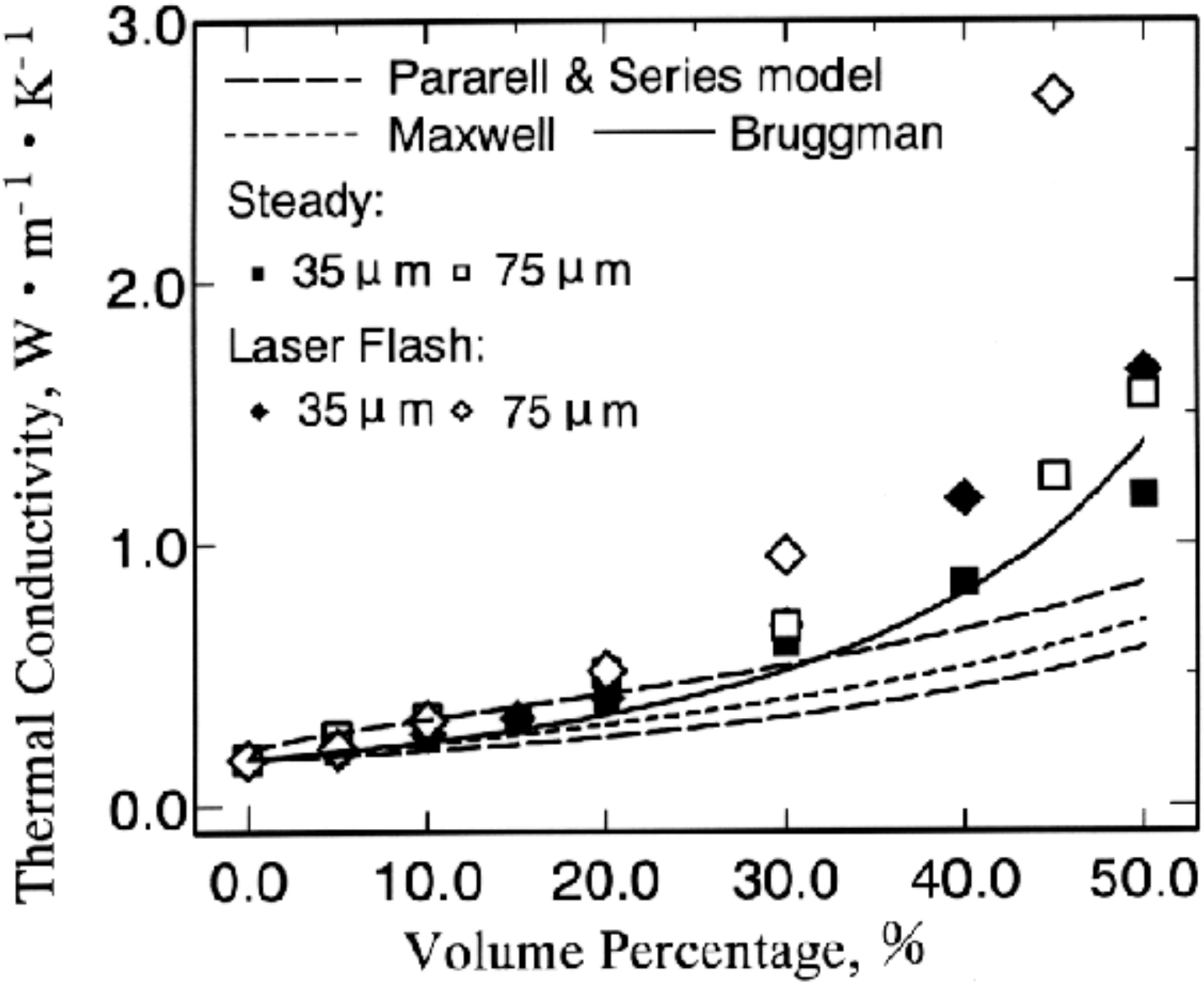
Pararell & Series model
Maxwell
Bruggman
Steady:
35 μm
75 μm
Laser Flash:
35 μm
75 μm
3.0
2.0
1.0
0.0
0.0
10.0
20.0
30.0
40.0
50.0
Thermal Conductivity, W • m-1 • K-1
Volume Percentage, %

Figure 3

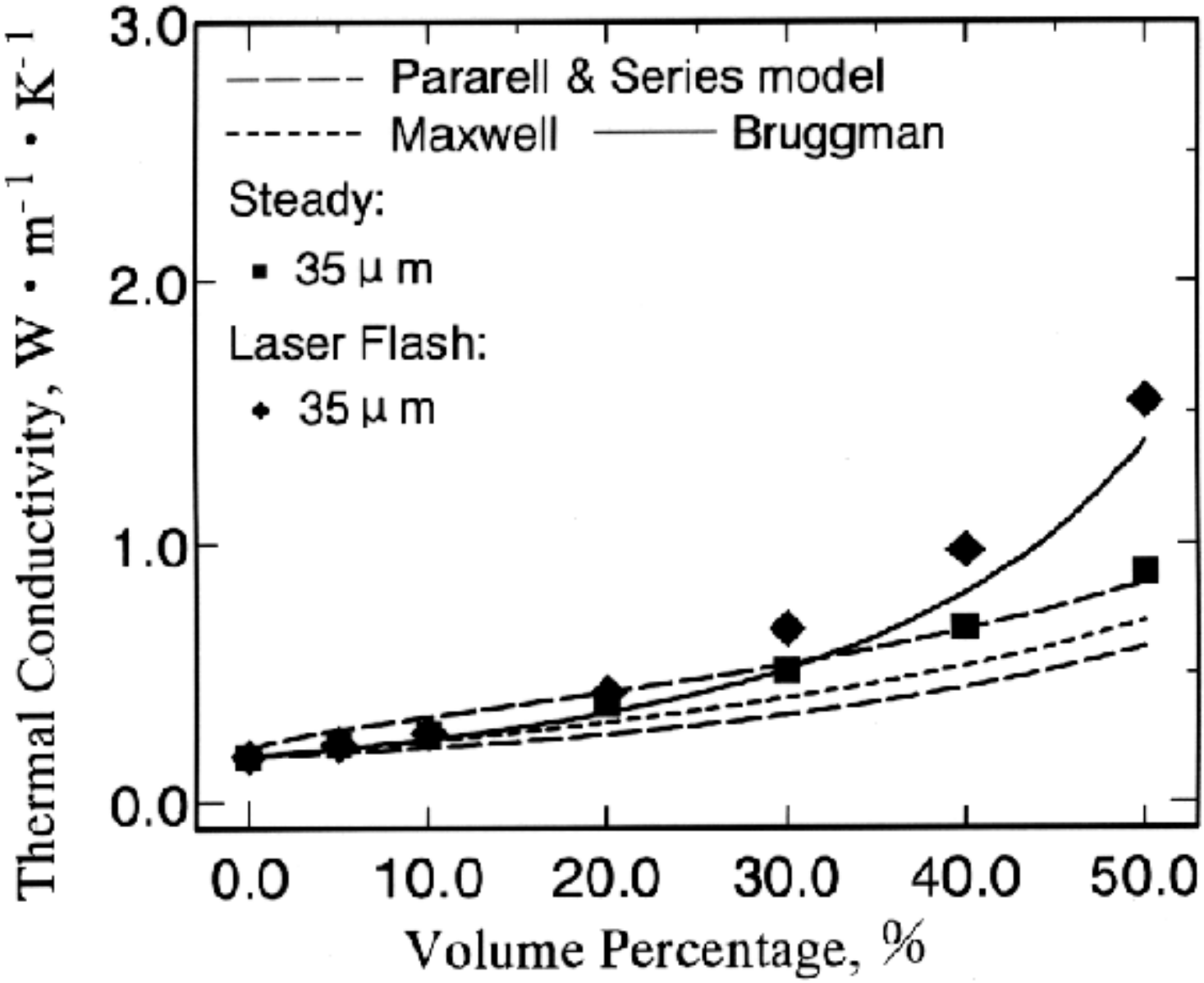
Pararell & Series model
Maxwell
Bruggman
Steady:
35 μ m
Laser Flash:
35 μ m
3.0
2.0
1.0
0.0
0.0
10.0
20.0
30.0
40.0
50.0
Thermal Conductivity, W • m-1 • K-1
Volume Percentage, %

Figure 4

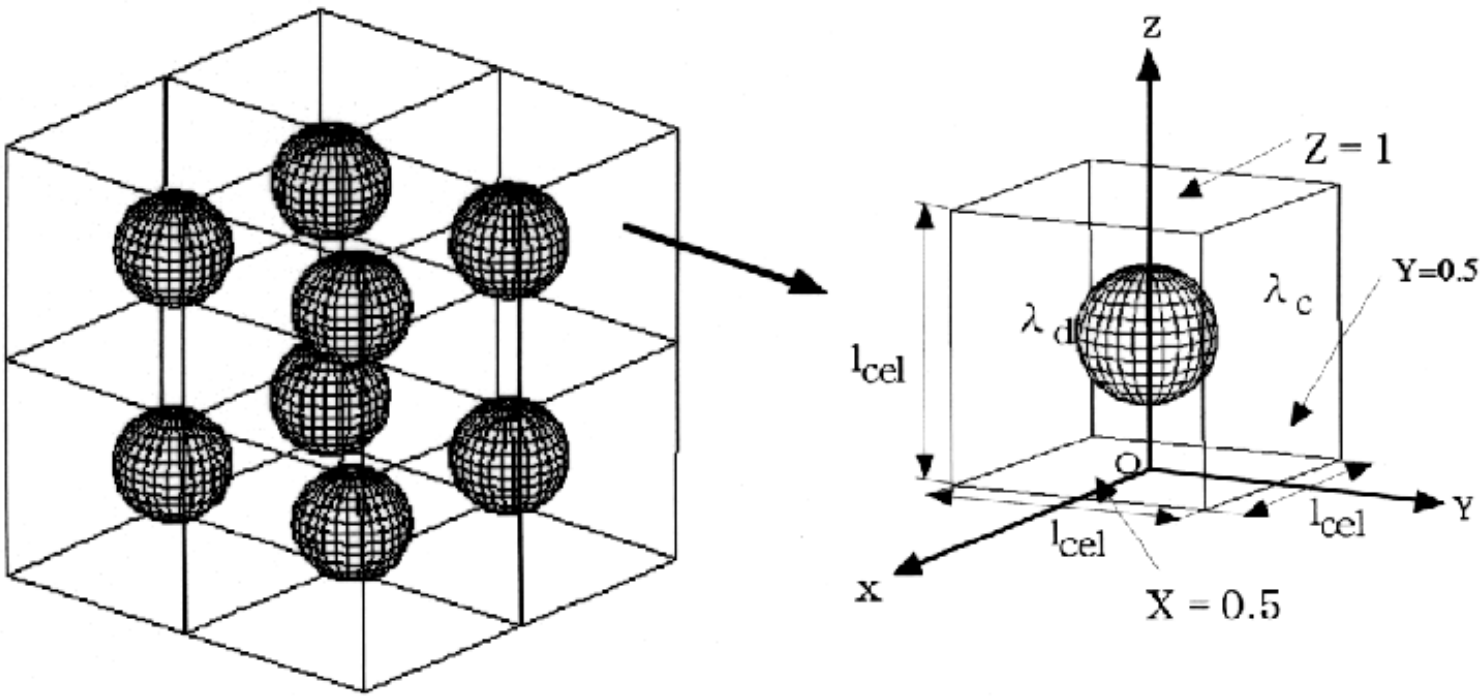

Figure 5

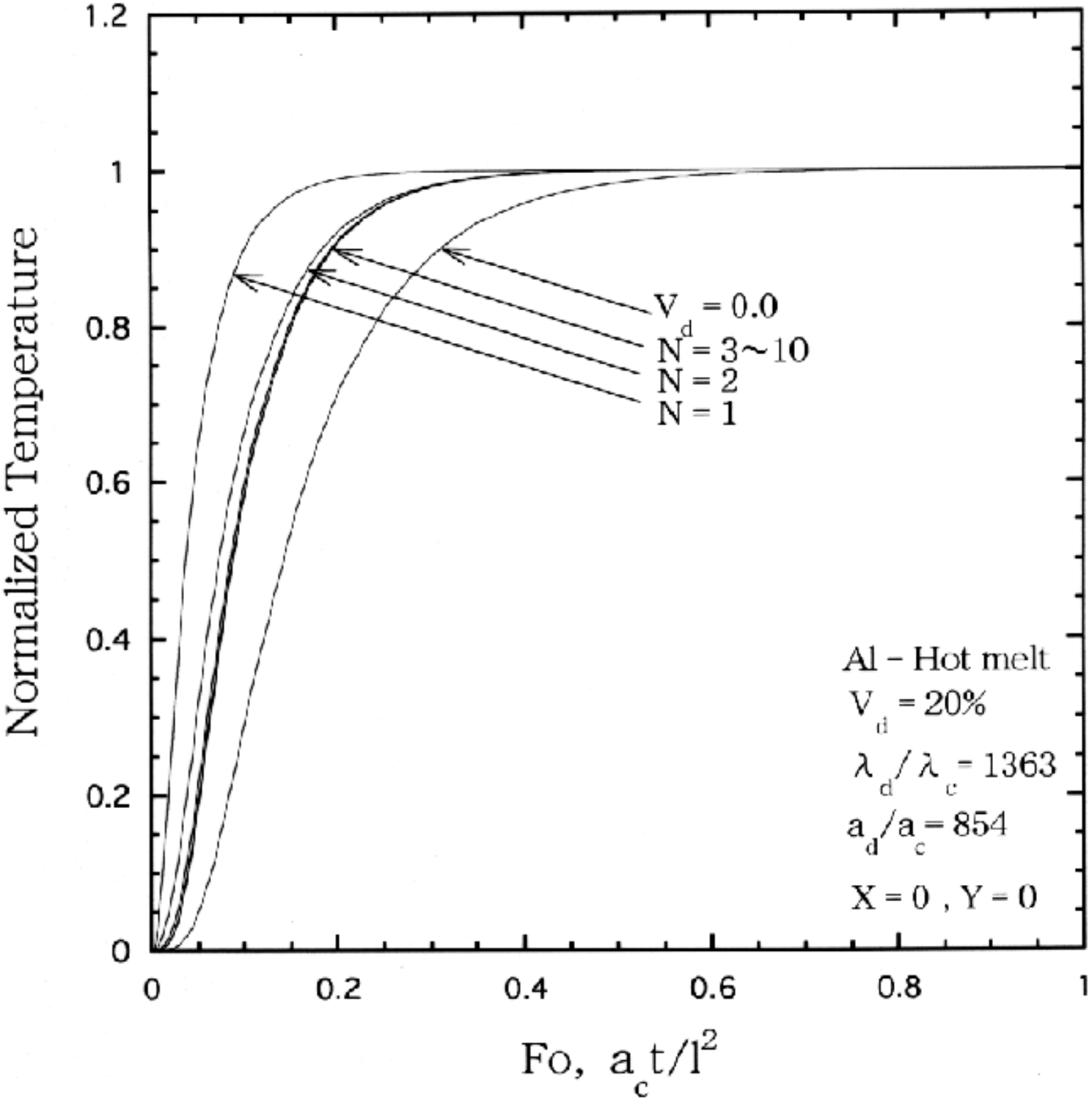
1.2
1
0.8
0.6
0.4
0.2
0
0
0.2
0.4
0.6
0.8
1
Normalized Temperature
Fo, $a_c t/l^2$
V_d = 0.0
N = 3~10
N = 2
N = 1
Al – Hot melt
V_d = 20%
λ_d/λ_c = 1363
a_d/a_c = 854
X = 0 , Y = 0

Figure 6

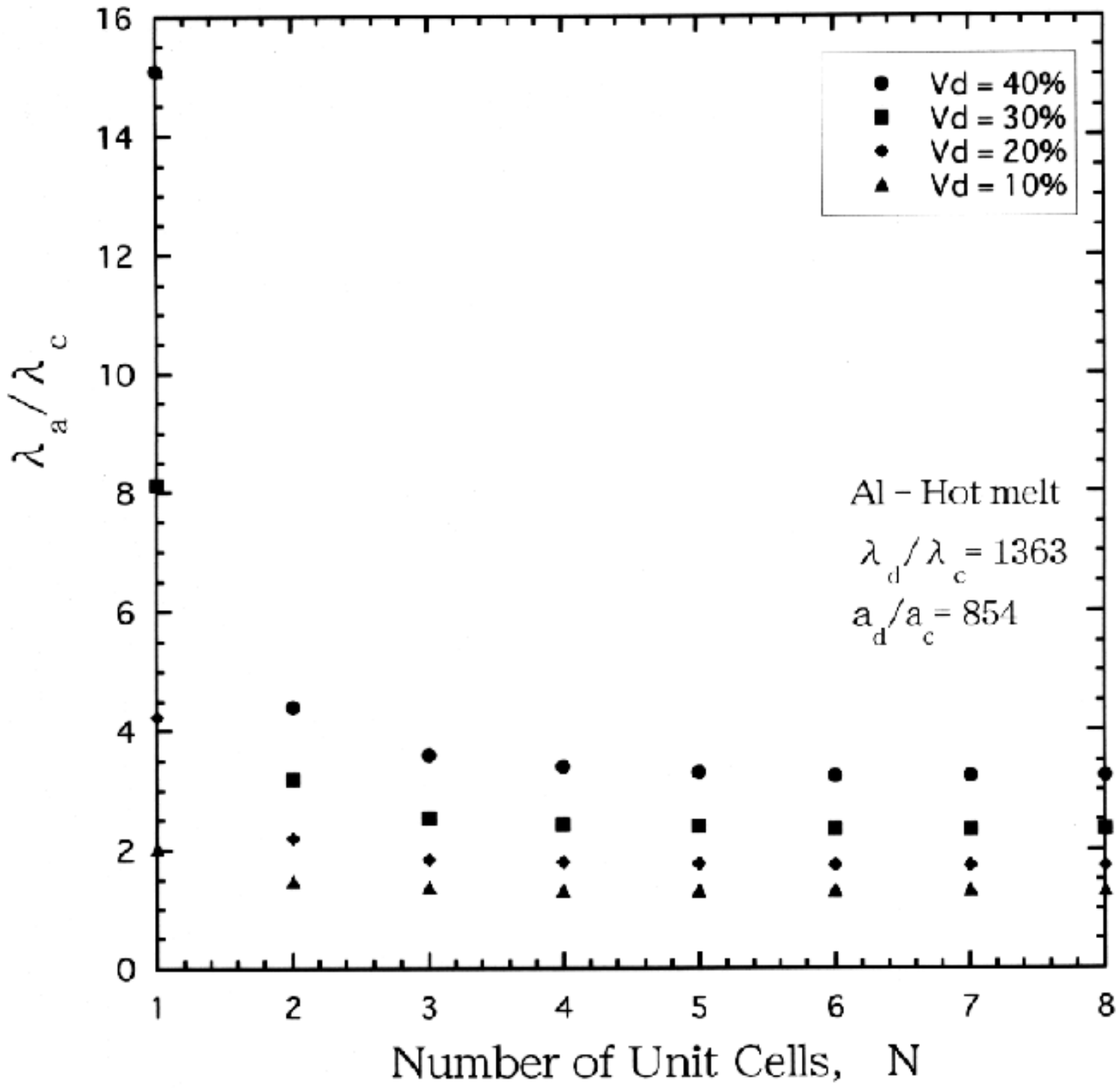
Vd = 40%
Vd = 30%
Vd = 20%
Vd = 10%
Al – Hot melt
λd / λc = 1363
ad / ac = 854
λa / λc
Number of Unit Cells, N
0
2
4
6
8
10
12
14
16
1
2
3
4
5
6
7
8

Figure 7

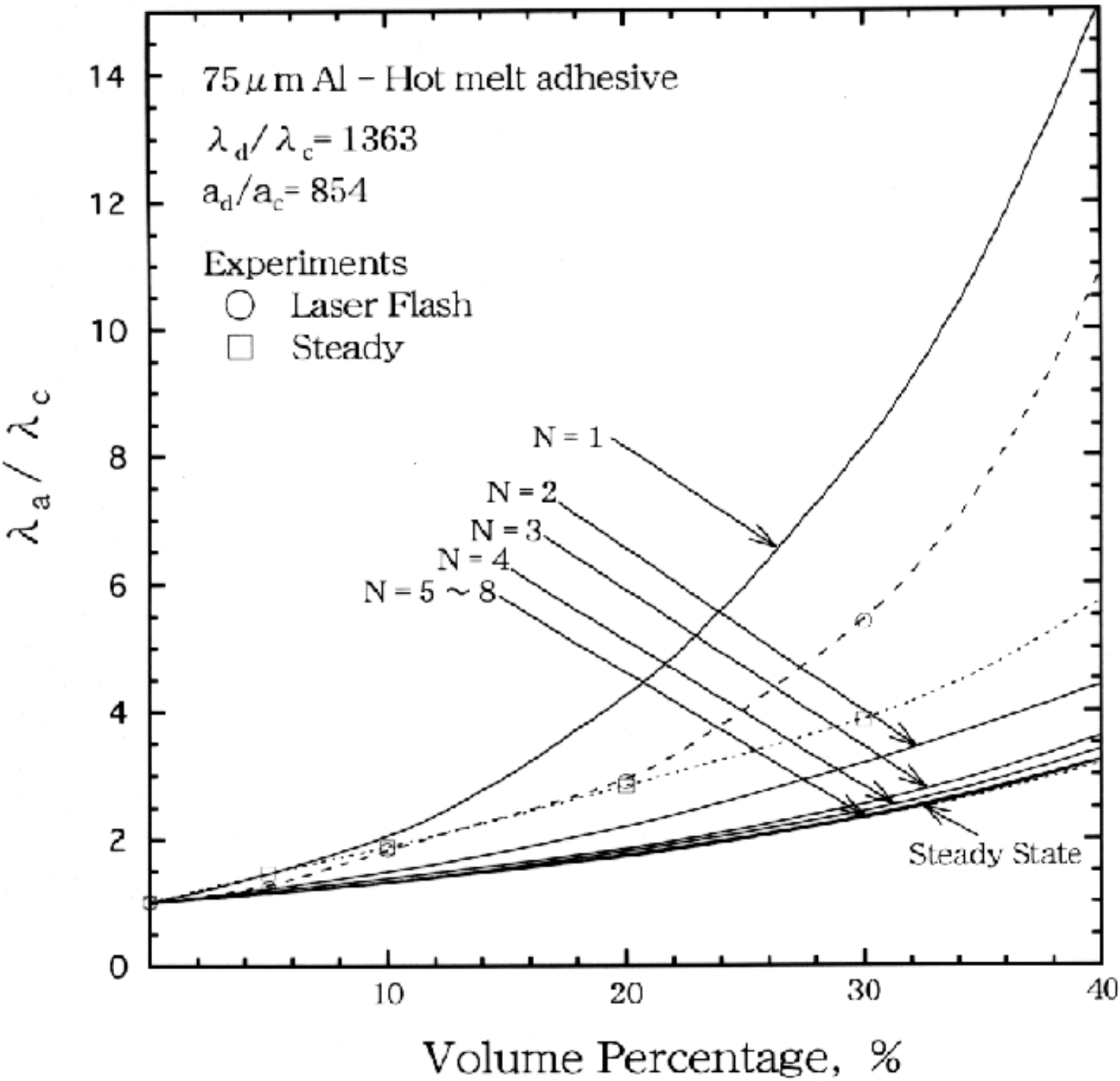
75 μm Al – Hot melt adhesive
λd/λc = 1363
ad/ac = 854
Experiments
Laser Flash
Steady
N = 1
N = 2
N = 3
N = 4
N = 5 ~ 8
Steady State
λa / λc
Volume Percentage, %
0
2
4
6
8
10
12
14
10
20
30
40

Figure 8