Breakaway properties of film formed on copper and copper alloys in erosion-corrosion by mass transfer equation

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Abstract

Erosion-corrosion tests on copper and three types of copper alloys in a 1 wt% solution of $CuCl_2$ were carried out at various flow velocities using a jet-in-slit testing apparatus, which is capable reproducing various hydrodynamic conditions. A damage profile of a specimen was developed using a surface roughness meter to evaluate locally occurring damage. The damage depth rate for copper, beryllium copper and a 70/30 copper nickel alloy increased with increasing flow velocity, and suddenly increased at a certain velocity, which is called the breakaway velocity. The breakaway velocities at the central and disturbed part of a specimen were different for each sample, indicating that the hydrodynamic conditions of the flowing solution had an effect. The damage depth rate, calculated from the mass transfer equation, which involved mass transfer in the concentration diffusion layer and in the corrosion product film, could be fitted to the experimental data, suggesting that the mass transfer equation can be applied to the evaluation of erosion-corrosion damage. The corrosion product film was exponentially broken away at velocities higher than the breakaway velocity. The breakaway properties of the corrosion product film were confirmed to be different for each material, since the power of the exponential equation was different for each sample.

1 Introduction

Localized corrosion frequently occurs near the inlet ends of heat exchanger tubes which are made of a copper alloy where seawater is used.   It occurs when the protective film of a corrosion product, formed on the surface of the copper alloy, is broken away by shear stress and turbulence, causing the underlying metal surface to come into direct contact with the corrosive liquid.   This phenomena is known as erosion-corrosion, flow induced localized corrosion, FAC as flow accelerated corrosion or flow assisted corrosion etc [1-3].   Damage by erosion-corrosion largely depends on hydrodynamic conditions such as the flow velocity of a liquid, and this type of corrosion is characterized by the "breakaway velocity" at which the surface protective film is destroyed as the flow velocity increases [4].

In order to predict the extent of damage to copper alloys under a flowing solution, it is imperative to elucidate relationships between damage occurring in the materials and the hydrodynamic conditions of a corrosive solution.   Erosion-corrosion of copper alloys proceeds via a diffusion controlled process in many cases, and the mass transfer equation for an oxidizing agent over the material surface is generally adopted.   To apply the mass transfer equation to erosion-corrosion damage, both mass transfers in the concentration boundary layer and in the corrosion product film on the material need to be considered, because a corrosion product film formed on the material confers a resistance to corrosion [5, 6].   Flow velocity is generally used as a hydrodynamic parameter to relate erosion-corrosion damage, because it is quite simple.   However, it is not sufficient for the accurate prediction of damage, since erosion-corrosion frequently occurs in the turbulent portion, where the direction of flow changes, such as a pipe bend, an elbow and tee pipe fittings.   Several papers have reported that the Sherwood number, a dimensionless number used in mass transfer operations, is useful

for the mass transfer coefficient in the concentration boundary layer [7-11].   Poulson reported that the Sherwood number in many flowing conditions can be estimated through electrochemical measurement [9].   However, the Sherwood number might not also be accurate, in terms of describing the condition of a corrosion product film.   Nešićet et al. conducted a numerical simulation of turbulent flow, when a rust film was present, and found that the turbulent flucutations affect both mass transfer through the boundary layer and the removal of the film [12].   A numerical simulation of a pipe flow, to investigate erosion-corrosion, has also been reported [13-15].

In this study, erosion-corrosion tests for copper and three types of copper alloys were conducted in a 1wt% $CuCl_2$ solution under various flow velocities using a jet-in-slit testing apparatus.   A damage profile of a specimen was prepared using a surface roughness meter to evaluate local damage.   The depth of the damage, calculated using the mass transfer equation was related to the experimental data to confirm the applicability of the equation. Using the mass transfer coefficient in corrosion products film obtained from mass transfer, the condition of the film and the breakaway properties were compared for each material.

2 Experimental

The jet-in-slit testing apparatus used in the erosion-corrosion test is shown in Fig. 1.   The testing apparatus consisted of a test solution reservoir, a pump, a flow meter and a test cell. Figure 2 shows a detailed schematic rendering of the test cell.   In this apparatus, the test solution was allowed to flow from the periphery of the slit between the specimen and the nozzle into the nozzle mouth.   The diameter of the specimen was 16 mm.   The nozzle was made of a polymethyl-methacrylate resin with a bore diameter of 1.6 mm.   The gap between the nozzle top and the specimen was 0.4 mm.   As the solution was injected from the nozzle

mouth into the slit, the solution filled the slit and flowed radially over the specimen surface. As it approached the periphery of the specimen, the cross-sectional area of the flow increased, and accordingly the flow velocity decreased. The rapid reduction in flow velocity created a shear stress and an intense turbulence in the flow [16]. As a result, localized corrosion damage in the jet-in-slit test can be accounted primarily by shear stress and the turbulence of the flow.

A 1 wt% $CuCl_2$ solution saturated with air was used as the test solution. Cu^{2+} was used as the oxidizing agent to accelerate the corrosion reaction. The temperature of the test solution was maintained at 40ºC. The flow velocities at the nozzle outlet were varied from 0.2 to 7.5 m s^{-1}. At a flow rate of 0.4 L min^{-1}, the fluid velocity at the nozzle outlet was 3.3 m s^{-1} and the Reynold's number at that point was 8100. The test duration was 1h.

The materials used in the investigation were pure copper (Cu) and three types of copper alloys, namely beryllium copper alloy (BeCu) and two types of copper nickel alloys (70CuNi and 30CuNi). Their chemical compositions are shown in Table 1.

Damage depth was determined by comparing the difference in the specimen surface profile before and after the test using a surface roughness meter and measurement of the mass loss of the specimen. The damage depth rate was obtained by converting the maximum damage depth into mm y^{-1}.

3 Mass transfer equation

Various hydrodynamic parameters have been proposed to control the occurrence and extent of erosion-corrosion. The mass transfer coefficient is a parameter relating the rate of a diffusion controlled reaction to the concentration driving force incorporated from both diffusional and turbulent transport processes. Erosion-corrosion of copper alloys mainly

proceeds under cathodic control because the rate controlling step in corrosion is the transport of the oxidizing agent from the bulk of the fluid to the metal surface. When the surface of the copper alloy is exposed to a flowing fluid, a concentration boundary layer is formed in the bulk of the fluid outside of the corrosion product film, as shown in Fig. 3. The diffusion rates of the oxidizing agent, r_1 and r_2 in the concentration boundary layer and in the corrosion product film, respectively, can be determined as follows;

$$r_1 = k_c \, (c_b - c_d) \tag{1}$$

$$r_2 = k_d \, (c_d - c_w) \tag{2}$$

where c_b, c_d and c_w (mol L^{-1}) are the oxidizing agent concentration in the bulk of the flowing fluid, at the outside surface of the corrosion products film and at the metal surface respectively. k_c and k_d (mm y^{-1}) are the mass transfer coefficient in the concentration boundary layer and in the corrosion products film respectively.

The corrosion rate should be proportional to the diffusion rate of the oxidant. In the steady state, the mass transfer rates in the concentration boundary layer and that of the corrosion product film are equal to each other. Accordingly, the corrosion rate, Rc (mm y^{-1}), can be given as follows by using a conversion factor, K (L mol^{-1}),

$$Rc = K \, r_1 = K \, r_2 \tag{3}$$

The concentration of oxidizing agent at the metal surface, c_w, may be zero ($=0$), since a very rapid electrochemical reaction is assumed there. Equations (1)-(3) then give

$$Rc = K \, c_b \, / \, (\, 1/k_c + 1/k_d) \; = c_b \, / \, (\, 1/Kk_c + 1/Kk_d) \tag{4}$$

Equation (4) indicates that the corrosion rate is directly proportional to the concentration of the oxidizing agent, c_b, and inversely proportional to the combined resistance to mass transfer, $1/Kk_c+1/Kk_d$. The concentration of the oxidizing agent, c_b, is 0.075 mol L^{-1} which corresponds to a 1 wt% CuCl$_2$.

The issue of whether the mass transfer equation might be applied to the experimental results was examined. The problem is how to determine the mass transfer coefficients, k_c and k_d. According to the definition of the mass transfer coefficient, the coefficient in the concentration boundary layer, k_c, is inversely proportional to the thickness of the concentration boundary layer. It was previously determined that the thickness is dependent on the flow velocity and is inversely proportional to the velocity to the power of 0.5 for a laminar flow and of 0.8 for a turbulent flow [17]. Accordingly,

$$k_c \quad u^{0.5} \quad \text{(for laminar flow, Re<2300)} \tag{5}$$

$$k_c \quad u^{0.8} \quad \text{(for turbulent flow, Re>2300)} \tag{6}$$

It may be assumed that the mass transfer coefficient in the corrosion product film, that k_d, is also inversely proportional to the thickness of the corrosion product film, but was originally independent of flow velocity, because the thickness of the corrosion product film was maintaned nearly constant. After the increase of the corrosion rate, it was assumed that k_d depends on the flow velocity to the power, i.e., k_c. This is because the surface after the breakaway of the corrosion products film consisted of a completely naked area and at the same time the area is still covered with the residue of the corrosion product [16]. Accordingly,

$$Kk_d = \alpha \qquad (\text{constant}, < \text{breakaway velocity}) \qquad (7)$$

$$Kk_d = \beta u^n \qquad (> \text{breakaway velocity}) \qquad (8)$$

where α, β *and* n are constant.   Under thes assumptions, Kk_c and Kk_d were determined and fitted experimental data.

4 Results and discussion

4.1 Damage profile

Cross-sectional profiles of Cu and BeCu specimens after the test at a flow rate of 0.8 L min^{-1} are shown in Fig. 4.   The dotted line indicates the profile before the test determined as volume loss calculated from mass loss and the density of a specimen coinciding to the volume loss calculated from the surface profile before and after the test.   Both Cu and BeCu specimens were largely damaged at the central part of the specimen (A) and an area approximately 2 mm from the center of the specimen (B).   The damage at part A is due to shear stress and part B to turbulence, as described above [16].   The ratio of damage in the central part A and the disturbed part B in Cu specimen is approximately two thirds.   On the other hand, the ratio for the BeCu specimen is about half.   Thus, the damage to the BeCu specimen was considerably larger at the central part.   This indicates that the resistance to the corrosion for a corrosion product film depends on the hydrodynamic conditions of flowing solution.   In order to evaluate the hydrodynamic effect on erosion-corrosion damage in detail, the central part within a 1 mm radius of the specimen and the disturbed part, 2 to 3 mm from the center of the specimen were chosen and the maximum damage depth at both parts of each

specimen at various velocities were measured.

4.2 Damage depth rate and fitting by mass transfer equation

Figure 5 shows the relationship between the damage depth rate at the central part and the disturbed part of a Cu specimen and the flow velocity.   The solid curves indicated in the figure were calculated from mass transfer equation and fitted to the experimental data. According as the same procedure, the experimental data and the fitted lines for BeCu, 70CuNi and 30CuNi are shown in Figs. 6, 7 and 8 respectively.   The coefficient in the concentration boundary layer, Kk_c, was determined and fitted to all the experimental data.   Thus the constants $α$, $β$ and n in equation (7) and (8), which are related to the mass transfer coefficient in a corrosion product film, are listed in Table 2.   These parameters are discussed below.

The damage depth for the Cu specimen increased slightly with increasing flow velocity at lower velocities (Fig. 5).   The damage depth increased rapidly at a certain velocity, namely the breakaway velocity [4].   The breakaway velocity at the central part is 2 m s^{-1} and at the disturbed part, 0.8 m s^{-1}, quite different values.   The damage depth at a velocity lower than the breakaway velocity was on same curve for both the central and disturbed parts.   The damage depth doubled at the breakaway velocity at both parts, and then became deeper with increasing flow velocity.   This indicates that the corrosion product film formed on the Cu was easily broken away by turbulence occurring at the disturbed part in preference to shear stress.   It was confirmed that the damage depth, determined by the mass transfer equation, was well fitted to experimental damage depth of Cu, although the damage at each part of the specimen was different.

The damage depth rate for the BeCu specimen increased with increasing flow velocity, but was relatively low, compared to the rate for the Cu specimen (Fig. 6).   The breakaway

velocity at the central part is 0.8 m s^{-1} and at the disturbed part was 0.5 m s^{-1}, and the breakaway velocity was lower than that for the Cu specimen. The damage depth rate was very low at a velocity lower than the breakaway velocity, compared to the rate for the Cu specimen. At a velocity higher than the breakaway velocity, the damage depth rate increased slightly with increasing flow velocity, however, the behavior of the damage depth rate was similar at both the central and disturbed parts. The corrosion behavior of the BeCu specimen was different from the Cu specimen. The damage depth rate, calculated from the mass transfer equation, could also be fitted to the experimental data for the BeCu sample.

Concerning 70CuNi, a breakaway velocity of 3 m s^{-1} at the central part and 1 m s^{-1} at the disturbed part were observed (Fig. 7). The damage depth rate became three times higher at the breakaway velocity, and then increased with increasing flow velocity. Although the damage was low compared to the damage for the Cu specimen, the corrosion behavior was similar to that of Cu specimen. This was caused by the formation of a good quality film against corrosion, due to the addition of nickel. The damage depth rate at a velocity lower than the breakaway velocity came into line, but the curve calculated from the mass transfer equation did not coincide to experimental damage rate. This actually seems to be caused by a slight breakaway of the film, even though the damage was not fatal. The damage depth rate at a velocity higher than the breakaway velocity simulated the experimental rate.

The damage depth rate for 30CuNi was constant at both the central and disturbed parts at all velocities (Fig. 8). This shows that 30CuNi is an excellent film for protecting against erosion-corrosion, even if the flow velocity is high. The damage depth rate calculated from the mass transfer equation was fitted to the experimental results well.

As a result, the damage depth rate was different at the central and disturbed parts, and was dependent on hydrodynamic conditions. However, it was confirmed that the mass transfer equation and the assumption concerning the corrosion products film as described by equations

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(7) and (8) can be applied to erosion-corrosion damage.

4.3 Characterization of film

Relationships between flow velocity and the mass transfer coefficient in a corrosion product film, Kk_d, at the central part of each specimen are shown in Fig. 9. The mass transfer coefficient in the concentration boundary layer, Kk_c, is shown as a dotted line in the figure. The dotted line of the Kkd curves shows the breakaway velocity for each material. The mass transfer coefficient in the corrosion product film, Kk_d, of Cu is close to the value of the mass transfer coefficient of the concentration boundary layer, Kkc, at a velocity lower than the breakaway velocity. Therefore, the damage rate for Cu depends on the concentration boundary layer at a lower velocity, however, the damage rate of the other copper alloys is determined by the condition of the corrosion product film, because the Kk_d of other copper alloys was very low compared with Kkc. This is equivalent to α as shown in Table 2, and the resistance to the corrosion of the film formed on BeCu, 70CuNi and 30CuNi was enhanced by more than twice compared to the film formed on the Cu. At a velocity higher than the breakaway velocity, the Kkd for copper alloys was always lower than Kkc. Because of this, the damage rate is mainly dependent on the mass transfer rate in the corrosion product film, namely the damage rate must be determined only by the corrosion products film. Concerning the Kkd at a velocity higher than the breakaway velocity, the slope of the line, listed in Table 2 as a power of n, was quite different for each material. The constant, n, appears to be the breakaway property of the film formed on each material. The constant, n, for Cu was 0.6, similar to the change in the thickness of concentration boundary layer, the same as Kkc. The film formed on BeCu was breakaway with difficulty, since the constant for BeCu was as low as 0.3. The breakaway property of the film formed on

70CuNi was almost proportional to the flow velocity, since the constant, *n*, was 0.9.   Thus, this confirms that the breakaway property of each material was different at the central part, where the shear stress was dominant.

Relationships between flow velocity and mass transfer coefficient in the corrosion product film, Kk_d, at the disturbed part of each specimen are shown in Fig. 10.   At a velocity lower than the breakaway velocity, it was quite the same as the behavior at the central part.   At a velocity higher than the breakaway velocity, the *Kkd* for Cu became larger than the value of *Kkc*.   This indicates that mass transfer in the concentration boundary layer was dominant, although small amounts of corrosion product film might remain on the surface of a specimen. The damage for 70CuNi affected both mass transfer coefficients, since the *Kkd* for 70CuNi was comparable to *Kkc*.   The *Kkd* for BeCu is so low that mass transfer in the corrosion product film was dominant as well as that at the central part.   The power, *n*, of the exponential equation for *Kkd* at a velocity higher than the breakaway velocity was different for each material, however, it was the same as that at the central part.   Thus, although the extent of breakaway of the film formed on each material was dependent on hydrodynamic conditions, the increasing ratio for breakaway of the film on flow velocity does not depend on hydrodynamics but on the type of material.

As a result, in order to predict erosion-corrosion damage for a copper based material, the mass transfer equation can be used as a fundamental equation, however, flow velocity is not adequate for expressing various hydrodynamic conditions such as turbulence and shear stress. The Sherwood number seems to be more suitable than the flow velocity.   Of course, the mass transfer coefficient in concentration boundary layer might be predicted, however, it seems to be difficult to predict the mass transfer coefficient in a corrosion product film, because the Sherwood number was almost propotional to flow velocity in the range of less than 10000 for the Reynold's number in an impinging jet testing apparatus, similar as the

apparatus used in this study [8]. A parameter for hydrodynamic conditions instead of flow velocity and the Sherwood number would be desirable for predicting the breakaway property of a corrosion product film. Observing the corrosion product film formed on Cu the specimen tested at a lower flow velocity, it consisted of numerous particles, the size of which was approximately 5 μm in diameter. The breakaway of the corrosion product film was equivalent to removing particles due to the hydrodynamic action of the flowing solution. Concerning the removal of the particles, it is important to investigate relationships between their morphology and adhesive force. Considering these facts, selecting a hydrodynamic parameter related to erosion-corrosion was the most important initial issue. The mass transfer coefficient can be determined by electrochemical measurements, but it was thought that the force acted on the surface of the material, for instance, a pressure measurement was useful. It is reasonable to use the mass transfer equation as a basic equation. The prediction of erosion-corrosion damage in an actual machine should be developed, along with a numerical simulation [13-15].

5 Conclusions

Erosion-corrosion tests for copper and three types of copper alloys were carried out in 1 wt%CuCl$_2$ solution using a jet-in-slit testing apparatus, and the results of the test led to the following conclusions;

1. The damage depth rate of Cu, BeCu and 70CuNi increased with increasing flow velocity, and the breakaway velocity was clearly recognized. The breakaway velocity at the central and disturbed parts of the same material was different, i.e., 2 m s^{-1} and 0.8 m s^{-1} for Cu, 0.8 m s^{-1} and 0.5 m s^{-1} for BeCu, and 3 m s^{-1} and 1 m s^{-1} for 70CuNi. No breakaway velocity was recognized for 30CuNi. The film formed on Cu and 70CuNi was largely

damaged by turbulence, resulting in the hydrodynamic conditions of the flowing solution affecting the breakaway property of the corrosion product film.

2. Damage depth rate, calculated by the mass transfer equation, which involved mass transfer in the concentration diffusion layer and in the corrosion product film, could to be fitted to the experimental damage rate, and it was confirmed that the mass transfer equation can be applied to erosion-corrosion damage for copper and copper alloys.

3. The film condition was almost constant at a velocity lower than the breakaway velocity, and the film formed on BeCu, 70CuNi and 30CuNi was enhanced by more than twice compared to the film formed on the Cu, as evidenced by an analysis of the mass transfer coefficient in the corrosion product film.   The corrosion product film was exponentially broken away at a velocity higher than the breakaway velocity.   The breakaway property of the corrosion product film was confirmed to be different for each material, since the power of the exponential equation was different for each material.

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Captions

Fig. 1.   Schematic diagram of a jet-in-slit testing apparatus.

Fig. 2.   Detail of the test section in the jet-in-slit testing apparatus.

Fig. 3.   Distribution of oxidizing agent concentration in a flowing solution over a metal surface.

Fig.4.   Cross-sectional profile of a copper specimen (upper) and a BeCu specimen (lower) tested in a solution flowing at 0.8 L min^{-1} for 1 h.   The dotted line is the profile before the test.   A and B are the central and disturbed parts.

Fig.5.   Relationship between flow velocity and damage depth rate at the central and disturbed parts of a pure copper (Cu) specimen tested in a jet-in-slit testing apparatus. Lines were calculated using mass transfer equation as fitted to the experimental data.

Fig. 6.   Relationship between flow velocity and damage depth rate at the central and disturbed parts of a beryllium copper alloy (BeCu) specimen tested in a jet-in-slit testing apparatus.   Lines were calculated using mass transfer equation as fitted to the experimental data.

Fig. 7.   Relationship between flow velocity and damage depth rate at the central and disturbed parts of a copper nickel alloy (70CuNi) specimen tested in a jet-in-slit

testing apparatus. Lines were calculated using mass transfer equation as fitted to the experimental data.

Fig. 8. Relationship between flow velocity and damage depth rate at the central and disturbed parts of a copper nickel alloy (30CuNi) specimen tested in a jet-in-slit testing apparatus. Lines were calculated using mass transfer equation as fitted to the experimental data.

Fig. 9. Values of *Kkc* and *Kkd* determined as fitted to the damage depth rate at the central part of a specimen.

Fig. 10. Values of *Kkc* and *Kkd* determined as fitted to the damage depth rate at the disturbed part of a specimen.

Table 1 Chemical composition of the copper alloys used in the tests.

Table 2. Constants for the mass transfer equation determined as fitted to the damage depth rate of each copper alloy.

Fig. 1. Akihiro Yabuki et. al.

Slit adjuster

Specimen holder

Moving rod

Specimen

Nozzle

⇐ Test solution

⇓

Test solution
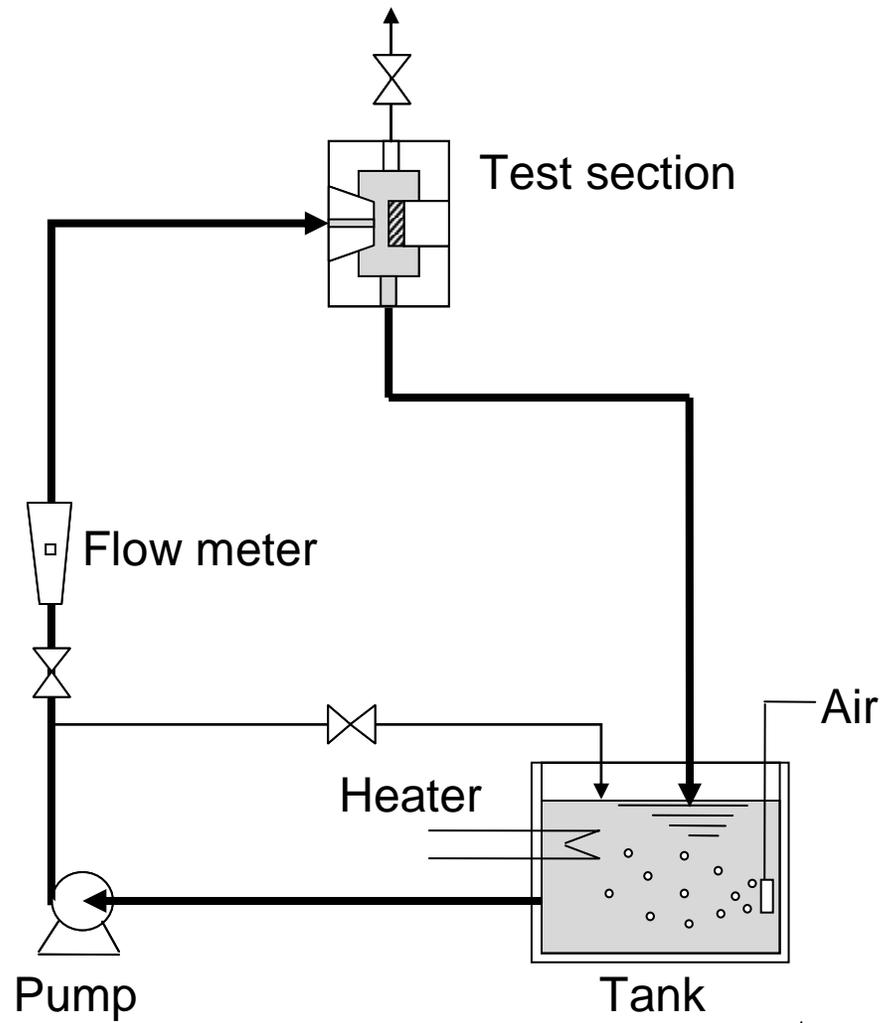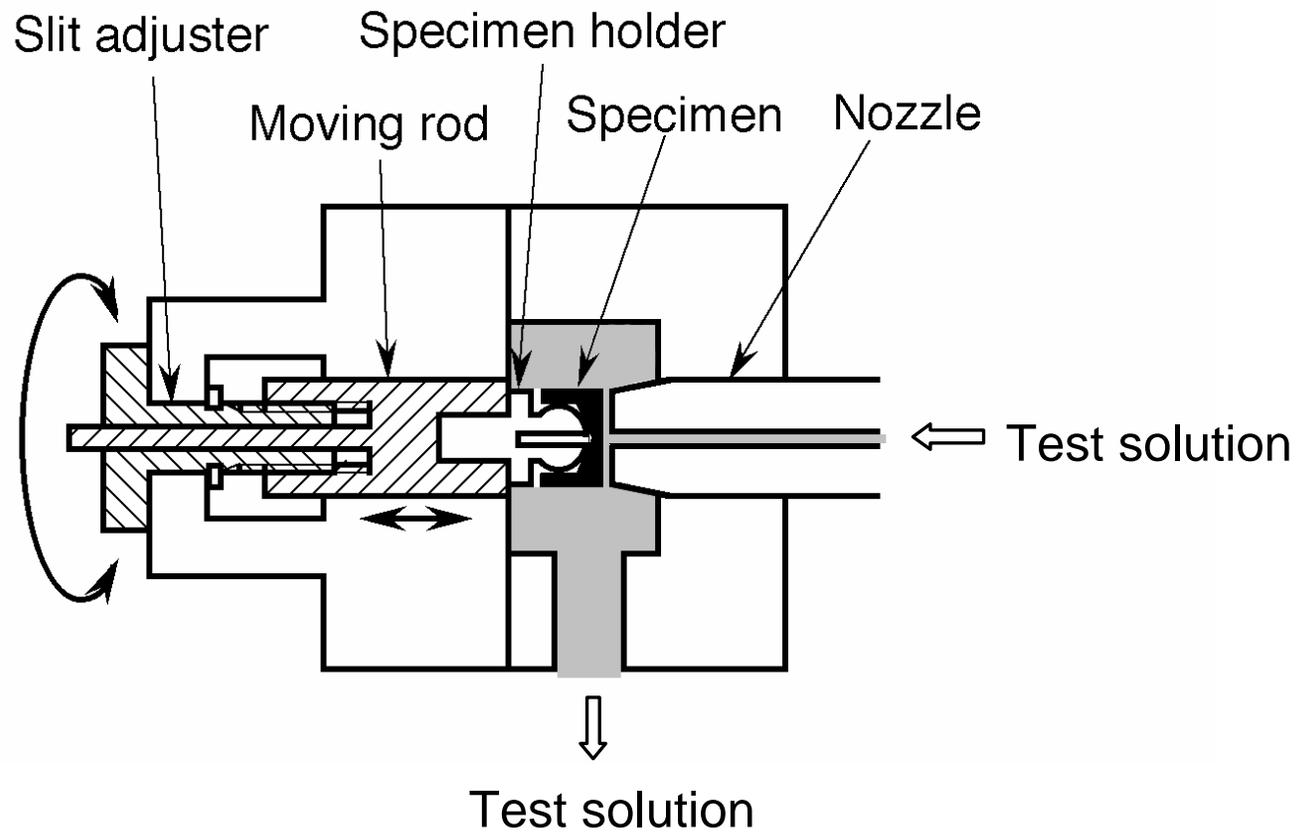
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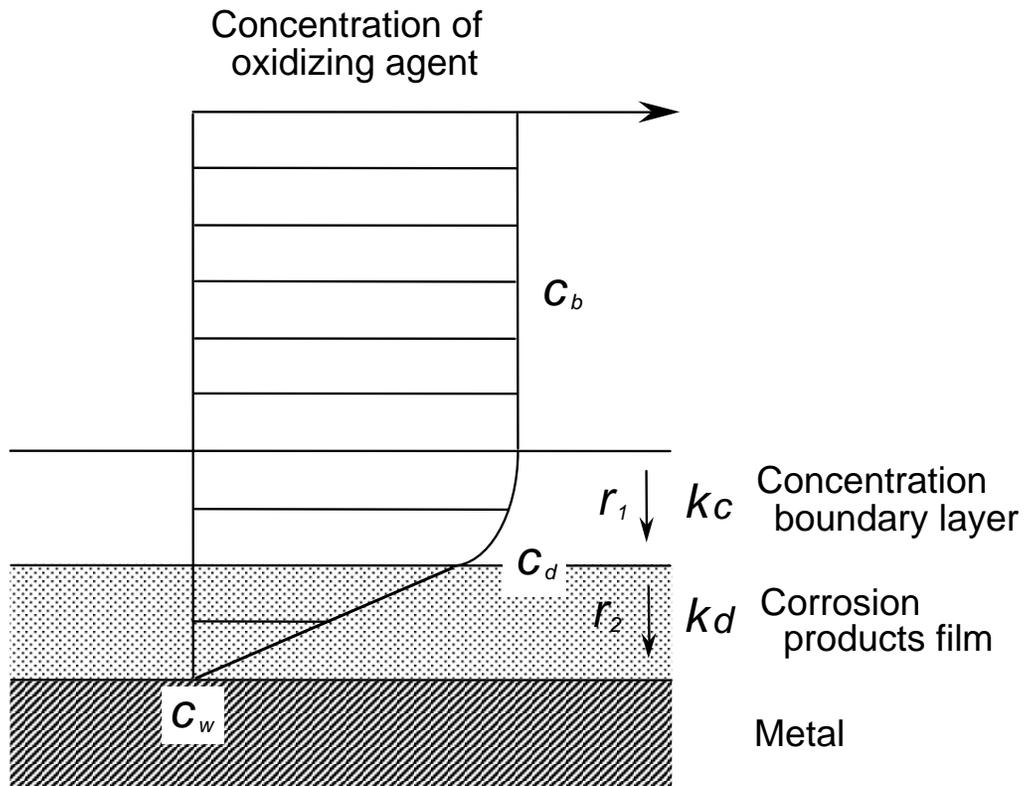
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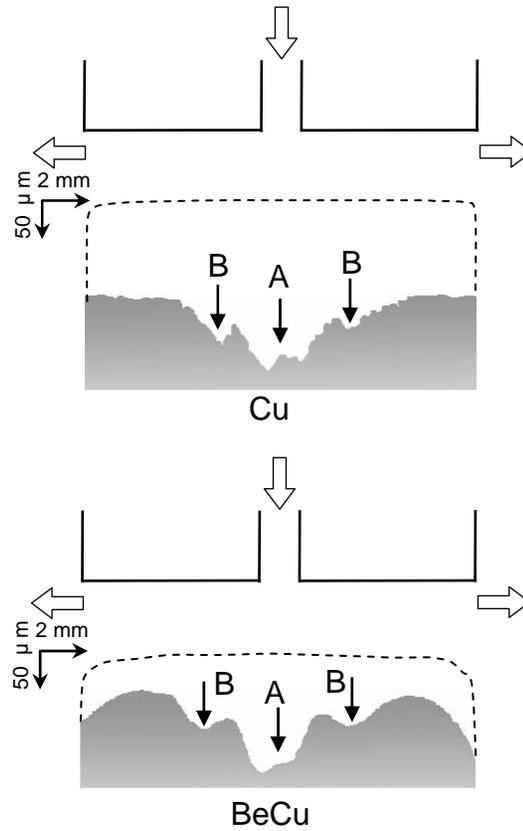
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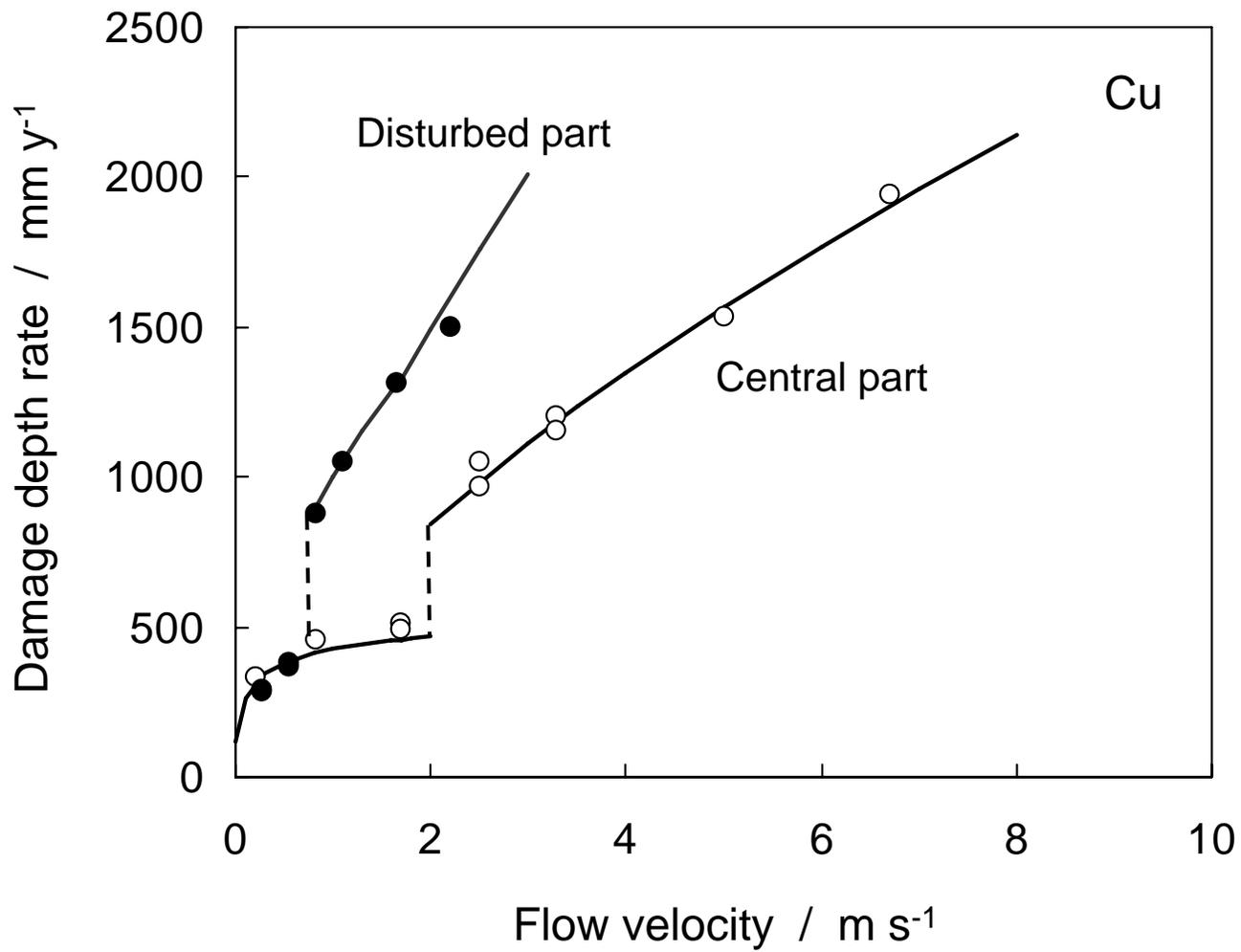
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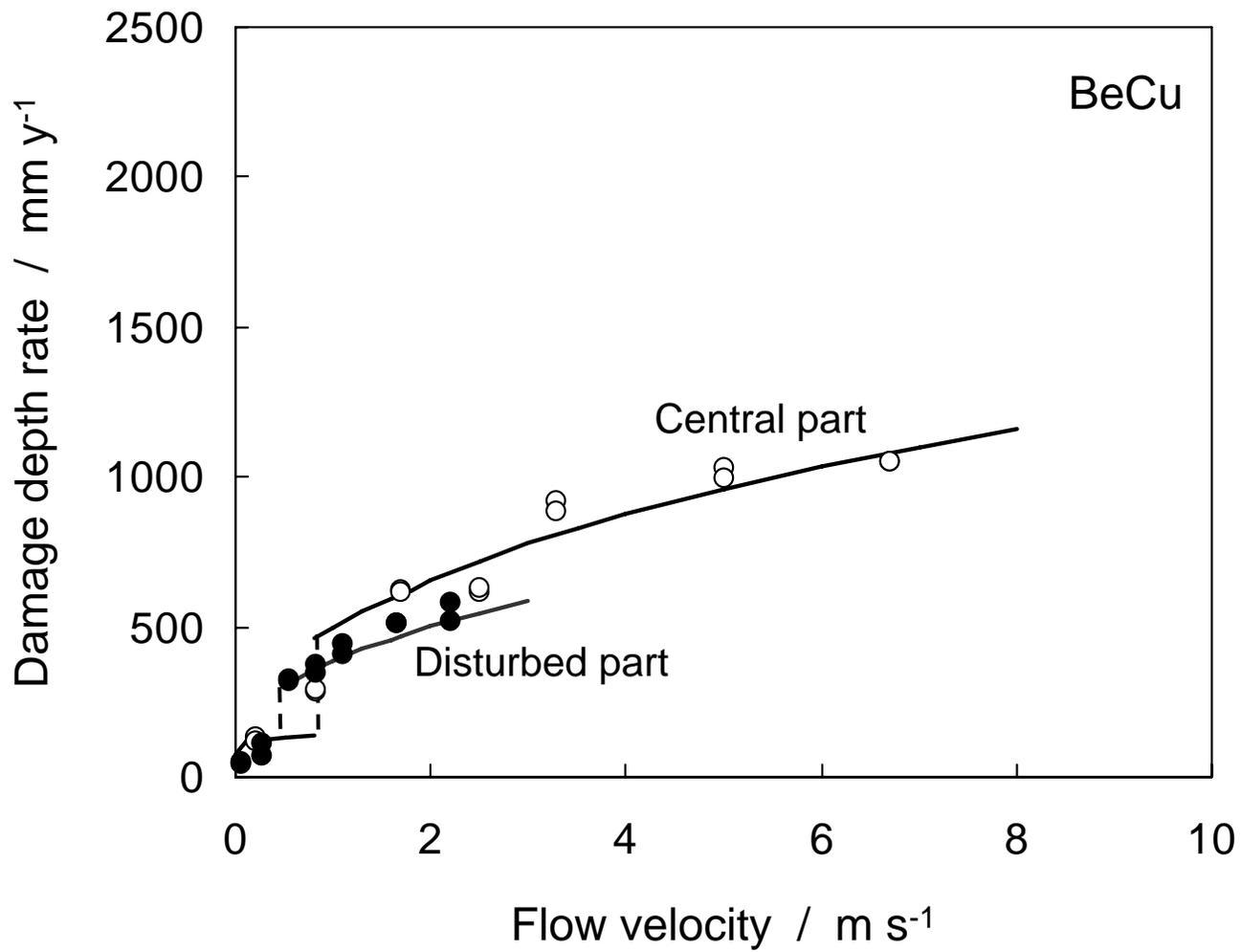
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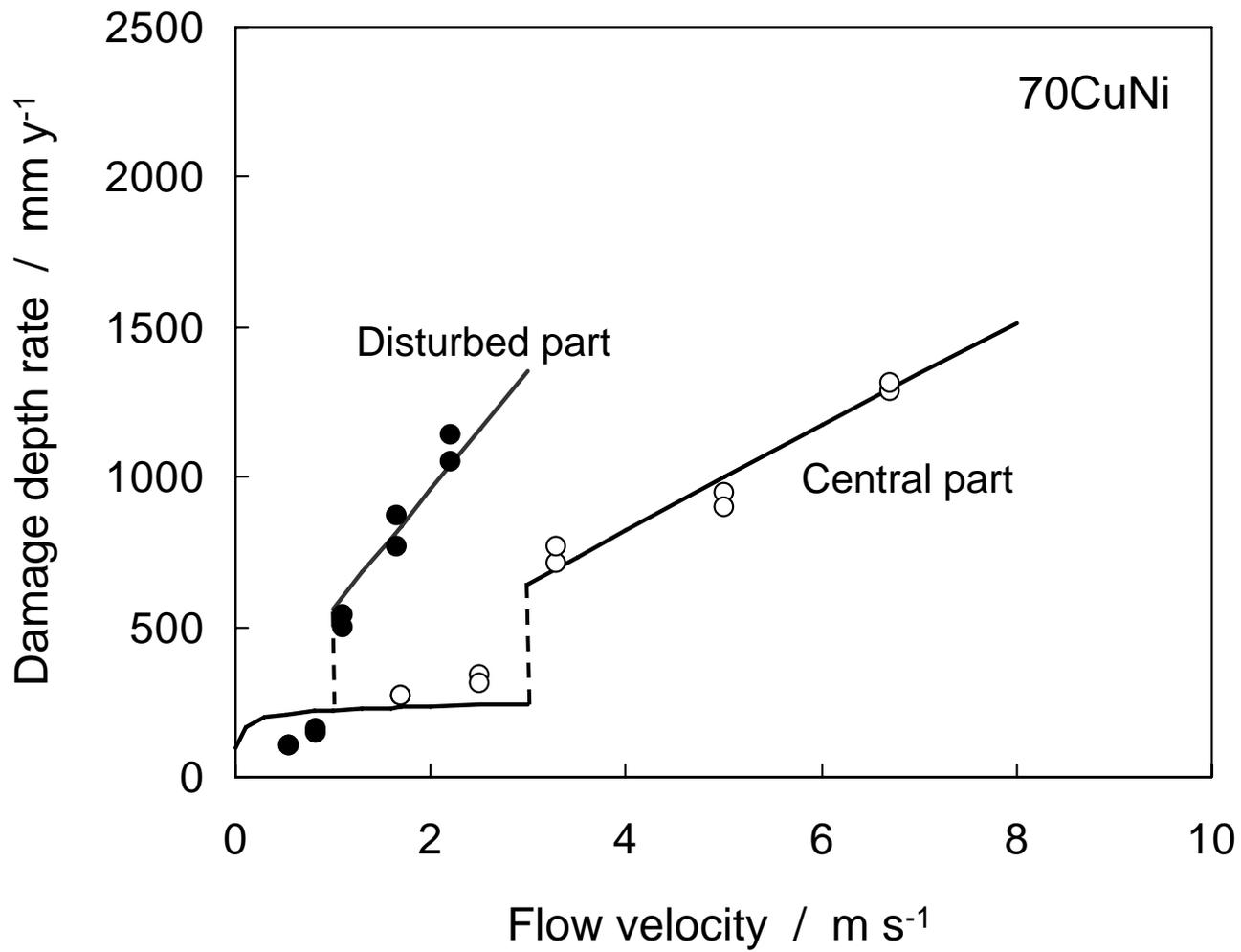
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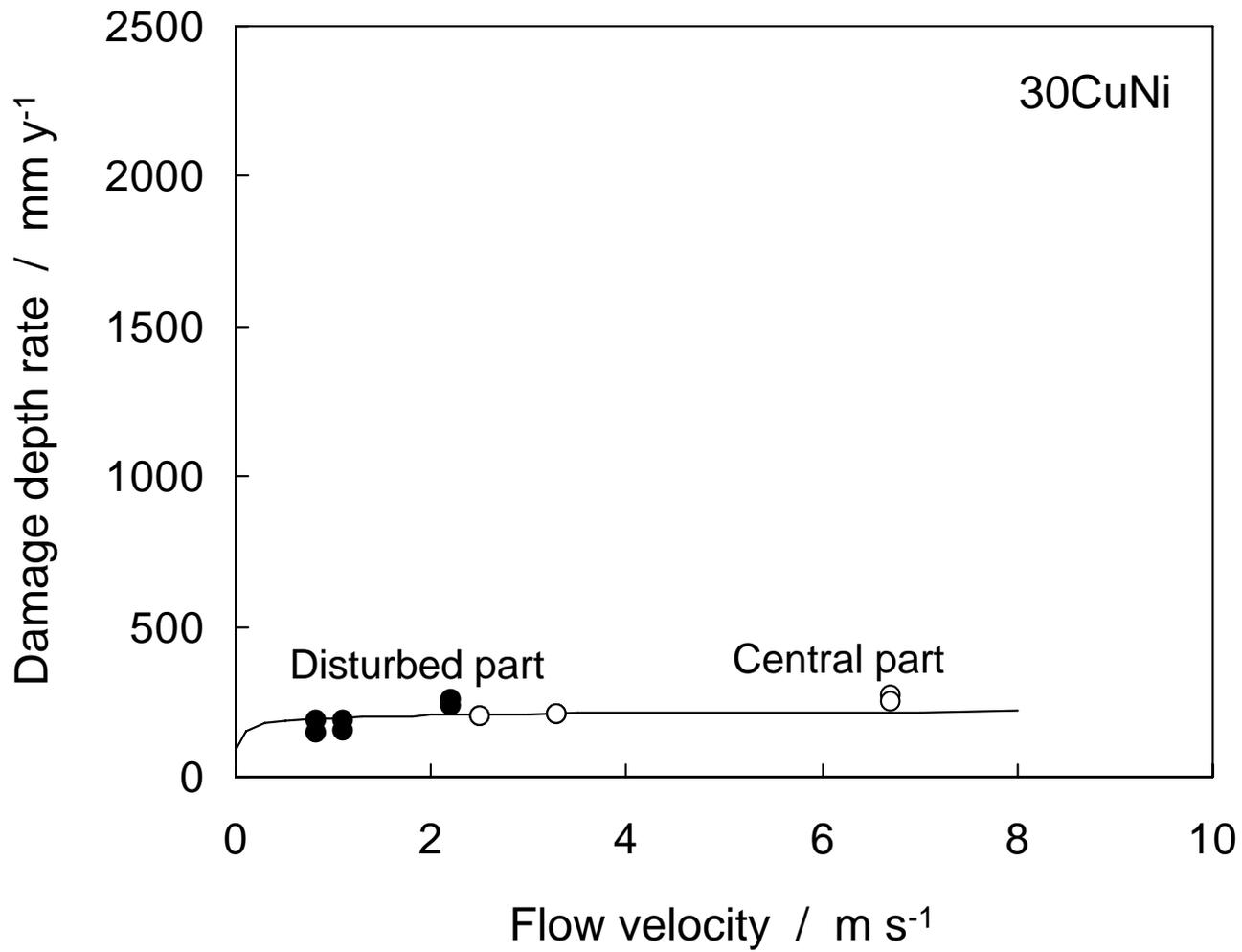
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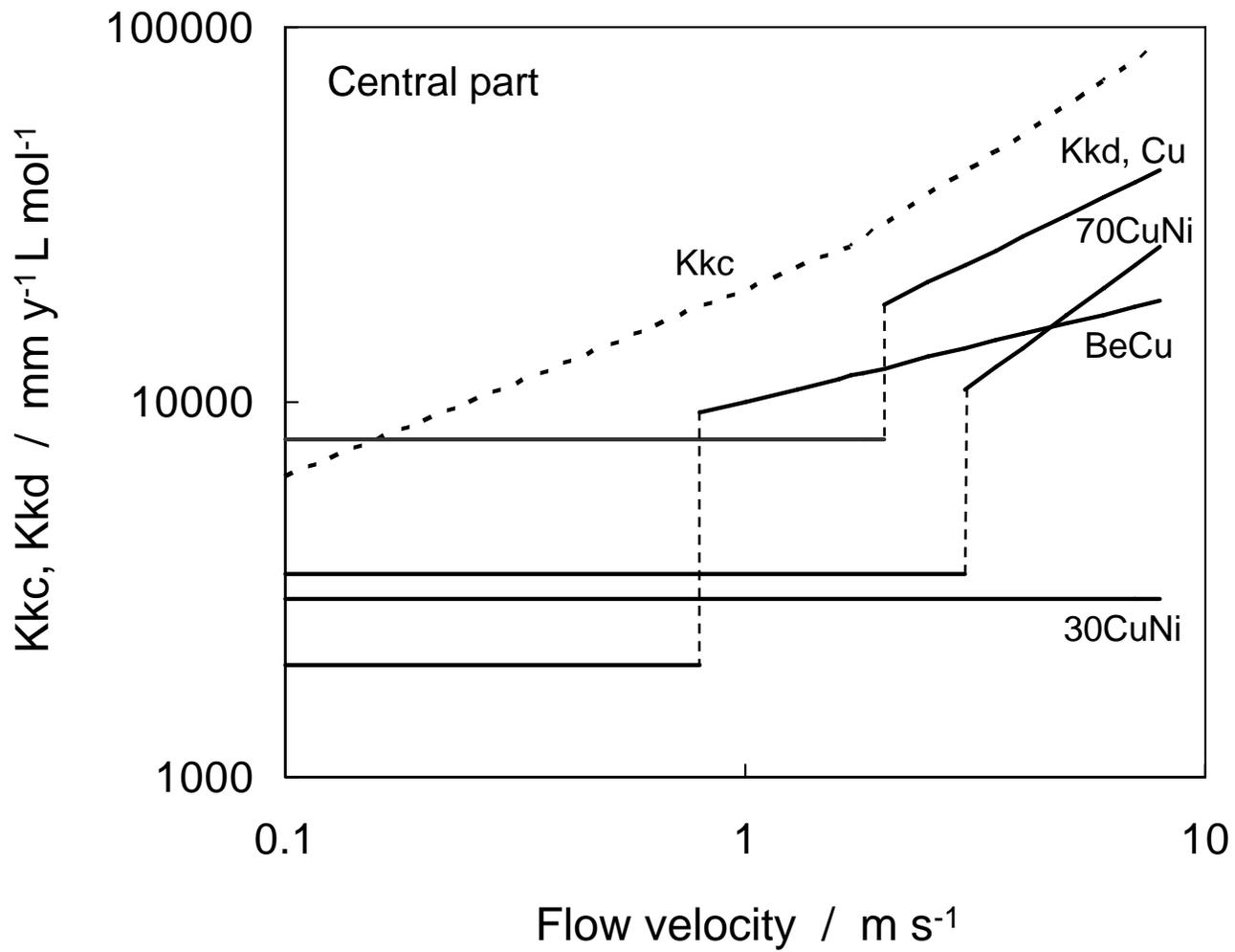
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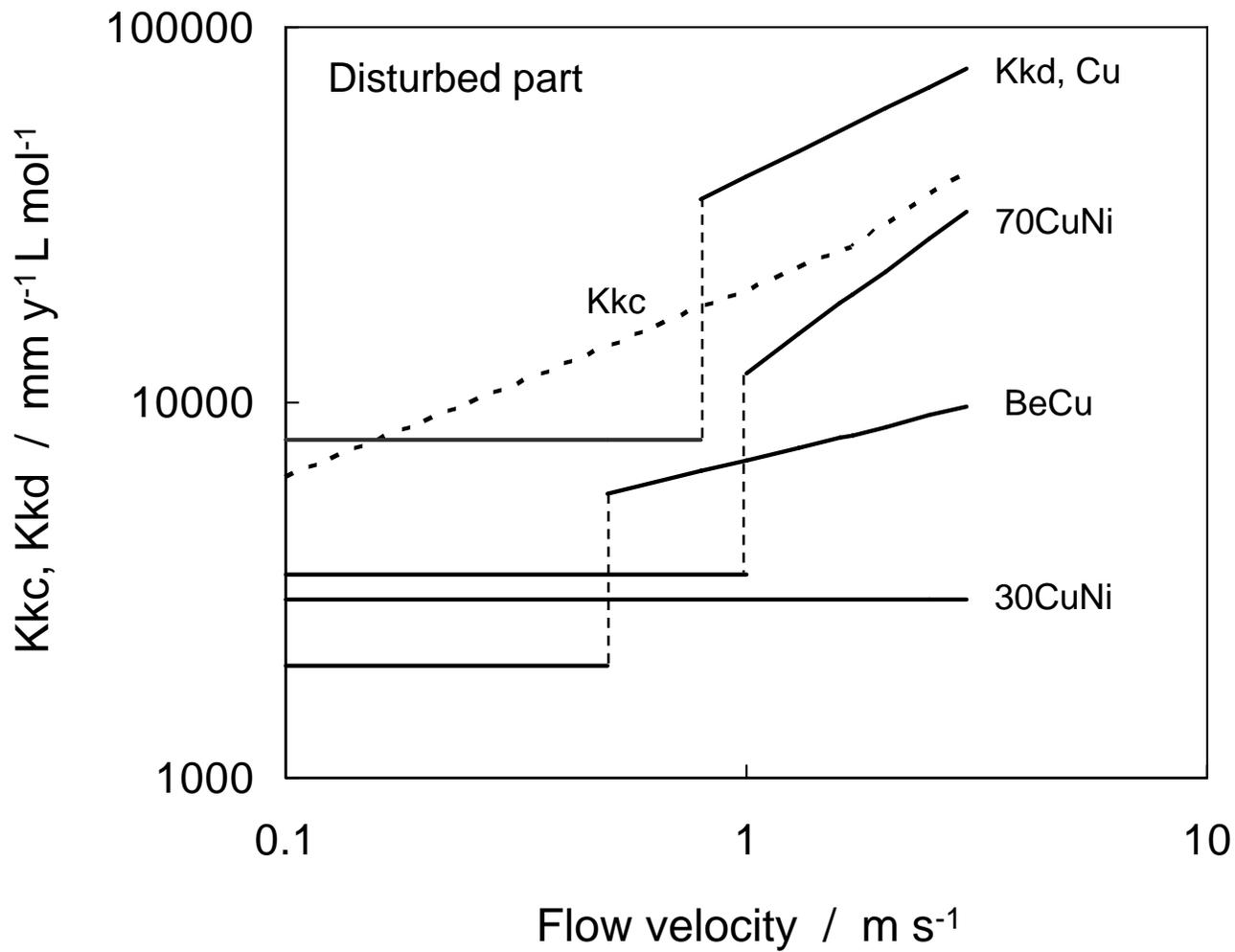
Fig. 10. Akihiro Yabuki et. al.

Table 1. Akihiro Yabuki et. al.

Symbol	Primary chemical composition / mass%
Cu	99.99Cu
70CuNi	30.2Ni-Cu
30CuNi	31.6Cu-Ni
BeCu	1.85Be-Cu

Table 2. Akihiro Yabuki et. al.

Symbol	Part	V_b / m s^{-1}	α	β	n
Cu	Central	2	8000	12000	0.6
	Disturbed	0.8		40000	
BeCu	Central	0.8	2000	10000	0.3
	Disturbed	0.5		7000	
70CuNi	Central	3	3500	4000	0.9
	Disturbed	1		12000	
30CuNi	Central	-	3000	-	-
	Disturbed	-		-	-