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Performance Analysis of Thermal Conductivity of Epoxy Matrix Composites Filled With Aluminium Powder

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ABSTRACT: The thermal conductivity of an AlN and an alumina-particulate-filled epoxy resin was investigated as a function of their volume content ratios. AlN composite exhibited thermal conductivity of 7.15W/mK at 68.5 vol% filler content but that of alumina composite was lower than 2.68W/mK even if the filler content was as much as 63.8 vol%. The thermal conductivity of the composites is strongly dependent on that of the filler. In the AlN filler properties, the particle size of the filler used affected the thermal conductivity of composites, and composites with higher thermal conductivity could be obtained by using filler with large particle size. The thermal conductivity of composites is governed by the number of resin layers on the surface of an AlN particle. The composite thermal conductivities decreased with increasing the oxygen content of the filler. The thermal conductivity of an AlN particle depends on the oxygen content of the particle itself and the decrease of the composite thermal conductivity with the increase of the oxygen content of the filler is due to the increase of the aluminum oxide layer thickness produced on the surface of particles. In the thermal conductivity model, the measured thermal conductivity of AlN-ground-powder-filled composites disagreed with the ones calculated using Bruggeman's expression. However, the measured values of alumina spherical-powder-filled composites agreed with the calculated ones. It is considered that the results depended on the sphericity of the used powder.

KEYWORDS: Aluminium nitride, Epoxy resin, AlN filler, Alumina filler, Thermal conductivity, Thermal conductivity model, Composite.

I. INTRODUCTION

The latest miniaturization of electronic devices and integrated circuits has caused an increase of power densities. The current trend in the field of microelectronic parts requires an epoxy resin with high thermal conductivity for encapsulating these heat-dissipating electronic components. [1-3] Epoxy resin has a low thermal conductivity, about 0.15 to 0.25W/mK, [4, 5] which can be increased by the addition of particulate inorganic filler materials with a high thermal conductivity. [6-8] Since the thermal conductivity of the inorganic filler is much larger than that of the epoxy resin, the addition of fillers is an effective way of raising the thermal property of the epoxy resin. Silicon oxide (silica), in the fused or crystalline state, and aluminium oxide (alumina) particles have been used as the resin. [9-12] Recently, the addition of silicon nitride (SiN) or aluminium nitride (AlN) particles with higher thermal conductivity has been attempted. [3-5] In this study, the thermal conductivities of the epoxy resins containing two kinds of particulate fillers were measured. Ground AlN and spherical alumina particles were employed as the filler. For AlN filler, in particular, the influence of the filling ratio and of the characteristics such as particle size and oxygen content on the composite thermal conductivities were studied.

Epoxy resins have been widely used in coatings, adhesives, highly-integrated memory chips, electronic packaging and light emitting diodes (LEDs) because of their chemical stability, adhesive properties, excellent

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resistance to several solvents, good mechanical properties and superior electrical resistivity [4–6]. However, epoxy resins have a poor thermal conductivity (0.15–0.25 W/mK [7,8]). In order to enhance the thermal conductivity, thermally conductive but electrically insulative materials should be introduced to form composite materials. Silica and alumina particles have long been used as fillers for epoxy resin composites in industry. However, the thermal conductivities of these oxides are one to two orders lower than that of some nitrides, i.e., aluminium nitride (AlN) and boron nitride (BN) [9–11]. Since AlN is reactive to moisture and water, but BN is chemically stable and inert to environment, BN has been considered as a potential filler candidate for the fabrication of high thermal conductivity epoxy resin composites for industrial applications.

II. EXPERIMENTAL PROCEDURES

To prepare test specimens, thermosetting epoxy resin powder modified with imido radical (Best Lex LS, Sumitomo Chemical Ind., Co., Ltd.) and particulate AlN material (D50=23.7m, R335 Toyo Aluminium K. K.) made from direct nitridation of aluminium metal were mixed in a glass container. Properties of the particulate AlN material used in this experiment are shown in Table 1, and a scanning electron microscope (SEM) image of the AlN material is shown in Fig. 1. The mixed materials were put into circular cylinders with 12mm diameter and 40mm height made of stainless steel and were pressed under 29MPa. The green bodies were cured in an oven at 180 for 30min. Specimens, with 10mm diameter and 2mm height, were obtained for the measurement of thermal conductivity. For comparison, the same procedure was carried out to obtain test specimens filled with the particulate alumina material. Spherical alumina particles (D50=10m alumina beads CB, Showa Denko Co., Ltd.) were used. A SEM image of the alumina particles is also indicated in Fig. 1. To investigate the effect of the particle size of AlN filler on the thermal conductivity of epoxy resin-AlN composites, various powders with different particle sizes were prepared by grinding a coarse AlN powder and adjusting the average particle size of ground powders by classification. The properties of the powders are also shown in Table 1 and their particle size distributions are shown in Fig. 2. Furthermore, in order to study the influence of oxygen content in AlN powder on the composite thermal conductivities, that of AlN powder with average particle size of 20.4m was adjusted by controlling heat treatment time at 450. Densities and thermal conductivities of specimens were measured by the water displacement and laser flash methods (TC-3000H-NC, Sinku-Riko, Inc.), respectively. Before measuring thermal conductivity, the surfaces of specimens were covered with carbon by spray coating.

Table 1 Properties and Oxygen Content of Al Powders

Average Particle Size (μm)	Specific Surface Area (m^2/g)	Oxygen Content (mass %)
5.7	1.29	0.76
10.3	0.47	0.45
15.6	0.35	0.43
19.0	0.25	0.36
23.7	0.18	0.30
26.1	0.14	0.30
27.6	0.13	0.29

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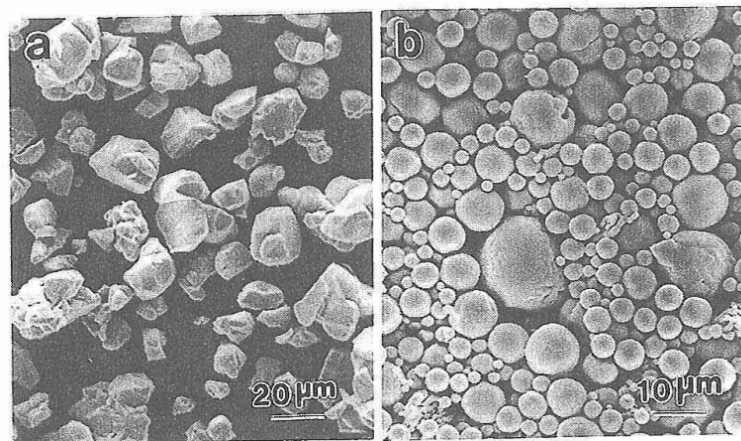


Fig 1 Microstructures of (a) Al and (b) Al₂O₃

III. MATERIALS AND METHODS

MATERIALS

Matrix Material:

Epoxy LY 556 resin, chemically belonging to the „epoxide“ family is used as the matrix material. Its common name is Bisphenol-A-Diglycidyl-Ether. The low temperature curing epoxy resin (Araldite LY 556) and the corresponding hardener (HY 951) are mixed in a ratio of 10:1 by weight as recommended. The epoxy resin and the hardener are supplied by Ciba Geigy India Ltd. Epoxy is chosen primarily because it happens to be the most commonly used polymer and because of its insulating nature (low value of thermal conductivity, about 0.363 W/m-K).

Filler Material (Aluminium Powder):

It is a silvery white metal. Aluminium is remarkable for the metal's low density and for its ability to resist corrosion. Its thermal conductivity and density values are 250 W/m.K and 2.7 g/cc. Structural components made from aluminium and its alloys are vital to the aerospace industry and are very important in other areas of transportation and building. Its reactive nature makes it useful as a catalyst or additive in chemical mixtures, including ammonium nitrate explosives, to enhance blast power

Composite Fabrication:

The low temperature curing epoxy resin (LY 556) and corresponding hardener (HY951) are mixed in a ratio of 10:1 by weight as recommended. Aluminium powder with average size 100-200 μm are reinforced in epoxy resin (density 1.1 gm/cc) to prepare the composites. The dough (epoxy filled with Al powder) is then slowly decanted into the glass tubes, coated beforehand with wax and uniform thin film of silicone-releasing agent. The composites are cast by conventional hand-lay-up technique in glass tubes so as to get cylindrical specimens (dia. 9 mm, length 120 mm). Composites of four different compositions (with 0.4, 1.4, 3.34 and 6.5 vol % of Al respectively) are made. The castings are left to cure at room temperature for about 24 hours after which the tubes are broken and samples are released. Specimens of suitable dimension are cut using a diamond cutter for further physical characterization and thermal conductivity test.

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Vol. 7, Issue 11, November 2018

Samples	Composition
1	Epoxy + 0 vol% (0 wt%) Filler
2	Epoxy + 0.4 vol% (1.01 wt%) Filler
3	Epoxy + 1.4vol% (3.4 wt%) Filler
4	Epoxy + 3.34 vol% (7.8wt%) Filler
5	Epoxy + 6.5 vol % (14.7wt%) Filler

Fig 2 List of particulate filled composites fabricated by hand-lay-up technique

THERMAL CONDUCTIVITY CHARACTERIZATION

Experimental Determination of Thermal Conductivity:

Unitherm™ Model 2022 is used to measure thermal conductivity of a variety of materials. These include polymers, ceramics, composites, glasses, rubbers, some metals and other materials of low to medium thermal conductivity. Only a relatively small test sample is required. Non-solids, such as pastes or liquids can be tested using special containers. Thin films can also be tested accurately using a multi-layer technique. The tests are in accordance with ASTM E-1530 Standard.

Operating principle of Unitherm™ 2022:

A sample of the material is held under a uniform compressive load between two polished surfaces, each controlled at a different temperature. The lower surface is part of a calibrated heat flow transducer. The heat flows from the upper surface, through the sample, to the lower surface, establishing an axial temperature gradient in the stack. After reaching thermal equilibrium, the temperature difference across the sample is measured along with the output from the heat flow transducer. These values and the sample thickness are then used to calculate the thermal conductivity. The temperature drop through the sample is measured with temperature sensors in the highly conductive metal surface layers on either side of the sample.



Fig 3 Determination of Thermal Conductivity Using Unitherm™ Model 2022

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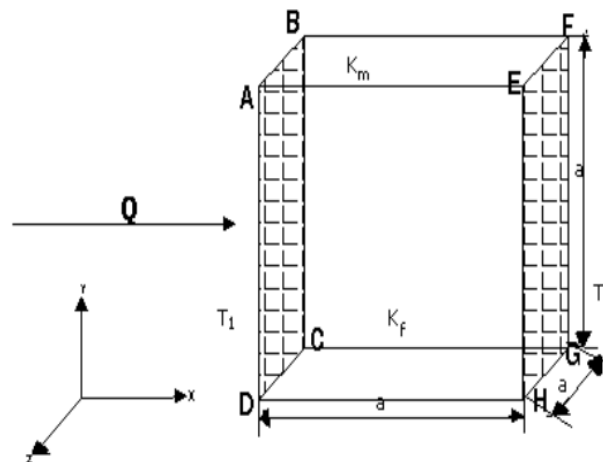
Vol. 7, Issue 11, November 2018

IV. RESULTS AND DISCUSSION

The determination of effective properties of composite materials is of paramount importance for functional design and application of composite materials. One of the important factors that influence the effective properties and can be controlled to an appreciable extent is the microstructure of the composite. Here, microstructure means the shape, size distribution, spatial distribution and orientation distribution of the reinforcing inclusion in the matrix. Although most composite possess inclusion of random distributions, great insight of the effect of microstructure on the effective properties can be gained from the investigation of composites with periodic structure. System with periodic structures can be more easily analyzed because of the high degree of symmetry embedded in the system. Using the finite-element program ANSYS, thermal analysis is carried out for the conductive heat transfer through the composite body. In order to make a thermal analysis, three-dimensional physical models with spheres-in-a-cube lattice array have been used to simulate the microstructure of composite materials for four different filler concentrations. Furthermore, the effective thermal conductivities of these epoxy composites filled with Aluminium particle up to about 6.5% by volume is numerically determined using ANSYS.

Numerical Analysis

In the numerical analysis of the heat conduction problem, the temperatures at the nodes along the surfaces ABCD is prescribed as T_1 ($=1000\text{C}$) and the convective heat transfer coefficient of ambient is assumed to be $2.5 \text{ W/m}^2\text{-K}$ at ambient temperature of 27°C . The heat flow direction and the boundary conditions are shown in Fig. 4.1. The other surfaces parallel to the direction of the heat flow are all assumed adiabatic. The temperatures at the nodes in the interior region and on the adiabatic boundaries are unknown. These temperatures are obtained with the help of finite-element program package ANSYS.



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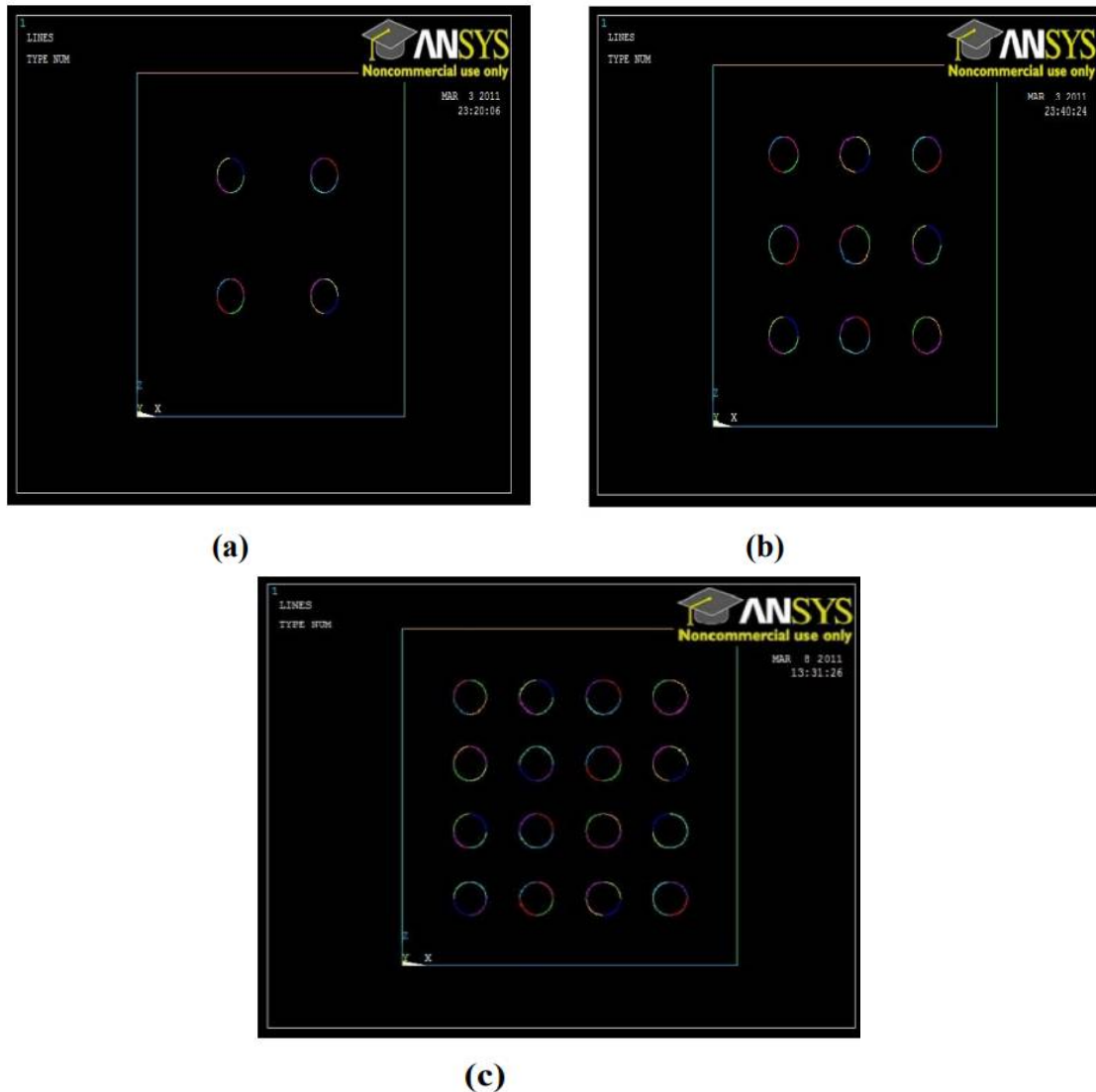


Fig 4 Typical 3-D spheres-in-cube models with particle concentration of Aluminium powder
(a) 0.4 vol% (b) 1.4 vol% and (c) 3.34 respectively.

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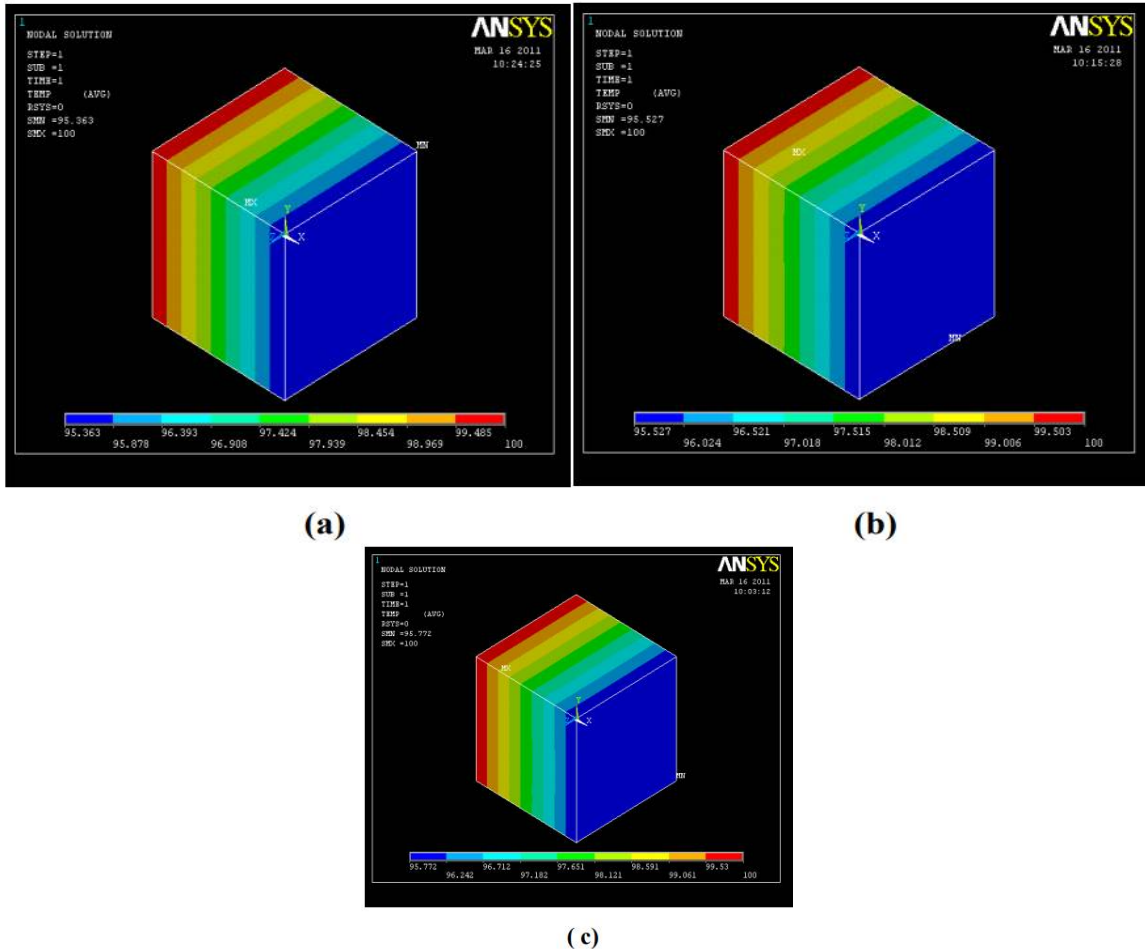


Fig 5 Temperature profiles for Aluminium filled epoxy composites with particle concentration of (a) 0.4 vol% (b) 1.4 vol% and (c) 3.34 vol% respectively

Sample	Al Content (vol %)	Effective thermal conductivity of composites K_{eff} (W/m-K)	
		FEM (Spheres-in-cube Model)	Experimental Value
1	0	-	0.363
2	0.4	0.369	0.364
3	1.4	0.382	0.369
4	3.34	0.408	0.385

Fig 6 Thermal conductivity for composites obtained from FEM and Experiment

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Vol. 7, Issue 11, November 2018

Composite Sample	Al Content (Vol. %)	Percentage errors associated with FEM results w.r.t. the experimental value (%)
1	0.4	1.3
2	1.4	3.5
3	3.34	5.9

Fig 7 Percentage errors with respect to the experimental value GRAPH:

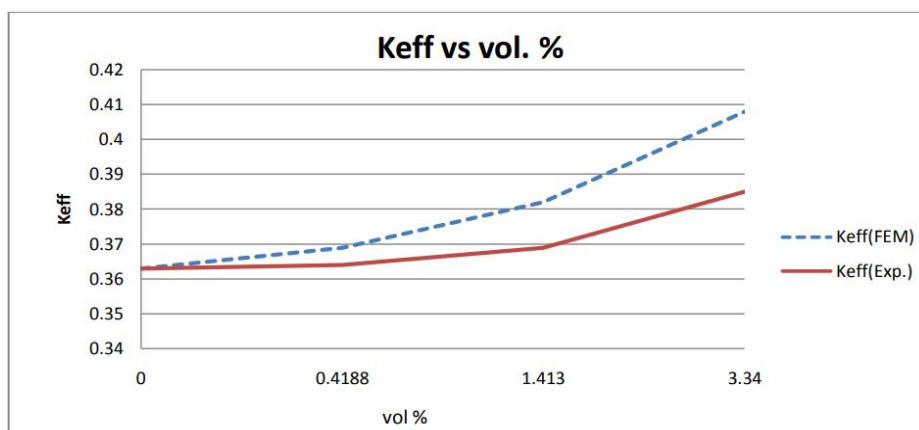


Fig 8 Comparison between Experimental results and FEM Analysis

Thermal conductivities of epoxy composites filled with aluminium particles to 6.5% by volume are numerically estimated by using the spheres-in-cube model and the numerical results are compared with the experimental results and also with some of the existing theoretical and empirical models. The temperature profiles obtained from FEM analysis for the composites with particulate concentrations of 0.4, 1.4, and 3.34 vol. %.

This study shows that finite element method can be gainfully employed to determine effective thermal conductivity of these composite with different amount of filler content. The value of equivalent thermal conductivity obtained for various composite models using FEM are in reasonable agreement with the experimental values for a wide range of filler contents from about 0.4 vol.% to 3.34 vol.%. Incorporation of Al results in enhancement of thermal conductivity of epoxy resin. With addition of 1.413 % and 3.34 vol. % of Al, the thermal conductivity improves by about 3.4 % and 6.3 % respectively with respect to neat epoxy resin.

VI.CONCLUSION

This numerical and experimental investigation on thermal conductivity of aluminium filled epoxy composites have led to the following specific conclusions: Successful fabrication of epoxy based composites filled with micro-sized Al by hand-lay-up technique is possible. Finite element method can be gainfully employed to determine effective thermal conductivity of these composite with different amount of filler content. The value of equivalent thermal conductivity obtained for various composite models using FEM are in reasonable agreement with the experimental values for a wide range of filler contents from about 0.4 vol.% to 3.34 vol.%. Incorporation of Al results in enhancement of thermal conductivity of epoxy resin. With addition of 3.34 vol. % of Al, the thermal conductivity improves by about 6.3 % with

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respect to neat epoxy resin. These new class of Al filled epoxy composites can be used for applications such as electronic packages, encapsulations, die (chip) attach, thermal grease, thermal interface material and electrical cable insulation.

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