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EFFECTIVE THERMAL CONDUCTIVITY OF ALUMINUM OXIDE WITH METALLIC FILLERS IN GASEOUS MEDIA AND A VACUUM AT VARIOUS TEMPERATURES

M. M. Safarov and Kh. Madzhidov

UDC 536.21

Experimental data on λ of granular porous aluminum oxide as a function of copper concentration and temperature in various gaseous media at atmospheric pressure and in a vacuum ($P = 8 \cdot 10^{-3}$ mm Hg) are presented.

Granular porous aluminum oxide is used in the production of ceramics, refractories, forms, catalysts, etc. Despite its wide use, the literature offers practically no data on its thermophysical properties. At present only the thermophysical properties of monolithic aluminum oxide have been studied sufficiently thoroughly [1].

The effective thermal conductivity coefficient of the aluminum oxide most widely used in high-temperature catalytic processes was studied (specific surface, 123 m^2/g; total pore volume, 0.35 cm^3/g; bulk density, 1 g/cm^3; cylindrical granule dimensions, 0.8-1.25 mm). The copper-containing specimens were prepared by steeping the aluminum oxide in a solution of copper in nitric acid with subsequent thermal processing in air and hydrogen at 673°K.

Effective thermal conductivity was measured by a regular thermal regime cylindrical bicalorimeter [2]. The experimental arrangement consisted of the cylindrical bicalorimeter, temperature stabilization system, vacuum system, and filling system. The calorimeter consisted of two coaxially arranged copper cylinders: internal (diameter 15.95 mm) and external (diameter of inner and outer sections 28.28 mm and 90.0 mm). The free space between the cylinders was filled with the specimen under study. The specimen thickness was 6.165 mm, and the temperature head across its boundaries with 1.78-0.90°K. During experiments the bicalorimeter temperature was maintained constant to an accuracy of 0.005-0.02°K. Relative measurement uncertainty at a confidence level of $\alpha = 0.95$ was 3.2%.

The specimen effective thermal conductivity was determined for the freely poured state. According to Table 1, the effective thermal conductivity increases with increase in copper content.

T. G. Shevchenko State Pedagogical Institute, Dushanbe. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 50, No. 3, pp. 465-471, March, 1986. Original article submitted December 5, 1984.

TABLE 1. Effective Thermal Conductivity of Aluminum Oxide in Air vs Copper Concentration at 293°K

Copper concn., %	0	4,5	12,0	15,5	18,6	23,4	28,8
$\lambda\cdot10^3$, W/(m·K)	168	176	181	202	215	234	247

TABLE 2. Basic Characteristics of Aluminum Oxide Containing Various Quantities of Copper

Copper concn., %	Specific surface, m²/g	Total pore volume, cm³/g	Bulk density g/cm³
4,5	119	0,310	1,073
12,0	112	0,272	1,121
15,5	105	0,260	1,148
18,6	100	0,246	1,186
23,4	99	0,218	1,277
28,8	84	0,196	1,344

During the steeping process the metal is deposited on the granule and introduced into its pores in the form of fine crystalline formations, separated from each other by the material of the granule, which has poor thermal conductivity. Naturally such metallic impregnations cannot produce a marked contribution to effective thermal conductivity. However, as experiment shows, they nevertheless lead to a slight increase in effective thermal conductivity with increase in metal content. This can partially be explained by the fact that upon metallization of the granules a reduction in pore volume occurs. In fact, according to Table 2, with increase in copper content on the aluminum oxide surface a reduction in granule pore volume occurs.

A decrease in granule porosity leads to an increase in effective thermal conductivity of the charge (Fig. 1), which changes with increase in copper content in all gaseous media and a vacuum by a linear law (see Fig. 4).

In fact, according to [3], with decrease in porosity the effective thermal conductivity of powder charges increases.

As is evident from Fig. 2, with increase in temperature the effective thermal conductivity coefficient in the various media and in vacuum increases linearly.

The studies show that with increase in thermal conductivity of the gaseous filler the effective thermal conductivity of the specimens increases. The highest thermal conductivity is found in the hydrogen and helium media with significantly lower values in argon and nitrogen (Table 3).

The smallest increase in thermal conductivity with temperature was found in a vacuum. With increase in thermal conductivity of the filler-gas, the increase in specimen conductivity with increase in temperature becomes larger. For example, for heating by 1°C the thermal conductivity of the specimen containing 4.5% copper increases in the following manner in the gases and vacuum: in vacuum, 0.057; argon, 0.157; nitrogen, 0.197; helium, 0.295; hydrogen, $0.363\cdot10^{-3}$ W/(m•K).

According to [4], the thermal conductivity coefficients of helium and hydrogen increase somewhat more rapidly with temperature than those of argon and nitrogen. Therefore, the effective thermal conductivity of the specimens in helium and hydrogen media increase much more rapidly than in nitrogen and argon.

The studies show that the effective thermal conductivity of the specimens depends significantly on the filling of their internal pore volume by gases and is determined to a lesser extent by the presence of gas filling the space between the specimen granules. For example, after creation of a deep vacuum the bicalorimeter was filled with argon, then for a specimen containing 4.5% copper at 293°K an effective thermal conductivity of 0.167 W/(m•K) was obtained. After removing the gas from the bicalorimeter (by shallow vacuuming so that argon

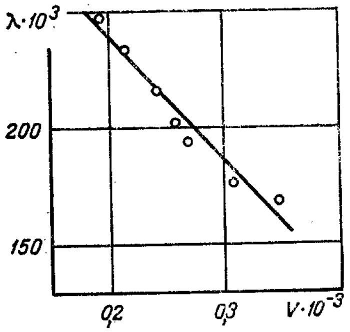

Fig. 1

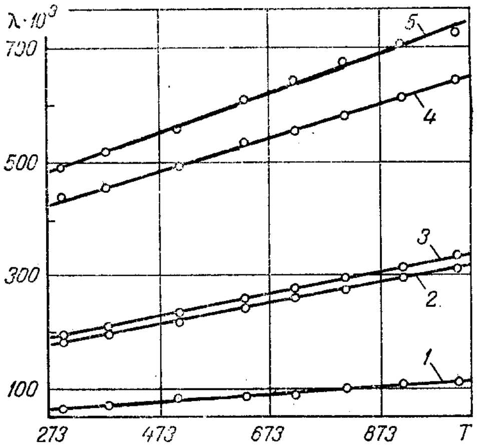

Fig. 2

Fig. 1. Effective thermal conductivity of aluminum oxide containing various quantities of copper at 293°K vs pore volume. λ, W/m•K; V, m^3/kg.

Fig. 2. Effective thermal conductivity of aluminum oxide containing 12% copper vs temperature in various gaseous media and in vacuum: 1) vacuum; 2) argon; 3) nitrogen; 4) helium; 5) hydrogen. T, °K.

remains within the pores) we filled the chamber with another gas, for example, helium. Measurements then produced results close to the effective thermal conductivity with the bicalorimeter filled with argon. This indicates the small effect of the gas filling the bicalorimeter and the dominant influence of the gas within the pores on effective thermal conductivity.

It should be noted that the effective thermal conductivity of the specimens in vacuum is insignificant in comparison to their conductivity in a gaseous medium. This indicates that in the gaseous medium heat transfer is accomplished mainly through the gas filling the specimen pores and the free space between granules, and to a significantly smaller degree by contacts between the granules of solid phase.

Thus, it may be concluded that the absence of gas within the pores significantly increases the thermal resistance of a layer of the specimen material and thus reduces the effective thermal conductivity. The increase in effective thermal conductivity of charges of the materials studied in a vacuum with increase in temperature can be explained by an increase in the area of the contact spots between granules and an increase in the fraction of radiant heat exchange.

To generalize the experimental data on effective thermal conductivity coefficients of the specimens studied, the law of corresponding states was used in the form

$$\frac{\lambda}{\lambda_1} = f\left(\frac{T}{T_1}\right), \tag{1}$$

where λ and λ_1 are the effective thermal conductivity coefficients at temperatures T and T_1; T_1 = 673°K.

Tests of Eq. (1) for the specimens studied herein revealed that it describes the effective thermal conductivity coefficients of these materials qualitatively and quantitatively.

The satisfaction of Eq. (1) by the specimens studied is shown in Fig. 3. As is evident, all the experimental points fit well on a common straight line which is described by the equation

$$\lambda = \left(0{,}40\frac{T}{T_1} + 0{,}59\right)\lambda_1. \tag{2}$$

Many points coincided in the figure, so experimental data for some specimens in various media are not shown.

TABLE 3. Experimental Values of Effective Thermal Conductivity of Aluminum Oxide vs Temperature and Copper Concentration in Various Gaseous Media and Vacuum $\lambda \cdot 10^3$, W/(m•K)

T, K	Gaseous medium				
	vacuum	argon	nitrogen	helium	hydrogen
Al_2O_3					
293,8	47,0	153	171	358	400
363,7	50,4	161	182	377	420
489,7	56,0	179	200	415	468
595,0	61,0	191	215	440	500
698,2	66,0	204	230	472	539
821,1	72,0	217	240	504	580
915,8	77,0	232	260	532	608
1010,8	83,0	243	276	559	649
Al_2O_3+4,5%Cu					
300,1	53	167	178	390	464
384,2	57	181	195	425	490
509,9	66	202	220	453	536
607,2	71	217	238	480	570
709,7	77	231	260	510	610
846,1	84	253	286	550	660
935,1	87	267	306	577	693
1019,3	94	280	320	602	725
Al_2O_3+15,5%Cu					
300,5	69	187	204	455	520
384,6	75	203	220	479	553
515,7	83	227	242	514	598
631,4	91	250	269	552	640
733,3	98	275	283	579	672
838,5	104	295	299	610	716
918,5	109	307	319	636	736
995,6	116	322	338	655	764
Al_2O_3+18,6%Cu					
297,8	74	190	221	468	549
388,8	81	210	238	494	583
570,7	93	248	272	546	645
671,2	99	269	290	575	682
781,3	107	293	310	610	720
902,0	115	317	332	654	760
1011,7	123	338	353	673	789
Al_2O_3+23,4%Cu					
295,7	78	199	240	490	574
396,6	85	222	258	508	610
522,4	94	249	283	553	650
658,5	103	278	309	586	694
805,3	114	310	335	628	742
899,6	120	331	354	653	772
1016,3	128	356	375	685	809
Al_2O_3+28,8%Cu					
290,4	85	214	252	522	595
364,9	92	229	268	543	623
491,2	102	257	295	574	667
598,8	110	280	317	598	700
705,5	119	304	339	624	735
819,2	128	329	362	653	774
939,5	137	355	388	683	813
1013,4	144	372	400	702	838

Equation (2) describes the temperature dependence of effective thermal conductivity coefficient of the specimens studied over the range 293-1073°K, generally to an accuracy of 2-5%. For individual points, the error of Eq. (2) reaches 8%.

With the aid of Eq. (2) one can calculate the thermal conductivity of the specimens studied as a function of temperature, if the value of λ_1 is known.

It is interesting to attempt to relate λ_1 in Eq. (2) with the percentage content of copper in the aluminum oxide (Fig. 4). As is evident from the figure, with increase in percentage content of copper λ_1 in a vacuum and gaseous media increases linearly. These straight

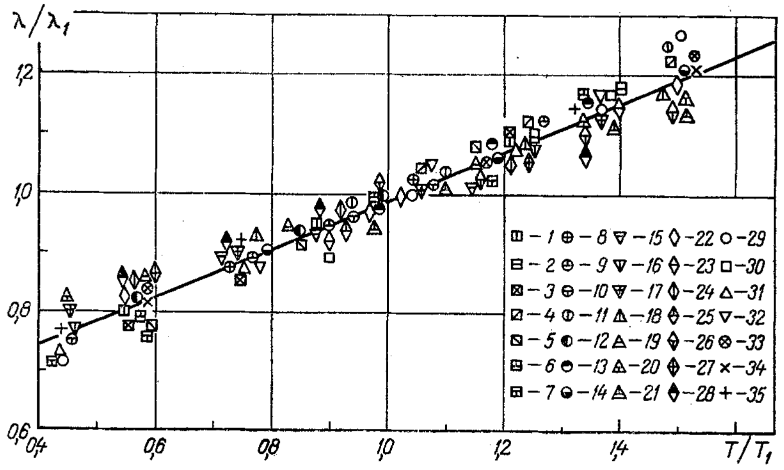

Fig. 3. Effective relative thermal conductivity vs relative temperature of pure aluminum oxide and oxide with various quantitites of copper in various media: argon (1, Al_2O_3; 2, 4.5%; 3, 12; 4, 15.5; 5, 18.6; 6, 23.4; 7, 28.8%); nitrogen (8, Al_2O_3; 9, 4.5%; 10, 12; 11, 15.5; 12, 18.6; 13, 23.4; 14, 28.8%); helium (15, Al_2O_3; 16, 4.5%; 17, 12; 18, 15.5; 19, 18.6; 20, 23.4; 21, 28.8%); hydrogen (22, Al_2O_3; 23, 4.5%; 24, 12; 25, 15.5; 26, 18.6; 27, 23.4; 28, 28.8%); vacuum (29, Al_2O_3; 30, 4.5%; 31, 12; 32, 15.5; 33, 18.6; 34, 23.4; 35, 28.8%).

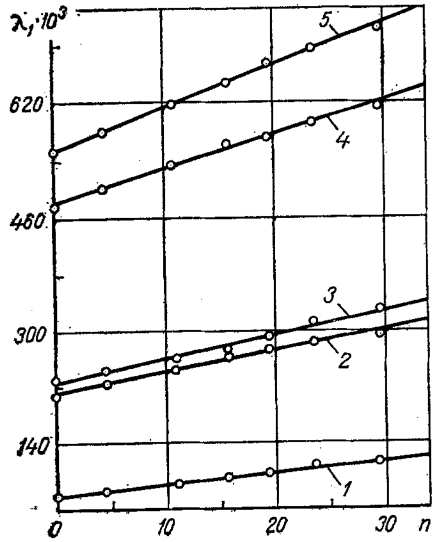

Fig. 4. Dependence of λ_1 on percentage of copper content: 1) vacuum; 2) argon; 3) nitrogen; 4) helium; 5) hydrogen; n, %.

TABLE 4. Values of Coefficients A and B in Eq. (3)

Medium	Vacuum	Argon	Nitrogen	Helium	Hydrogen
$A\cdot 10^3$, W/(m·K)	1,376	2,963	3,069	4,709	6,243
$B\cdot 10^3$, W/(m·K)	72,809	213,67	239,19	481,8	551,9

lines can be described by the equation

$$\lambda_1 = (An + B)\cdot 10^{-3},\quad \mathrm{W/(m\cdot K)}, \tag{3}$$

where n is the percentage content of copper and values of A and B in various media are presented in Table 4.

From Eqs. (2) and (3) we obtain

$$\lambda = \left(0{,}40\frac{T}{T_1} + 0{,}59\right)(An + B)\cdot 10^{-3}. \qquad (4)$$

With the aid of Eq. (4) one can calculate the effective thermal conductivity coefficient of aluminum oxide with various copper contents as a function of temperature without experimental study. For such a calculation only a knowledge of the copper content is required.

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IDENTIFICATION OF A SPECIFIED THERMAL REGIME IN A STRUCTURE ON THE BASIS OF EXPERIMENTAL DATA OBTAINED IN OTHER REGIMES

I. E. Balashova

UDC 536.24

A method is proposed for calculating the temperature at a given point of a complex structure with a specified heating regime on the basis of experimental data obtained in other regimes.

During the development and testing of new equipment and the modernization of existing equipment operating at elevated temperatures, it is necessary to determine the temperature at individual points inside the structure in given thermal regimes. Difficulties in allowing for all features of heat transfer make it difficult to solve this problem by direct numerical methods through solution of the heat-conduction equation for complicated, multilayered structures. The quality of the results obtained is significantly affected by the shortage — and in some cases, complete lack — of information on the laws of distribution of contact heat-transfer resistance between the layers and heat transfer in air gaps.

In connection with this, it is very important to determine the temperature inside an object on the basis of temperature data obtained during experiments in other heating regimes. Searches for a solution to this problem have led to the idea of replacing the actual complex structure by a simpler mathematical model with fewer layers characterized by a certain effective heat-transfer coefficient [1, 2].

The method employed in [1] is based on the use of a so-called "reference" regime and conversion factor in the calculation of prescribed surface temperature regimes. The conversion factor is calculated from known empirical temperature data at a given point of the structure by analytical solution of a unidimensional heat-conduction equation for a one-layer wall. This method gives good results in several cases. However, it has certain limitations in terms of its application, indlucing the fact that it is possible to calculate only monotonic regimes the length τ^e of which does not exceed the length of the reference regime.

Another study [2] proposed that the computational model be the heat-conduction equation for fewer number of layers of the same geometry, with a certain constant effective thermal

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Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 50, No. 3, pp. 471-476, March, 1986. Original article submitted January 14, 1985.