Estimation of 2nd Phase Particle Size on Hot Crack of High Cr-Ni Steel Weld Heat Affected Zone Caused by Constitutional Liquation.

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【ABSTRACT】

Hot crack of high Cr-Ni stainless steel weld heat affected zone (HAZ) caused by liquationis closely related with constitutional liquation of 2nd phase particles in base metal. If particle size can be reduced in such degree that 2nd phase particles are decomposed during weld thermal cycle due to welding before constitutional liquation occurs and diffused into base metal matrix and are turned to solid solition, hot crack sensitivity of HAZ may be immunized. In the prsent study, we obtained critical particle size of such 2nd phase particles using the solution of the case where the particles were brought into contact with semi-infinite solid (base metal) according to the Second Low of Fick in diffusion, and HAZ hot crack sensitivity estimated from the theoretical critical particle size determined by this method was compared with the value obtained by hot ductility test. As a result, very good correlation was found between these two values, and it has become clear that constitutional liquation was a principal cause of HAZ hot crack in this type of material. At the same time, it was demonstrated that the estimation based on critical particle size of 2nd phase in the present study is practically suitable as a method to assess hot crack sensitivity caused by liquation of high Cr-Ni steel weld HAZ.

I. INTRODUCTION

In case base metal contains unstable compound phase (or precipitated phase) as 2nd phase at high temperature in high Cr-Ni steel having austenite structure, hot crack due to constitutional liquation of weld is very likely to occur on the weld heat affected zone(HAZ)[(1)~(4)], and high HAZ hot crack sensitivity is often shown together with weld thermal stress. In the past, hot crack sensitivity of HAZ has been evaluated by the extent of the brittle temperature range (BTR) in hot ductility test using a weld thermal cycle simulater, whereas the test procedure is complicated, and it is not very adequate method in case the material must be evaluated within short time such as in acceptnce test. On the other hand, the phenomenon of constitutional liquation can be avoided in HAZ hot crack caused by constitutional liquation of 2nd phase particles ("particles") are decomposed during thermal cycle of welding before constitutional liquation occurs and is diffused and turned to solid solution in the base metal matrix, and this may lead to the possibility that immunity could be given to HAZ hot crack based on this phenomenon. B. Weiss et al.[(5)] already reported on the estimation of particle size, but the particle size determined by the method of Weiss et al. does not necessarily agree with the results of HAZ hot

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crack test such as hot ductility test. In this connection, evaluation has been made in the present study on the method of Weiss et al., in particular on the method to obtain diffusion length, and a modified equation to estimate critical particle size has been proposed. At the same time, hot ductility test was performed on 21%Cr - 32%Ni steel including 3 series of materials, to which Nb, Ti and B were separately added, and the results were compared with critical particle size determined by the estimation method, and the validity of the present method was evaluated.

II. EXPERIMENTAL PROCEDURE

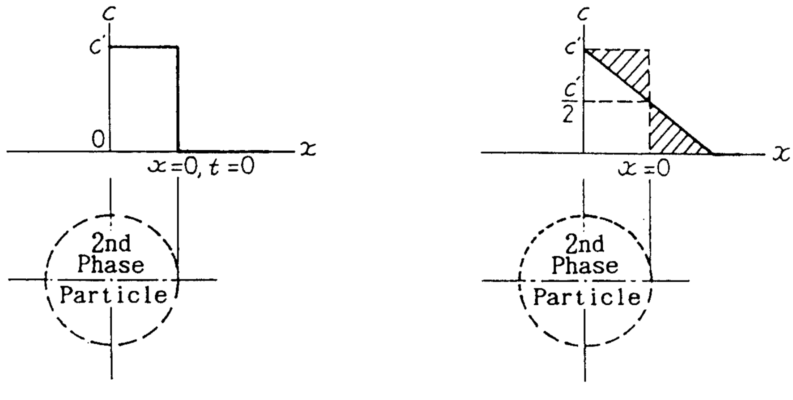
2-1 Determination of Critical Particle Size showing no Constitutional Liquation

The critical particle size showing no constitutional liquation can be basically obtained by a method similar to that of Weiss et al. Specifically, if it is assumed that a particle is of spherical shape and concentration of component element is C', the solution of the Second low of Fick can be expressed by the equation (1) by introducing error integration of Gauss when boundary condition at the interface between particle and matrix phase is $t = 0$ and $X < 0$ and $C = C'$, or it is $t = 0$ and $X > 0$ and $C = 0$.

$$C_{(x,t)} = C'/2[1 - \mathrm{erf}(X/2\sqrt{Dt})] \qquad (1)$$

Here, C is solute concentration at a distance "X" from the interface between particle and matrix. D is a diffusion coefficient and t is diffusion time. If it is assumed that, in order to obtain critical particle size, interface concentration when $X = 0$ is always based on $C'/2 = C_0$ as boundary condition when concentration C' of component element of the particleis linearly diffused and decreased from the center of the particle on the matrix as shownin Figure 1 and the particle is turned to solid solution, the equation (1) can be expressed by the equation (2).

$$C_{(x,t)}/C_0 = 1 - \mathrm{erf}(X/2\sqrt{Dt}) \qquad (2)$$



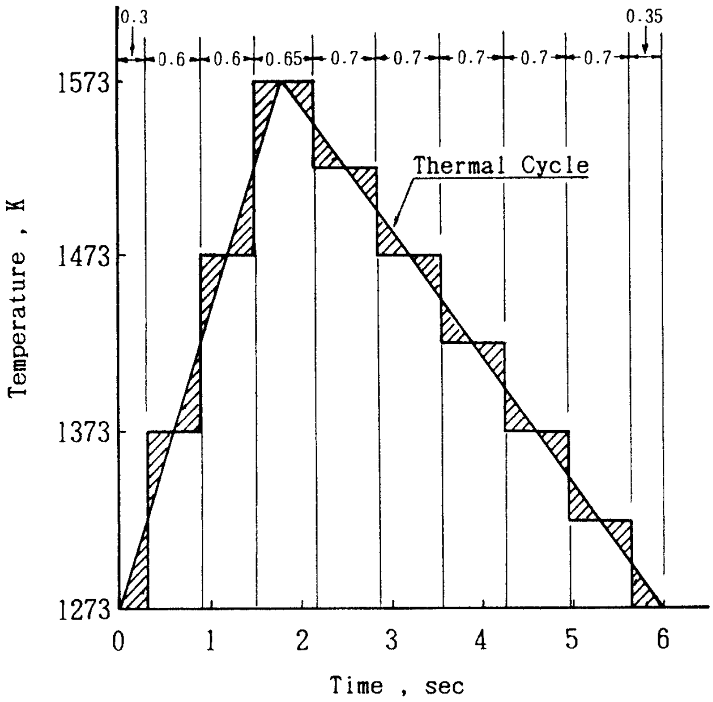
(a) Initial and boundary condition (b) The particle has just dissolved

Figure 1 Diffusion model of perticle approximated for determination of total diffusion length "X".

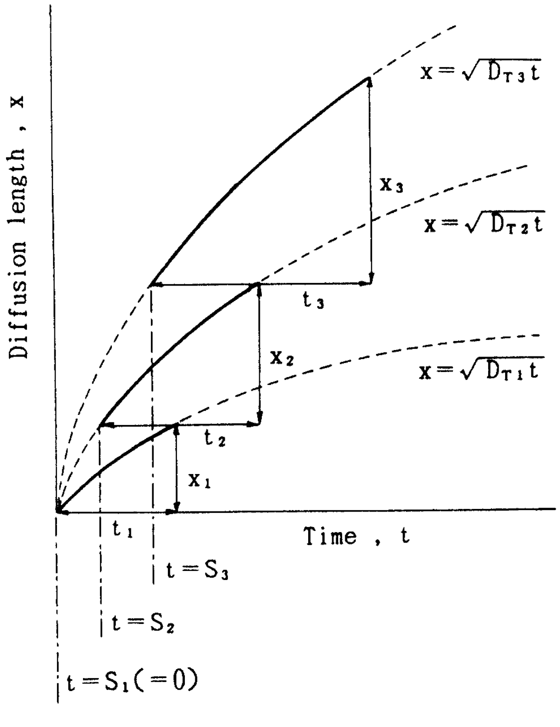
When the value of $X/2\sqrt{Dt}$ corresponding to the value $C_{(x,t)}/C_0 = 1/2$ is obtained from the error function table of Gauss, diffusion length "X" can be approximately expressed by the equation (3) because $X/2\sqrt{Dt} \doteqdot 1/2$.

$$X \doteqdot \sqrt{Dt} \qquad (3)$$

On the other hand, diffusion coefficient D_T at the temperature T can be given as : $D_T = D_0 \exp(-Q/RT)$ where D_0 is frequency factor, R is gas constant, T is temperature and Q is activation energy of diffusion. If the weld heat cycle measured for the present study is approximated by step function as shown in Figure 2, total diffusion length X_{tot} in the thermal cycle is a sum of diffusion length $X_i = \sqrt{D_{Ti} t_i}$ at each step. However, in the diffusion length X_i at each temperature, as shown in Figure 3, when the



-Figure 2-
Illustration of the step function method for determining the total diffusion length "X" for the weld thermal cycle.



-Figure 3-
Introduction of the time compensation term "S" for diffusion during weld thermal cycle.

temperature T is changed from i-th step to "i+1"-th step, the diffusion length X_{i+1} at the temperature T_{i+1} should be calculated by subtracting a time segment "S_i", during which the same diffusion lengt has the diffusion X_i at the temperature T_i is reached at the temperature T_{i+1} ("time compensation term"), from the step time at the temperature T_{i+1}. In other words, if it is supposed that diffusion length at each step is X_1, X_2, X_3,……, X_i, the diffusion length approximated by step function is as given by the equation (4) (where $S_1 = 0$),and total diffusion length X_{tot} is expressed by the equation (5).

$$\left.\begin{aligned} X_1 &= \sqrt{D_{T1}(S_1+t_1)} - \sqrt{D_{T1}\cdot S_1} \\ X_2 &= \sqrt{D_{T2}(S_2+t_2)} - \sqrt{D_{T2}\cdot S_2} \\ X_3 &= \sqrt{D_{T3}(S_3+t_3)} - \sqrt{D_{T3}\cdot S_3} \\ &\cdot\quad\cdot\ \cdot\ \cdot\quad\ \cdot\ \cdot \\ &\cdot\quad\cdot\ \cdot\ \cdot\quad\ \cdot\ \cdot \\ X_i &= \sqrt{D_{Ti}(S_i+t_i)} - \sqrt{D_{Ti}\cdot S_i} \end{aligned}\right\} \qquad (4)$$

$$X_{tot} = \Sigma_i X_i = \Sigma_i\{\sqrt{D_{Ti}(S_i+t_i)} - \sqrt{D_{Ti}\cdot S_i}\} \qquad (5)$$

In this case, the compensation term S_i is given by the equation (7) from the equations (4),(5) and (6).

$$\left.\begin{aligned} \sqrt{S_{T2}\cdot S_2} &= \sqrt{D_{T1}(S_1+t_1)} \\ \sqrt{D_{T3}\cdot S_3} &= \sqrt{D_{T2}(S_2+t_2)} \\ \sqrt{D_{T4}\cdot S_4} &= \sqrt{D_{T3}(S_3+t_3)} \\ \cdot\ \ \cdot\quad &\quad\ \cdot\ \ \cdot\ \ \cdot \\ \cdot\ \ \cdot\quad &\quad\ \cdot\ \ \cdot\ \ \cdot \\ \sqrt{D_{Ti}\cdot S_i} &= \sqrt{D_{Ti-1}(S_{i-1}+T_{i-1})} \end{aligned}\right\} \qquad (6)$$

$$S_i = \{D_{Ti-1}(S_{i-1}+T_{i-1})\}/D_{Ti} \qquad (7)$$

Therefore, the critical particle size d_c can be obtained by the equation (8) from the equation (5).

$$d_c = 2\Sigma_i X_i \qquad (8)$$

2-2 Correlation between Critical Particle Size showing no Liquation and the Results of Hot Ductility Test

To determine correlation between theoretical size d_c as described in 2-1 above and actual hot crack caused by liquation,the test materials were prepared, which have 21%Cr-32%Ni as basic composition and to which Nb, Ti and B are added by 2.87%, 1.94% and 0.290% at maximum respectively. These materials were melted in a high frequency furnace (including products melted by Heroult furnace and ESR furnace), and these were hot rolled to round bars each 10 mm in diameter, and hot ductility test was performed. The hot ductility test was carried out with heating and cooling process of measured thermal cycles as shown in Figure 2, and BTR of each test material was obtained.

Chemical compositions of these test materials are summarized in Table 1-1, 1-2 and 1-3. Further, mean particle size in these test materials was obtained from SEM image, and these particles were identified by electron diffraction pattern of extracted replica.

On the other hand, based on the results of identification of the extracted particles found in the test materials from the equation (8), critical particle size was obtained as diffusivity in the atom of the particle with slower diffusion speed from the diffusion data[6], and the relationship between mean particle size of 2nd phase particle in the base metal and BTR obtained from hot ductility test was assessed.

Table 1-1 Chemical composition of Nb bearing 21%Cr-32%Ni steel tested.

Steel No.	Aim of Nb	C	Si	Mn	P	S	Cr	Ni	Nb
1	—	0.058	0.43	0.76	0.011	0.005	20.89	32.04	—
2	0.05	0.045	0.45	0.72	0.010	0.005	20.93	32.27	0.06
3	0.10	0.048	0.46	0.78	0.011	0.008	20.87	31.84	0.12
4	0.30	0.050	0.48	0.74	0.012	0.008	20.91	32.48	0.33
5	0.50	0.049	0.46	0.76	0.012	0.008	20.91	32.34	0.58
6	1.00	0.050	0.46	0.72	0.014	0.008	20.64	32.62	1.09
7	2.00	0.052	0.56	0.77	0.011	0.009	20.57	32.04	2.18
8	3.00	0.055	0.40	0.66	0.009	0.013	20.61	31.90	2.87

Table 1-2 Chemical composition of Ti bearing 21%Cr-32%Ni steel tested.

Steel No.	Aim of Ti	C	Si	Mn	P	S	Cr	Ni	Ti
1	—	0.058	0.43	0.76	0.011	0.005	20.89	32.04	—
2	0.1	0.054	0.37	0.70	0.013	0.011	21.09	31.86	0.06
3	0.3	0.045	0.42	0.74	0.010	0.014	21.08	31.96	0.26
4	0.6	0.054	0.41	0.77	0.011	0.012	21.05	31.96	0.61
5	1.0	0.049	0.44	0.75	0.010	0.007	21.32	32.34	0.92
6	1.2	0.049	0.45	0.70	0.009	0.077	21.14	31.91	1.07
7	1.5	0.050	0.50	0.74	0.010	0.077	21.02	31.85	1.43
8	1.7	0.051	0.49	0.74	0.010	0.077	20.76	30.44	1.63
9	2.0	0.055	0.39	0.66	0.012	0.006	20.92	31.66	1.94

Table 1-3 Chemical composition of B bearing 21%Cr-32%Ni steel tested.

Steel No.	Aim of B	C	Si	Mn	P	S	Cr	Ni	B
1	—	0.058	0.43	0.76	0.011	0.005	20.89	32.04	—
2	0.003	0.055	0.41	0.74	0.006	0.014	20.84	31.96	0.003
3	0.006	0.051	0.42	0.74	0.005	0.014	20.59	32.10	0.006
4	0.009	0.054	0.41	0.74	0.005	0.014	20.84	32.10	0.009
5	0.050	0.045	0.33	0.58	0.006	0.005	20.86	32.10	0.060
6	0.300	0.056	0.39	0.75	0.007	0.014	20.64	31.82	0.290

Ⅲ EXPERIMENTAL RESULTS

Table 2-1, 2-2 and 2-3 show the results of identification of particles found in the test materials respectively. As it is evident from these tables, the main phase among the 2nd phase particle is NbC in Nb bearing material, TiC in Ti bearing material, and Cr_2B in B bearing material. From these results, critical particle size of each particle was obtained using the equation (8) with Nb, Ti and B as diffusivity element.

The results are given in Table 3. Critical particle size obtained by the method of Weiss et al. is also given in Table 3 for reference.

Table 2-1 Electron Diffraction Data from Extraction Replica of 21%Cr - 32%Ni Steel Containing 1.09%Nb.

d_{obs}	I_{obs} +)	ASTM X-Ray Data, Å							
		NbC		Nb_2C		Ni_3Nb		Cr_7C_3	
		d	I/I_0	d	I/I_0	d	I/I_0	d	I/I_0
2.58	S	2.58	100	-	-	-	-	-	-
2.22	M	2.23	80	-	-	-	-	-	-
2.13	W	-	-	-	-	2.124	100	2.176	100
2.02	W	-	-	-	-	2.001	75	2.052	100
1.97	W	-	-	-	-	1.975	75		
1.81	W	-	-	1.811	60	-	-	1.8016	22
1.57	W	1.58	50	1.551	60	-	-	-	-
1.35	W	1.348	65	1.344	15	-	-	-	-
1.28	W	1.280	13	1.299	60	-	-	-	-
1.03	W	1.025	25	1.043	55	-	-	-	-
		0.999	30	1.018	20	-	-	-	-
0.91	W	0.9124	26	0.928	60	-	-	-	-
0.84	W	0.8603	27	0.866	80	-	-	-	-

+) S : Strong, M : Medium, W : Weak

Table 2-2 Electron Diffraction Data from Extraction Replica of 21%Cr - 32%Ni Steel Containing 1.2%Ti.

d_{obs} Å	ASTM X-Ray Data, Å			
	TiC		Fe_2Ti	
	d	I/I_0	d	I/I_0
2.51	2.51	80	-	-
2.22	-	-	2.20	75
2.16	2.18	100	-	-
1.97	-	-	2.00	100
			1.96	50
1.73	-	-	1.83	50
1.52	1.53	50	-	-
1.38	-	-	1.38	10
1.33	1.31	30	1.34	75
			1.30	85
1.24	1.25	10	-	-
1.21	-	-	1.22	60

Table 2-3 Electron Diffraction Data from Extraction Replica of 21%Cr - 32%Ni Steel Containing 0.060%B.

d_{obs}	I_{obs} +)	ASTM X-Ray Data, Å					
		Fe_2B		Cr_2B		Ni_4B_3	
		d	I/I_0	d	I/I_0	d	I/I_0
2.97	W	-	-	2.947	14	2.99	60
2.54	W	2.554	30	-	-	2.53	60
2.46	W	-	-	-	-	2.471	80
2.31	W	-	-	2.300	57	-	-
2.25	W	-	-	-	-	2.249	80
2.10	M	2.124	40	2.1137	55	2.110	60
						2.070	100
2.00	S	2.011	100	2.0416	100	2.008	80
1.92	W	-	-	-	-	1.931	80
						1.918	80
1.90	W	-	-	-	-	1.906	80
1.83	W	-	-	1.8267	38	-	-
1.70	W	-	-	-	-	1.702	60
						1.691	41
1.65	W	1.634	18	1.6543	19	1.652	60
				1.6480	26		

+) S : Strong, M : Medium, W : Weak

Table 3 Critical Diameter of 2nd Phase Particles, obtained by Introduction of the Compensation Term "S" and Weiss's Equation.

Intermetallic Compound to form 2nd phase	Element dominating diffusion	D_0 (m^2/s)	Q (kJ/mol)	$d_{crit.}$ calc. by compen'n term"S"(μm)	$d_{crit.}$ calc. by Weiss's equa'n(μm)
NbC	Nb	5.6×10^{-4}	286	1.84	3.97
TiC	Ti	1.5×10^{-6}	251	1.15	2.67
Ni_4B_3	Ni	1.08×10^{-6}	273	0.61	0.84
$Cr_{23}C_6$	Cr	1.69×10^{-5}	264	0.99	1.75
Fe_2B	Fe	8.9×10^{-5}	291	0.77	1.36
Cr_2B	Cr	1.69×10^{-5}	264	0.99	1.75
Ni_3Nb	Ni	1.08×10^{-6}	273	0.61	0.84
Cr_3P	Cr	1.69×10^{-5}	264	0.99	1.75

As it is apparent from Table 3, the critical particle size obtained from the equation (8) is considerably smaller compared with that of the method by Weiss et al. On the other hand, Figure 4-1, 4-2 and 4-3 each represents relationship between BTR from hot ductility test on commercial products melted in Heroult furnace or ESR furnace and mean particle size of 2nd phase particles found in these test materials in addition to the materials given in Table 1. When the critical particle size shown in Table 3 are entered in Figure 4 together with the results obtained by the equation of Weiss etal.. the results are as shown by solid line (equation (8)) and one-dot chain line (equation of Weiss). As given in Figure 4, BTR of the test material having a particle size smaller than the critical particle size obtained from the equation (8) is within the range

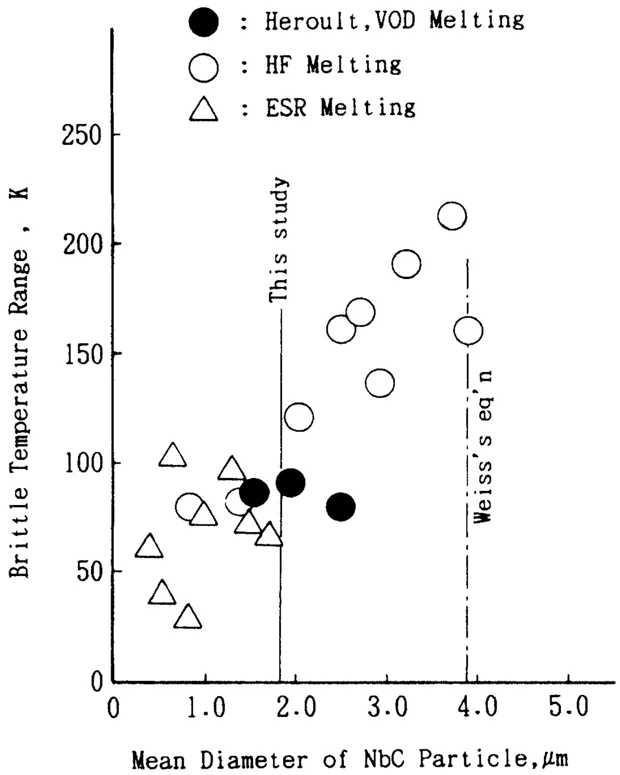


Figure 4-1 Effect of NbC Particle Size on the Brittle Temperature Range of Nb bearing Cr-Ni Steel.

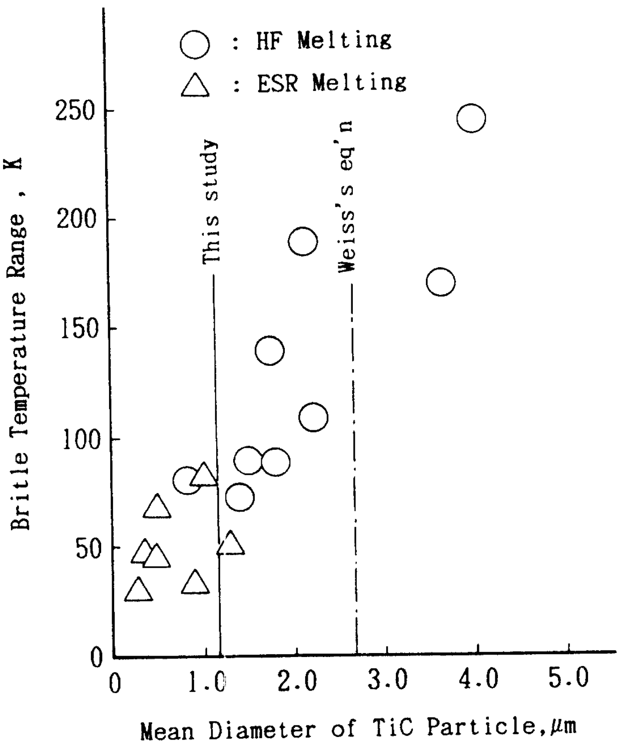


Figure 4-2 Effect of TiC Particle Size on the Brittle Temperature Range of Ti bearing Cr-Ni Steel.

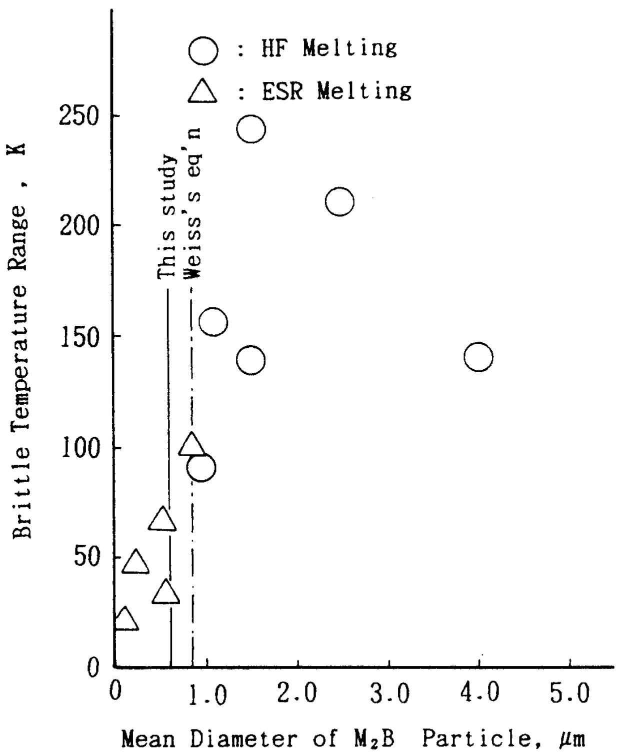


Figure 4-3 Effect of M_2B Type Particle Size on the Brittle Temperature Range of B bearing High Cr-Ni Steel.

of not higher than 100℃ [7] where HAZ hot crack sensitivity is negligible, and the critical particle size from the equation (8) agrees well with the results of hot ductility test. In contrast, the particle size according to the equation of Weiss et al. was considerably higher than the value estimated from the hot ductility test, and this appears to be insufficient to estimate HAZ hot crack of the test materials.

Ⅳ DISCUSSION

The result described in Ⅲ above reveal that the phenomenon of constitutional liquation of 2nd phase particles present in the base metal is a predominant factor in hot crack of high Cr-Ni steel weld heat affected zone. At the same time, it may be suggested that the critical particle size showing constitutional liquation can be estimated from the equation (8) and that the equation (8) induced from the Second Low of Fick is effective as an equation to estimate critical particle size. However, the above results are the results on Incoloy ally, which has 21%Cr - 32%Ni as nominal composition, and it is not certain whether it is applicable to high Cr-Ni steel other than the above composition or not. Under such circumstances, hot ductility test similer to that of Incoloy alloy and measurement of mean particle size of 2nd phase particle (NbC) were performed on URANUS 65 (Heroult and ESR melted products with nominal composition of 25%Cr - 20%Ni - Nb), which is being used for reprocessing of nuclear reactor fuel at prsent, and hot crack sensitivity was evaluated. Table 4 shows chemical composition of URANUS 65 under study, mean particle size of 2nd phase particle (NbC) and BTR obtained by hot ductility test. As it is evident from Table 4, mean particle size of NbC was 1.12μm(2.0μm max.) in Heroult furnace melted product, while it was 0.68μm(0.8μm max.) in ESR melted products, and these are all less than the critical particle size (1.84 μm). Also,BTR of these products are lower than 100℃,and it seems that occurrence of HAZ hot crack is negligible. In paticular, ESR melted products have smaller mean particle size, and this may be favorable melting condition from viewpoint of resistance to HAZ hot crack sensitivity. In fact,URANUS 65 materials are used as a dissolving tank for reprocessing system and appear to be suitable for practical application.

Table 4 Chemical Composition, BTR, Mean Diameter of NbC Particle of URANUS 65 for Reprocessing of Nuclear Reactor Fuel.

Melting	Chemical Composition, (Wt%)								BTR (°C)	Mean Diameter of NbC Particle(μm)
	C	Si	Mn	P	S	Cr	Ni	Nb		
Heroult	0.009	0.26	0.69	0.007	0.001	25.15	20.30	0.24	103	1.12 (max. 2.0)
E S R	0.012	0.18	0.68	0.008	0.001	24.61	20.97	0.22	68	0.68 (max. 0.8)

V CONCLUSION

Hot crack of high Cr-Ni steel weld heat affected zone was identified as a phenomenon caused by constitutional liquation of 2nd phase particles present in base metal, and a method has been proposed to obtain critical particle size decomposing and diffusing before constitutional liquation occurs in 2nd phase particles by using the solution of the Second Low of Fick. Using Incoloy alloy as test material, critical particle size was compared with BTR of hot ductility test. As a result, it was found that BTR where hot crack of HAZ could be avoided was correlated well with critical particle size, and that hot crack sensitivity of high Cr-Ni steel HAZ could be estimated from the critical particle size obtained by the method proposed in the present study. The present estimation method is also applicable to high Cr-Ni steel other than Incoloy alloy.

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