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DEVELOPMENT OF A STEADY STATE CREEP BEHAVIOR MODEL OF POLYCRYSTALLINE TUNGSTEN FOR BIMODAL SPACE REACTOR APPLICATION

by

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EXECUTIVE SUMMARY

The fuel element for one of the many reactor concepts being currently evaluated for bimodal applications in space consists of spherical fuel particles clad with tungsten or alloys of tungsten. The fuel itself consists of stabilized UO_2.

One of the life limiting phenomena for the fuel element is failure of the cladding because of creep deformation. This report summarizes the information available in literature regarding the creep deformation of tungsten and its alloys and proposes a relation to be used for calculating the creep strains for elevated temperatures in the low stress region ($\sigma \leq 20$ MPa). Also, results of the application of this creep relation to one of the reactor design concepts (NEBA-3) are discussed.

Based on the traditional definition of creep deformation, the temperatures of 1500 K to 2900 K for tungsten and its alloys are considered to be in the "high" temperature range. In this temperature range, the rate controlling mechanisms for creep deformation are believed to be non-conservative motion of screw dislocations and short circuit diffusional paths. Extensive theoretical work on creep and in particular for creep of tungsten and its alloys have been reported in the literature. These theoretical efforts have produced complex mathematical models that require detailed materials properties. These relations, however, are not presently suitable for the creep analysis because of lack of consistent material properties required for their use. Variations in material chemistry and thermomechanical pre-treatment of tungsten have significant effects on creep and the mechanical properties.

Analysis of the theoretical models and limited data indicates that the following empirical relation originally proposed by M. Jacox of INEL and the Air Force Phillips Laboratory, for calculating creep deformation of tungsten cladding, can be used for the downselection of preliminary bimodal reactor design concepts.

$$\dot{\epsilon} = 1.3 \times 10^3 \left[e^{\frac{-53370}{T}} \right] \times \sigma^{3.53}$$

(1)

where

$\dot{\epsilon}$ = steady state creep rate, h^{-1}

σ = creep stress, MPa, $\sigma \leq 20.0$ MPa

$1500 \leq T \leq 2900$, K

As stated before, equation (1) is an empirical one and is to be used only for the conceptual design of the fuel element. For validating the final design, an additional experimentally verified database must be created. The relation in equation (1) is to be used for comparison of various fuel types and operational schemes; it is not for actual construction of the fuel elements.

The relation of equation (1) was applied to the NEBA-3 reactor concept. The results of this application indicate that tungsten-clad

stabilized-UO_2 cermet fuel will be able to perform well, for many operational parameters, from the creep strength considerations. When the peak pulse is applied near the beginning of life (when burnup and therefore, fission gas inventory is low) the creep strains due to the pulse alone are calculated to be small–less than 5% for all cases analyzed.

Despite the lack of extensive and specific creep data, based on the sum total of available knowledge base pertaining to tungsten and its alloys, and the overall conservatism built into the fuel element model, it is reasonable to state that the use of equation (1) above would lead to justifiable selection of bi-modal reactor designs with respect to the creep deformation of the fuel element.

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1.0 Introduction

Creation of high specific energy sources–nuclear powered or otherwise–necessitates use of materials that can withstand high temperatures for the designed life of the source. Tungsten metal and several of its alloys meet the requirements for many of the potential applications; their features include very high melting points and excellent thermal and mechanical properties. For these reasons several space nuclear reactor concepts of the past as well as the present have considered tungsten based cermet fuel elements. One of the life limiting phenomena for such fuels is cladding failure because of creep deformation.

In this paper, a zeroeth order model for the evaluation of creep strain in cermet-fuel element concepts is developed. This model uses a proposed empirical relation for creep strain in tungsten. The available published theories and data were analyzed and indicate that this proposed empirical creep strain relation is applicable for the range of stress and temperature relevant to several reactor concepts.

2.0 Description of Typical Nuclear Fuel

The fuels being considered are, typically, made of tungsten or tungsten alloy coated spheres of stabilized UO_2. UO_2 is typically stabilized by the addition of a small quantity–usually less than 2%–of Gd_2O_3, Y_2O_3, H_fO_2, or a combination of these three oxides. These oxide additions help suppress the formation and migration of free uranium. Tungsten clad stabilized UO_2 spheres are compacted into various shapes and sizes through a powder metallurgy type process. Typical sphere diameter is of the order of 100 to 300 μm with cladding thickness of the order of 10 to 30 μm. The details of the fuel geometries being considered are provided in the design descriptions of the concepts.

3.0 Literature Search: Theories of Creep Deformation and Published Data

Tungsten is a strategic metal used extensively for industrial as well as military applications. Over 90% of incandescent light bulbs are based on tungsten filaments. Many heating elements for furnaces are tungsten based. Tungsten constitutes an important alloying element for turbine blades for industrial and military engines. Furnaces and anvils to make industrial diamonds and other high temperature materials are made of tungsten or its compounds. Given all of its applications, it is not surprising to find that tungsten has been the object of extensive study. However,

because of difficulties involved in high temperature materials preparation, instrumentation and long term maintenance of the same, only a limited amount of creep data are available in published literature. The data available are limited in stress and temperature values, environmental conditions and material types. Also, no data are available, in the literature reviewed, for in-reactor creep of tungsten.

Creep is a thermally activated time dependent process. During the stage II or steady state creep, work hardening is balanced by thermal recovery. Thermal recovery is achieved by dislocation climb, short path diffusional processes at grain surfaces and cross-slip of dislocations.

Detailed discussion of theories of creep are provided in Reference 1. Without going into details, it is noted here that for steady state creep, work hardening must exactly equal thermal recovery. It is further noted that the number of dislocation sources that can be activated, and therefore, the creep rate, is proportional to some power of the normalized stress; that is,

$$\dot{\epsilon} \propto \left(\frac{\sigma}{E} \right)^n$$

where,

$\dot{\epsilon}$ = creep rate, σ = applied stress, E = elastic modulus, and n is a temperature and material dependent constant.

Thermal recovery is an activated process and depends on diffusional characteristics of the material. Three important diffusional paths are of relevance to elevated temperature creep. They are: (i) lattice diffusion, (ii) grain boundary diffusion and, (iii) dislocation diffusion. Without going into the actual derivation, the relation between creep rate ($\dot{\epsilon}$) for the steady state creep deformation can be expressed as [see Robinson and Sherby (2)]:

$$\dot{\epsilon} = S \, \lambda^2 \, Deff \left(\frac{\sigma}{E} \right)^n$$

(1)

where

$\dot{\epsilon}$ = strain rate, s^{-1}

S = material constant, cm^{-4}

λ = grain or sub-grain dimension, cm

$Deff$ = effective diffusion rate, $cm^2 \cdot s^{-1}$

σ = applied stress, MPa

E = Elastic modulus, MPa

Further, $Deff = D_L \, F_L + D_{gb} \, F_{gb} + D_d F_d$ (2)

where,

D_L = lattice diffusion coefficient, $cm^2 \cdot s^{-1}$

F_L = fraction of active lattice atoms/sites

D_{gb} = grain boundary diffusion coefficient, $cm^2 \cdot s^{-1}$

F_{gb} = fraction of active grain boundary sites

D_d = Dislocation diffusion coefficient, $cm^2 \cdot s^{-1}$

F_d = fraction of active dislocation sites

The general validity of equation (1) for polycrystalline tungsten has been demonstrated by Robinson and Sherby (2). The diffusion coefficients are the rate controlling factors for creep deformation. Based on the creep data from several types of tungsten specimen—wrought, re-crystallized,

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powder metallurgy–having different grain sizes and initial dislocation densities, it has been shown (2) that grain boundary diffusion is not critical or rate controlling. Therefore, the relation for Deff can be simplified to:

$$Deff = D_L F_L + D_d F_d$$

Furthermore, it has been experimentally determined that the activation energies for lattice and dislocation diffusion are 140,000 kcal/mole and 90,500 kcal/mole respectively (2, 3). In addition, the analysis of published data, as reported by Robinson and Sherby suggest that:

$$Deff = 5.6 \ \exp\left[\frac{-140000}{RT}\right] + 10^{-4} \ \exp\left[\frac{-90500}{RT}\right]$$

(3)

The creep equation (Eq. 1) then is:

$$\dot{\epsilon} = S \ \lambda^2 \left\{ 5.6 \ \exp\left[\frac{-140000}{RT}\right] + 10^{-4} \ \exp\left[\frac{-90500}{RT}\right] \right\} \left(\frac{\sigma}{\epsilon}\right)^n$$

(4)

Equation (4) is a complex relation that requires knowledge of several fundamental properties of the material. Robinson and Sherby (2) have derived another, very elegant equation for creep. This equation is stated below for the completeness of literature survey:

$$\dot{\epsilon} = D_d \left[3.36 \times 10^{-25}\right] \rho_o \ \epsilon^{B\sigma} \sigma^5$$

(5)

where,

$\dot{\epsilon}$ = creep rate, s^{-1}

D_d = dislocation diffusion constant, $cm^2 \cdot s^1$

ρ_o = initial dislocation density

B = material constant

σ = creep stress

Equation (5) above is based on a number of assumptions and on data from certain ferrous alloys. It is a complex equation and not well suited for design applications.

4.0 Creep Relation for Conceptual Design Analysis

In Section 3.0 above it was stated that the creep equation based on the combination of theory and experimental data are quite complex and require detailed knowledge of the material. For conceptual design, such an approach is not practical. Therefore, we have decided to use the following simple, empirical relation:

$$\dot{\epsilon} = 1.3 \times 10^3 \left[\epsilon^{\frac{-53370}{T}}\right] \times \sigma^{3.53}$$

(6)

where,

$\dot{\epsilon}$ = steady state creep rate, h^{-1}

T = temperature, K, $1500 < T \le 2900$ K

σ = creep stress, MPa, ≤ 20.0 MPa

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Equation 6 was first developed by M. Jacox, et. al (4a), and is based on a curve fit to some of the data in Reference 4b.

We have compared additional published experimental creep rate data with the values attained from equation (6). This comparison is summarized in Table 1. In all, the results of eight experiments are compared; five with creep stress being less than 20 MPa and three being higher. When $\sigma < 20$ MPa, the calculated value for $\dot{\epsilon}$ is nearly equal to or slightly higher than the experimental value. For $\sigma > 20$ MPa, the calculated value is higher or lower by a factor of about 3 than the experimental value. Thus, it is clear that the proposed strain rate relation should be used for $\sigma < 20$ MPa.

Several specific correlations were derived for explicit stress and temperature range. It was found that, for conceptual design comparisons, equation (6), the suggested relation, is acceptable over the entire range of parameters of interest, and it predicts creep rates comparable to the experimental values.

5. Brief Discussion of Radiation Effects and Suggested Approaches to Mitigate These Effects:

It is generally accepted that under a radiation environment, the diffusion rates could be significantly enhanced because the thermally activated atomic motion is enhanced by neutron bombardment. Hence, in-reactor creep rates could be significantly higher. However, based on in-reactor data from austenitic and ferritic steels it is our belief that these effects for tungsten at the high temperatures under consideration should be small. It is reasonable to postulate that radiation enhanced creep deformation of tungsten will not exceed that due to thermal creep by more than one order of magnitude

when compared with unirradiated tungsten. However, we strongly recommend that for the final design an adequate data base regarding in-reactor creep of tungsten be developed through carefully designed experiments.

It is comforting to know that there are two alternatives available to mitigate the radiation effect. The first alternative is to use a dilute alloy of tungsten instead of pure tungsten. The effect of alloying is such that the creep rates could be decreased by up to three orders of magnitude. This is verified experimentally for low temperature ($T \leq 1500$ K) creep of several alloys of polycrystalline tungsten (10). Elevated temperature data of several investigators for single crystal tungsten and its dilute alloys as analyzed by Gao and Zee (11), also suggest similar quantitative decreases in creep rates. The reason for such a dramatic decrease in creep rates for alloys is that the solute atom(s) are effectively able to pin down the dislocations. Not only are the diffusion rates decreased, but also the number of dislocation sources—for the same stress and temperature value.

The second approach for decreasing the in-reactor creep rate is based on grain size control. Unlike other high temperature metals, such as nickel based super alloys, the steady state creep rate of polycrystalline tungsten has been found to increase with the square of the grain size (2). This would suggest that grain boundaries in tungsten do not slide significantly during creep deformation. On the contrary, they appear to add significant resistance to the creep motion. Thus, the smaller the grain size the smaller the expected steady state creep rate.

In summary, the radiation enhanced creep deformation can be controlled through dilute alloying of tungsten and by having suitably small grain and sub-grain structure.

6.   Calculated Creep Deformation for the
      NEBA-3 Concept.

In this section we have calculated creep
deformation values for the NEBA-3 concept.
The fundamental fuel design assumptions for
NEBA-3 are as follows.

The fuel element is composed of tungsten-clad
stabilized UO_2 spherical kernels which are
agglomerated through a powder metallurgy
process. However, the geometry for the creep
calculation is assumed to be that of a free
standing spherical particle of stabilized UO_2
clad with tungsten.   Three fractions of
theoretical density (0.85, 0.9, and 0.95) were
considered for stabilized UO_2. The tungsten
cladding is assumed to be 100% of theoretical
density (TD).

The fission gas generation was calculated for
different burnups, using the computer code
ORIGEN (12). Fission gases are assumed to
occupy the void spaces in the fuel, and the
only fission products contributing to the gas
pressure are Xe and Kr (13, 14, 15). Further,
the temperature dependence of fission gas
release is assumed to be as shown in Figure 1
(14, 15).

The stress in the cladding is calculated by
using the simple hoop stress model, that is,

$$\sigma = \frac{PD}{2t}$$

(7)

where,

σ = hoop stress

D = diameter of the sphere

P = pressure,

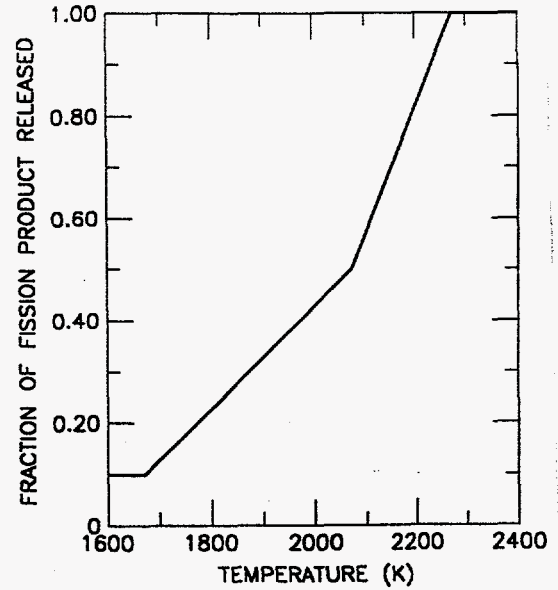t = thickness of the cladding

Figure 1

Fission Product Release

Parametric analyses of creep strain at the end
of life for various cores are shown in Figures
2 through 6. The parameters studied include
burnup, cladding thickness, fuel theoretical
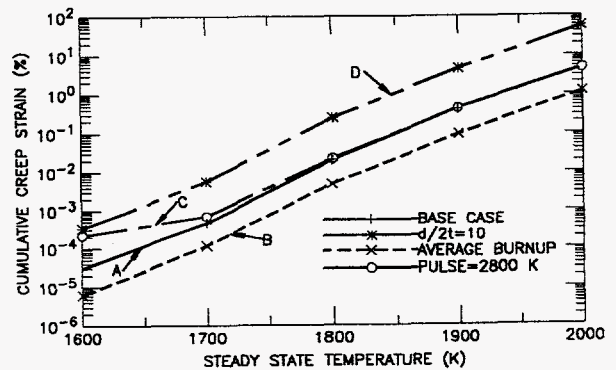density and operating temperatures.

Figure 2

Sensitivity Analysis For NEBA-3

Figure 2 shows results of calculations for a
specific design. The base case (curve A)
assumes the following:

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a) Power History ⇒ Peak pulse is assumed at the beginning of life with power at peak equal to about eight times the power at steady state. The temperature at peak power is 2300 K and stays at this level for one hour. The steady state temperature values for the 10 year life were varied from 1600 K to 2000 K.

b)   Fuel density = 0.9 of theoretical density

c)   D/2t = 5.0

d)   Peak burnup = 0.2%, with peak to average = 1.5.

For the base case (curve A), the 1600 K steady state temperature produces a negligible cumulative strain (3×10^{-5} %) at the end of life for the peak burnup locale. Raising the steady state temperature to 2000 K increases the cumulative strain at peak burnup location to about 5%, at the end of life.

Figure 2 also shows sensitivity to other parameters. Curve B shows the cumulative strain for the average (burnup) particle. The cumulative creep strain at the end of life for 1600 and 2000 K steady state operation are about 7×10^{-6} % and 1.0%, respectively. Curve C is for the same conditions as the base case (curve A), except that the temperature at the peak of pulse is equal to 2800 K instead of 2300 K. There is no significant difference (in creep strain) between curves A and C. Curve D represents thinning of the cladding by a factor of 2. Obviously the cumulative strain values are higher by a factor of about 12 (~ $(2)^{3.53}$ = 11.6). The end of life stress (not shown in the figure) for the base case varies from 1.4 MPa at 1600 K to 6.7 MPa at 2000 K.

Figures 3 and 4 show creep strains for the average particle at different operating temperatures and burnups. In these sets of results the variables were as follows.
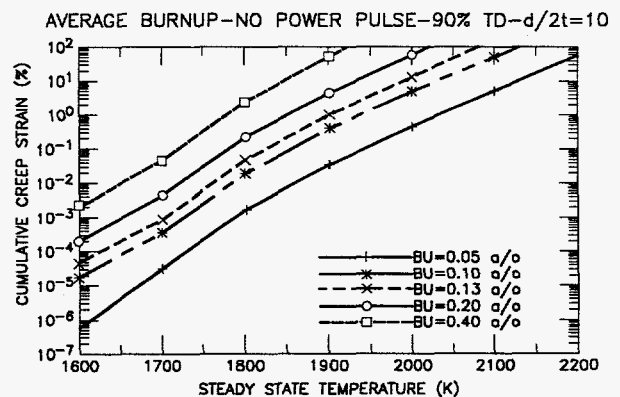

AVERAGE BURNUP–NO POWER PULSE–90% TD–d/2t=10

Figure 4

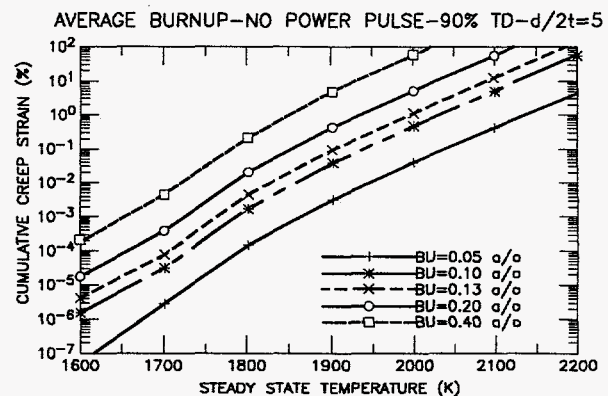Sensitivity Analysis For NEBA-3

AVERAGE BURNUP–NO POWER PULSE–90% TD–d/2t=5

Figure 3

Sensitivity Analysis For NEBA-3

a) No power pulse;

b) UO_2 density 90% of TD, and

c) D/2t = 5, and 10 for figures 3 and 4, respectively.

The results plotted show that increasing burnup results in increased cumulative strain values. For the same operating temperature the cumulative creep strain increases with burnup (stress)–through the stress exponent (equation 6).
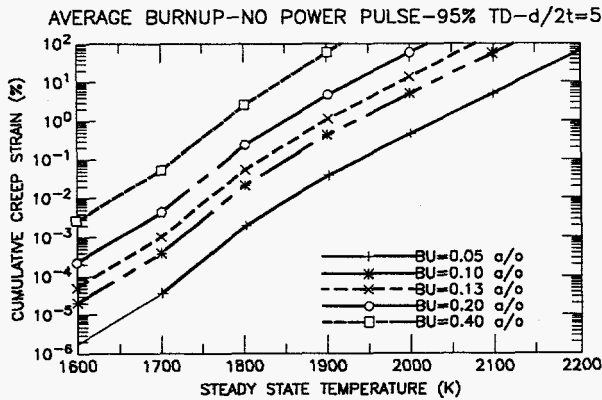
AVERAGE BURNUP-NO POWER PULSE-95% TD-d/2t=5

Figure 5

Sensitivity Analysis For NEBA-3
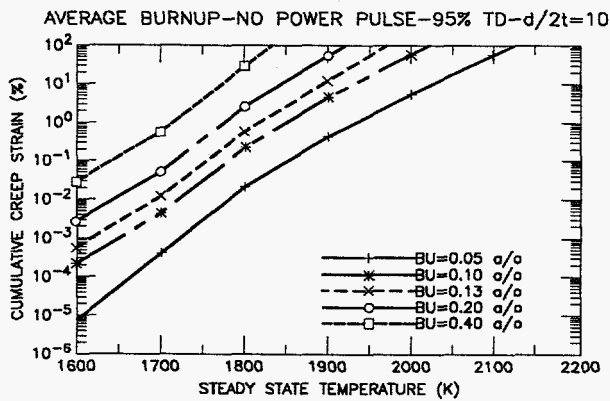
AVERAGE BURNUP-NO POWER PULSE-95% TD-d/2t=10

Figure 6

Sensitivity Analysis For NEBA-3

Figures 5 and 6 show the same sensitivity as for Figures 3 and 4 except that the fuel density is equal to 95% TD instead of 90% TD.

The results presented in Figures 2 through 6 can be used to set operating bounds on the fuel elements used in the conceptual designs. Note that, these curves are not restricted to use on NEBA-3 designs but are of more general validity for fuels of similar construction.

Summary:

Several nuclear reactor concepts are being developed for bi-modal operation at low steady state power for space applications. Some of these concepts use UO_2-tungsten cermet fuel. One of the life limiting phenomena is due to creep of the tungsten cladding. Creep is a very complex phenomenon and data for tungsten is limited even without considering the effects of radiation. However, in order to perform a downselection of different reactor concepts a zeroeth order model had to be developed even with this scarce data. An empirical relation for creep strain was suggested. Analysis of the available published theories and data show that the suggested relation seems to be valid for the range of stress and temperature applicable to the proposed concepts. This relation was therefore used to develop a zeroeth order model using the proposed creep strain relation and results of parametric analyses were provided. The parameters varied were burnup, fuel density, cladding thickness, power history and temperature. The results shown in Figures 2 through 6 can be used to set operating bounds in the fuel elements used in the actual designs. It is stressed that the proposed creep strain relation for tungsten can only be used for downselection of reactor designs. Final design will require detailed experimental validation.

The proposed creep rate relation as stated in this paper simply allows one to calculate cumulative strain. The design community has

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to agree on an acceptable value of creep strain–which may be much smaller than the rupture strain or total strain at the end of stage II creep and use that value consistently for all designs. Also, the computation of stress values should be carefully reviewed for the final design. The final design should be based on the concept of effective stress which would take into account all of the ramifications of three dimensionality of the fuel element including the subtle but significant differences between "thin shell" and "thick shell" considerations. The approximations used in the present paper, however, allow one to downselect the fuel and reactor concepts quickly.

Based on the analyses performed using model described, it is reasonable to conclude that tungsten based cermet type fuel elements are conceptually valid and procedure to the next step of preliminary design is encouraged.

Table 1. Comparison of Experimental Data with Calculated Values for Steady State Creep of Polycrystalline Tungsten.

Temp.,°K	Creep Stress, MPa	Creep Rate, h⁻¹/
		Experimental Data/ Calculated Value*
1923	27.6	$3.0 \times 10^{-4} / 1.4 \times 10^{-4}$ (5)
1923	75.8	$1.3 \times 10^{-2} / 5.0 \times 10^{-3}$ (5)
2073	34.5	$9.6 \times 10^{-4} / 2.3 \times 10^{-3}$ (9)
2203	13.8	$3.5 \times 10^{-4} / 4.1 \times 10^{-4}$ (8)
2473	6.9	$3.8 \times 10^{-4} / 5.0 \times 10^{-4}$ (5)
2473	9.7	$1.0 \times 10^{-3} / 1.6 \times 10^{-3}$ (5)
2473	10.3	$2.6 \times 10^{-3} / 2.1 \times 10^{-3}$ (7)
2473	13.8	$4.5 \times 10^{-3} / 5.8 \times 10^{-3}$ (5)

*Experimentall value is average of three tests. Number in parenthesis is the reference for the experimental value.

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