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THERMOELECTRIC HEAT FLUXMETERS FOR THERMAL CONDUCTIVITY AND INTERFACE THERMAL RESISTANCE MEASUREMENTS

P.M. SCHLICKLIN
Centre de Recherches de
Pont-à-Mousson
B.P 28
54700 PONT-A-MOUSSON, FRANCE

J.G. STOCKHOLM
TNEE - Air Industrie
Thermoelectrics
250, Route de l'Empereur
92508 RUEIL MALMAISON Cedex, FRANCE

Abstract

Heat fluxmeters for the measurements of thermal conductivities and interface resistances are presented. They have as the active part one or more pieces of bismuth telluride that are used to measure heat fluxes larger than 1 kW/m². Two sizes of units are described and the method of calibration is indicated. The small unit is used for homogeneous samples at the scale of cross sectional areas ranging from 1 to 2.5 cm². The larger unit which may be used for samples of cross sectional area of the order of 30 cm² is designed to accomodate materials such as concrete that are not homogeneous at the scale of 2 cm².
The fluxmeters are designed to operate at room temperature and to cover the range of thermal conductivities from 0.1 to 100 W/(m.K). Results are given for stainless cast irons, polymeric materials and concretes.

Heat fluxmeters are essentially used for low fluxes for measurements on insulating materials[1]
The present work concerns also intermediate thermal conductivity values of the order of 1 W/(m.K).
The reason for choosing a heat fluxmeter that is thermoelectric is twofold.
First : thermoelectric materials such as bismuth telluride have a thermal conductivity around 1.5 W/(m.K).
Second : these materials having large Seebeck coefficients, one may relate the heat flux to a voltage measurement.

1. Objective

A simple heat fluxmeter based on a bismuth telluride sample of 1.5 cm² and 1.5 mm thickness is appropriate for heat fluxes in the range of kilowatts per m².
Our reference consists of a piece of bismuth telluride soldered to two copper discs with small holes drilled along their diameter to accept thermocouple junctions for voltage and temperature measurements.
Our standard was calibrated by the Laboratoire de Physico-Chimie de l'Etat Solide headed by Pr. J.P. ISSI of the University of Louvain-la-Neuve (Belgium).
It is preferable to purchase the bismuth telluride already soldered to nickel plated copper discs, otherwise it must be pretinned. The main manufacturers of thermoelectric material are :

- MELCOR 990 Spruce Street Trenton New Jersey 08648, USA
- MARLOW INDUSTRIES INC. 10451 Vista Park Road, Dallas Texas 75238, USA
- KOMATSU ELECTRONICS INC. 2597 Shinomiya, Miratsuka - Shi, Kanagawa - Ken, Japan.

Several substandards at the Centre de Recherches de Pont-à-Mousson are available for other laboratories, for comparison with existing fluxmeters or to reference newly made fluxmeters.
When larger heat flux surfaces are needed several of the above mentionned pieces may be soldered between two parallel copper plates.

Three units are described here.
The first consists of a single sample of thermoelectric material (1.5 cm² area round or square and 1.5 mm thick) intercalated between two copper discs of 18 mm diameter. The second consists of five pieces similar to the above mentionned one, soldered between two copper plates of 55 x 55 mm cross section and 5 mm thick and the third one is analogous to the second one but with nine sample pieces instead.

2. Single unit and calibration

2.1. Description

The configuration illustrated in Fig. 1 will be described in detail, since it is used in the two other configurations.

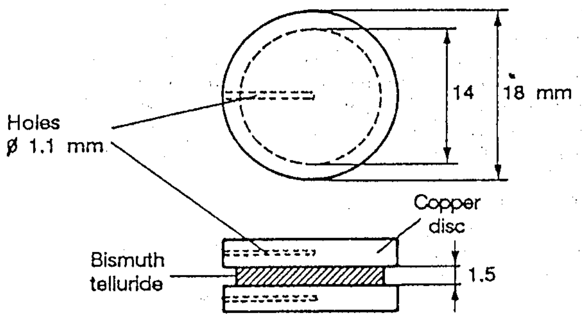

Fig.1 Single unit or substandard.

The thermoelectric material used between the two copper discs may be of N or P type and the inner surface of the copper disc must be coated with a nickel diffusion barrier (7 to 10 µm thick) to ensure stability in time.

The soldering of pretinned bismuth telluride material is performed with the bismuth-tin eutectic (58 % Bi, 42 % Sn, melting point 138°C). Care should be taken to avoid dissolving the nickel plating with the solder.
If one suspects that the nickel plating has been damaged, the completed set should be characterized (mV per ΔT), then the set should be put in an ordinary ambient air oven at 100°C for several weeks and the characteristics should be rechecked. If these characteristics are unchanged (within 3 %) then the part is considered to be stable in time.

2.2. Calibration

We have adopted a comparative method[2,3] that was developed for thermal conductivity measurements on bismuth telluride samples.

A schematic representation of the set-up is given below

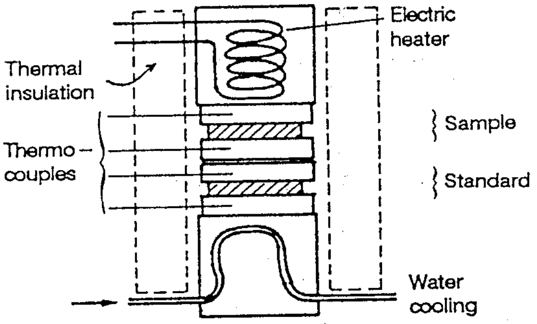

Fig. 2 Schematic representation of the principle of thermal conductivity measurement on the single unit shown in Fig. 1.

One of our standards is the part n° N 589 of N type with an area of 154 mm², thickness 1.5 mm and a thermal conductance of C = 0.161 W/K at 27°C (300 K).
This standard is used to calibrate numerous samples and substandards according to the following procedure.
A delicate problem in thermal conductivity measurements is the estimation of the heat losses, the geometry we are dealing with where the height (thickness) of the sample is small (1.5 mm). The heat flux through the specimens is large and the lateral heat losses are comparatively small. An estimation of heat losses for such a geometry[2,4], leads to a value between 2 and 5 % of the heat flux,giving the same range of accuracy by default for the measured value of thermal conductivity.
A standard and a sample are placed one above the other in the temperature differential unit. Heat sink compound is used at its interface in order to decrease the contact resistance.

The substandards are calibrated at 300 K, the temperature correction we use for these bismuth tellurides, was measured experimentally between - 10°C and + 60°C by J.P. ISSI, it is :

$$k_t = k_0 \ (1 - 0.0014 * t)$$

where t is the temperature in °C and k_0 the thermal conductivity at 0°C.

To eliminate the temperature correction, two sets of measurements must be made, one with the standard below the sample, and one with the standard above the sample.

The average values across the sample and across the standard are used to calculate the conductance C (W/K) :

$$C(Spl) = \frac{\Delta T(Std)}{\Delta T(Spl)} \ . \ C(Std)$$

The thermal conductivity of the sample is obtained from:

$$k(Spl) = \frac{e(Spl)}{e(Std)} . \frac{S(Std)}{S(Spl)} . \frac{\Delta T(Std)}{\Delta T(Spl)} . k(Std)$$

The samples and secondary standards, we call "susbstandards" can also be calibrated using the voltage generated by the Seebeck effect.

The relationship between the Seebeck voltage and the power (heat flux) is a linear relationship.
We can write : $\Delta V = S \ . \ \Delta T$
where S is the overall Seebeck coefficient
The standard gives the value for the thermal power (watts) :

$$P = C(Std) \ . \Delta T$$

so $$P = C \ . \ \Delta V/S$$

The characteristics of the standards include the contact resistances between the bismuth telluride and the copper discs and the thermal resistance of the copper discs. In fact the copper discs alone have a negligible influence but are necessary to facilitate the temperature and voltage measurements.
Applications are presented in paragraph 4.
A single piece substandard used for the measurements of thermal conductivity of glass has the following characteristics :
$P = 228 \ (\pm 21).\Delta V$ in W/V
$C = 0.165$ W/K.
The value of C is necessary to define the effective range of heat fluxes.

3. Fluxmeter with multiple elements

Thermal conductance measurements of heterogeneous materials require that the fluxmeter's area be several hundred times larger than the area of the homogeneous components. A fluxmeter size of 55 x 55 mm was chosen so that materials such as concrete with component areas of the order of 10 mm² can be measured.
To cover the range of heat fluxes, we built two units one with a thermal conductance of the order of 0.7 W/K with five 18 mm diameter substandards, the other with a thermal conductance of the order of 1.2 W/K with nine 18 mm substandards.
The substandards must all be of the same type N or P, for a given fluxmeter otherwise there is a cancellation of the Seebeck effects.

3.1 Description and construction of the fluxmeteers

In Fig. 3 we present a drawing of the two fluxmeters.

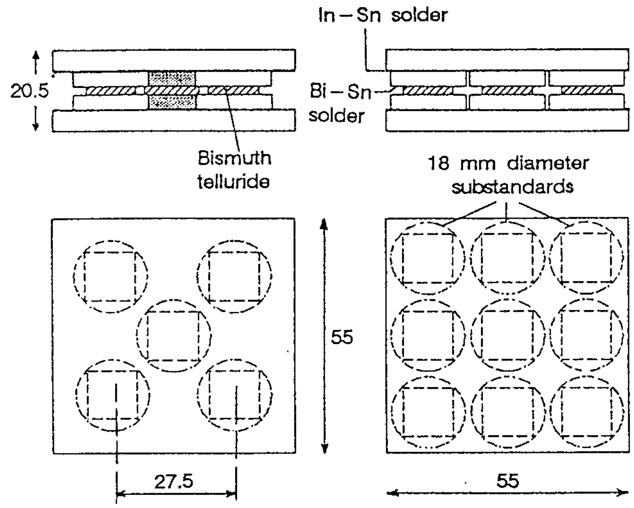

Fig. 3 - Drawing of the nine and five piece fluxmeters.

First the substandards are built and calibrated. The square copper plates of dimension 55 x 55 x 5 mm are made of electrolytic copper. In order to facilitate the mounting, the solder that will be used between the copper plates and the substandards must have a melting point lower than that of the solder used for manufacturing the substandards.
An Indium-Tin eutectic (52 % In, 48 % Sn, melting point 117°C) is used.

The procedures for soldering the copper discs to the copper plates are the following :

- pretinning with the above eutectic of both the external copper surfaces of the substandards and of one side of each copper plate

- a copper plate is placed on a hot plate with its pretinned surface on the upper side. The plate is heated till the solder melts, then the cold pretinned substandards are dipped in a liquid flux, giving no solid residue, and positionned on the copper plate. The precision of manual positionning can be enhanced using a template, the positions are shown in Fig.3. Once the substandards are all positionned, the heating is stopped to allow the system to cool down

- the structure is removed from the hot plate and the other plate with the pretinning on the upper side is now heated till the solder melts. The initial structure is turned over with the substandards facing downwards and placed on the second heated plate. It is positionned after slight back and forth movements to eliminate any air bubble and the sides are aligned up with those of the lower plate. The heating of the hot plate is stopped and the final structure is allowed to cool down.

3.2. Calibration

The overall thermal conductance consists of two parts, one relative to the plates and solder interfaces and one to the substandards.

The conductance of five or nine substandards is equal to the sum of the individual conductances of each substandard because they are thermally in parallel. The plates being thermally in series with the substandards the overall thermal resistance is the sum of the thermal resistances.

An approximate value of the thermal resistance of the plates is obtained by calculation using a thermal finite element computer program. We use "Titus" of Framatome - France.
There are two vertical planes of symmetry, so it is only necessary to calculate a quarter of each plate. The Fig 4 below shows the networks used for the calculations.

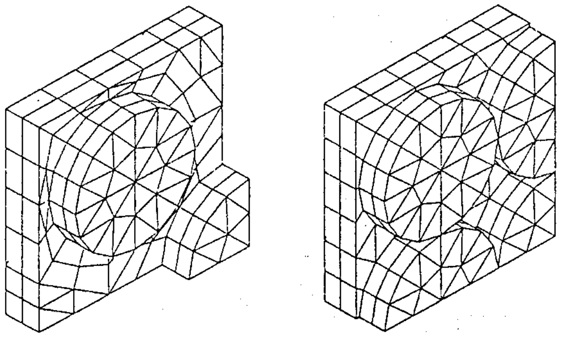

Fig 4 Schematic representation of the networks for thermal resistance calculations.

For these calculations, the temperatures on two of the areas are imposed : the outside flat area (55 x 55 mm) assumed to be at 10°C and the circular area, at the interface of the bismuth telluride assumed to be at 0°C.
The circular copper discs of the substandards are associated with the flat plates of 55 x 55 x 5 mm as can be seen in Fig 4.

The calculated thermal resistance of one plate is :
- with five discs R_t = 0.060 K/W
- with nine discs R_t = 0.040 K/W
In fact there is a solder (In-Sn) interface between each copper disc and the copper plane.

Individual measurements for the substandards used in the two fluxmeters are given in the following table where the values of Seebeck coefficient are also included.

Table 1 : Fluxmeters : characteristics of individual substandards

	5 pieces		9 pieces	
	S µV/K	C W/K	S µV/K	C W/K
Individual substandards	179	0.161	183	0.165
	179	0.165	185	0.169
	179	0.165	186	0.161
	179	0.160	181	0.169
	178	0.167	182	0.170
	-----	-----	185	0.167
			183	0.174
			186	0.166
			180	0.161
			---	------
av.S	178.4		183.4	
sum C		0.819		1.502

We have obtained the heat fluxmeter constant (W/V) by two methods.
The first method combines measurements and calculations. We obtain in this way for each of the two fluxmeters (5 and 9 piece) the overall thermal conductance. We also obtain an average Seebeck coefficient of all the substandards..
The second method consists in measuring directly on the fluxmeters their conductances and their Seebeck coefficients.
The overall thermal conductance is measured by putting in series a 18 mm diameter standard and the fluxmeter as shown in Fig.5

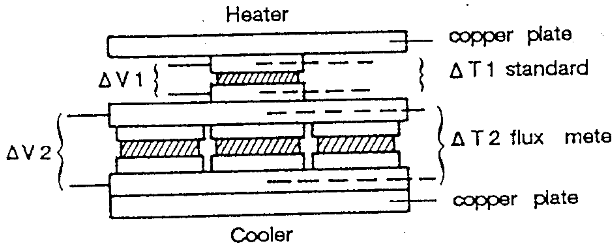

Fig. 5 - Fluxmeter direct calibration

At the bottom and at the top of the assembly are two copper plates of 55 x 55 x 5 mm.
These plates are necessary because, as we indicated in paragrah 2.2 that it is advisible to change the relative position of the fluxmeter and the standard. The change in cross section from the standard (18 mm diameter) to fluxmeter (55 x 55 mm) has no influence on the fluxmeter calibration because :
a) the lateral heat losses are negligible due to the black insulating material, which presents a thermal conductivity
k = 0.04 W/(m.K)
b) the temperature difference between the bottom of the standard and the top of the fluxmeter does not enter into the calibration.
The total measured conductance was obtained as shown in Fig. 5. The calculated and measured thermal conductances are given in Table 2.

Table 2 Fluxmeters conductance characterstics

	5 piece		9 piece	
	Calculated	Measured	Calculated	Measured
Substandard conductance W/K		0.819		1.502
Substandard resistance W/K		1.222		0.666
Plates resistance K/W	2 x 0.060		2 x 0.040	
Total resistance K/W (a)	1.342		0.746	
Total conductance C W/K	0.745	0.673 (b)	1.340	1.182 (b)

(a) The average measured values of the substandards are from Table 1
(b) Conductance measured with a known flux (W) from a calibrated standard.

The overall Seebeck measurements are made simultaneously with the conductance measurements.
In the coordinate system of ΔV = f(ΔT) the experimental points are remarkably aligned. The slope of the line (tangent of the angle with respect to the ΔT axis) is the Seebeck coefficient.
The values are shown in Table 3.

Table 3 Fluxmeters voltage calibration

		5 piece	9 piece
Calculated	Average substandard Seebeck coef. (µV/K)	178.4	183.4
	Calculated fluxmeter constant C/S (W/V)	4175	7305
Measured	Overall Seebeck (a) measurement (µV/K)	173.2	181.8
	Measured fluxmeter constant C/S (W/V)	3880	6500

(a) Least mean square linear correlation between ΔV and ΔT

The calculated average Seebeck is less than 3 % above the overall measured Seebeck.
In fact the overall Seebeck should be measured, especially as it is an easy measurement.
The precision on the thermal conductance (or resistance) is estimated to be ± 5 % .
The measured fluxmeter constants (C/S in W/V) are given in Table 3 using the overall measurements.
The object in having calculated the conductance and Seebeck values is to show that they are within 15 % of the measured values and can be useful for dimensioning various size fluxmeters.

4. Applications

Three fluxmeters have been presented, which can be characterized by their thermal conductances :

- 18 mm diameter C = 0.137 W/K
- 55 x 55 - 5 substandards C = 0.673 W/K
- 55 x 55 - 9 substandards C = 1.182 W/K

4.1 Single subunit

This is the easiest one to use. It is suitable for thermal conductances and interface conductance (or resistance) measurements on samples which may be considered as homogeneous on the scale of 18 mm diameter area.

One example of thermal conductivity example is that of austenitic cast iron with the following composition in weight (%) :
Ni = 30, Cr = 3, Nb = 1, C = 2,2
Sample dimensions : 18 mm diameter
20 mm length.
The stack is presented in the Fig. 6.

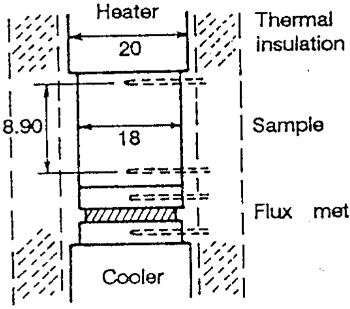

Fig 6 Thermal conductivity measurement of cast iron.

The average temperature of the sample is 28°C. The temperature drop across the substandard (C = 0.137 W/K) is 11.6 K and across the sample 10.5 K, the thermal conductance of the sample is therefore C = 0.149 W/K and its thermal conductivity k = 11.3 W/(m.K)

Another interesting example is the measurement of the interface thermal resistances between two materials with high thermal conductivities (above 10 W/(m.K)). It is relatively easy to make the measurement with the following stack

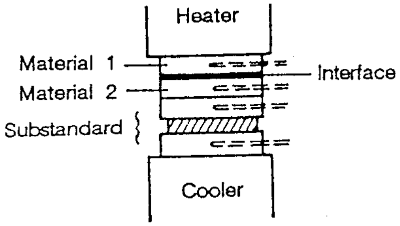

Fig 7 Interface thermal resistance measurement

A fundamental parameter in all interface thermal resistance measurements is the pressure. The unit containing the stack should therefore contain a calibrated tightening device. A simple solution is a calibrated spring.

When material 1 and material 2 are both aluminium alloys (ISO Al Mg Si - ASTM 6063) with a surface roughness (polished) $R_a = 3.10^{-8}$ m, for two dry surfaces the thermal resistance as a function of the pressure is given in Fig 8.

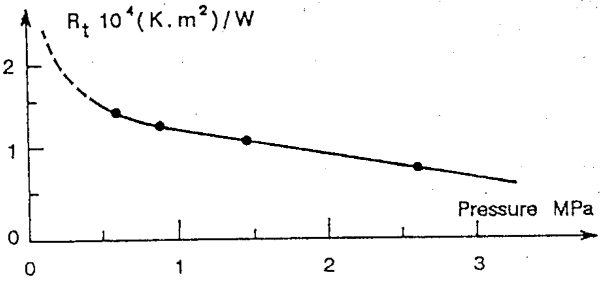

Fig 8 Interface thermal resistance versus pressure for a dry Al - Al - interface.

4.2. Fluxmeter with 5 or 9 substandards

4.2.1. Thermal conductivity of a polymeric material. We have measured the thermal conductivity of a PVC sample (poly-vinyl-chloride). The interface thermal resistance is determined by the measurements we made on several samples with the following thicknesses : e = 1, 5, 10, 20 mm. The plates on either side of the samples are made of electrolytic copper of 55 x 55 x 5 mm. One or more thermocouples are placed in each plate.

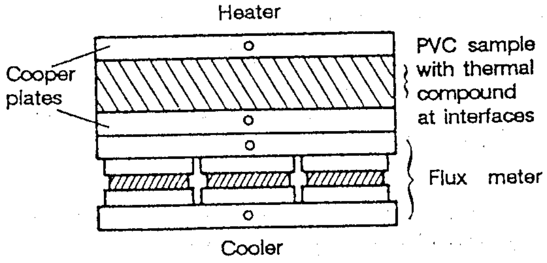

Fig 9 Thermal conductivity measurement of PVC.

The results of thermal conductivity data as a function of thickness for a pressure of 0.5 MPa are given below

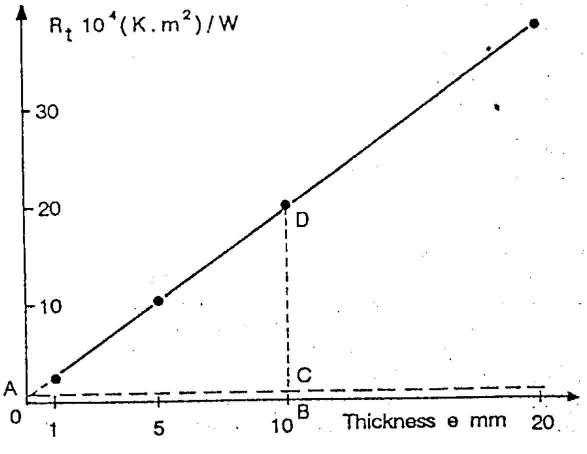

Fig 10 Thermal resistance of PVC samples

The intersection of the line with the Y ordinate,at O thickness:OA gives the thermal resistance of both interfaces. The thermal resistance being a linear function of the sample thickness, we can obtain the thermal conductivity of the sample by substracting the value of OA (=BC) from the measured value BD. The thermal resistance of a 10 mm thick material is then R_t = CD.

The thermal conductivity is equal to :

$$k = e/(R_t . A)$$

We find k = 0,172 W/(K.m). The thermal resistance on each side (OA divided by 2) is found to be : R_i = 0,30 K/W for the area of A = 30.2 cm² therefore the interface thermal resistance per unit area is :
$r_i = 9.10^{-4}$ (K.m²)/W

4.2.2. Thermal conductivity of a concrete

The problem with concrete is that it is not a homogeneous material. Some of the aggregates may reach dimensions as large as 5 to 10 mm. The sample has the following dimensions :

55 x 55 x 10.5 mm

The measured sample is a glass-cement composite (6 % weight of glass) cured 28 days at 20°C, 50 % rh.
The stack is similar to the one shown on Fig 9.
We decided to insert the thermocouples in the copper plates because it is easier to drill holes in the copper plates than in the concrete samples.
A thermal compound (Dow Corning G 340 : silicone grease filled with zinc oxide) is inserted at the interfaces. The roughness of the concrete is measured with a sliding probe. The heights of the asperities are found to be 0.02 mm. The hollows were filled with the thermal compound and the copper plates are pressed with a circular motion to ensure a direct contact of the copper surface with the most prominent asperities.
Knowing the area of the interface (A = 30.2 cm²) and its average thickness (e = 0.01 mm) as well as the thermal conductivity of the grease (K = 0.15 W/(m.K) as given by manufacturer), we may compute :
Thermal resistance for each grease interface : R_t = 0.022 K/W
The overall measurement with the 2 interfaces is :
R_t = 4.41 K/W
The two interface resistances are substracted :
$R_t = 4.41 - 2 \times 0.022 = 4.37$ K/W

The thermal conductivity is therefore :
k = 0.79 W/(m.K)
It is advisible to measure the thermal conductances of several thicknesses and with the same surface roughness, so that one can obtain by extrapolation like in paragraph 4.2.1 the interface resistances. Samples of different thicknesses were not available for such measurements.

5. Conclusions

Three fluxmeters are presented with their characteristics.

Table 4

Nber of substandard	1	5	9
Thermal conductance W/K ± 5 %	0.137	0.673	1.182
Fluxmeters constant W/V ± 5 %	727	3880	6500

They are used for thermal conductivity measurement and interface thermal resistance measurements.

These fluxmeters are well suited for mesuring any thermal conductivity in the range from 0.1 to 100 W/(m.K). This covers the range of most polymeric materials, concretes, ceramics and most metals.
The advantage of these fluxmeters is that they are all derived from a basic substandard, unit consisting of a sample of bismuth telluride intercalated between two copper discs of 18 mm diameter.

The thermoelectric elements may be obtained from the manufacturers of thermoelectric materials and a few calibrated susbstandards are available from the authors.

Acknowlegdment

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