Elimination of Nitrogen Dissolved in Iron by Addition of Aluminium

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Synopsis

When nitrogen gas is reacted on melts of binary Fe-Al alloys under a pressure of one atmosphere, a part of the nitrogen enters the melts, (1) as non-metallic inclusions in the form of AlN compound, while the other part becomes (2) interstitially dissolved nitrogen in its atomic state. We measured the quantity of nitrogen in the two above states separately with respect to samples of Fe-Al alloys of various compositions. In particular, we investigated the relation between the quantity of the interstitially dissolved nitrogen and the aluminium content of the various samples, and arrived at important results suggesting a basic relation in the elimination of nitrogen in iron melt at variance with the theory accepted hitherto. A statistico-thermodynamical study of the results gave satisfactory answers.

I. Introduction

According to the generally accepted theory,[1] iron melts will be denitrified by addition of aluminium, or any other elements with strong affinity with nitrogen, such as Ti and Zr, in much the same way as they are deoxidized, as follows:

$$[N]^{**} + [Al] \rightarrow AlN \text{ (solid at 1,600°C)}.$$

In which the product [Al] [N] is a constant at a constant temperature, according to the law of mass action, so that a larger quantity of the aluminium added will result in better elimination of nitrogen. But, upon scrutiny of the results of the experiments on the nitrogen absorption of Fe-Al alloys conducted at our laboratory, it was found that the denitrification could not be explained fully by means of such a simple theory alone. We will hereunder report the statistico-thermodynamical study on the results of the denitrifying reaction, obtained by way of separate quantitative analysis of the nitrogen content in molten Fe-Al alloys partly in the form of AlN compounds (hereinunder referred to as AlN nitrogen, for brevity's sake) and partly solved interstitially in the quasi-crystal melt (hereinunder referred to as dissolved or interstitial nitrogen). This report forms a sequel to the series of the studies on the absorption of nitrogen by molten binary iron alloys conducted by one of the authers[2] of this report in recent years.

II. Methods of Experiments

1. Experiments on the Absorption of Nitrogen Gas

Using the same apparatus and procedures as described in the previous report, samples of molten Fe-Al alloys of various compositions were made to absorb pure nitrogen at a fixed temperature, and the equilibrium relation of the ternary system Fe-Al-N was studied. Some results of these experiments are shown in Fig. 1. As indicated in Fig. 1, the curves show that the quantity of formed AlN increases with time and with the rise in percentage of aluminium, so that no definite saturation value can be deduced from these curves. So, we proceeded to measure the quantity of absorbed nitrogen from the sum total of the gas evolved during the solidification of the melts and the values of nitrogen content obtained by analysis of the cooled solid, using the Method II described in a former report. When the result of such measurements is plotted against the percentage of aluminium content, the plotted points are scattered irregularly, as shown in Fig. 2 (points • and -•-), so that no definite correlation of the two quantities is evidenced. However, from the fact that solid
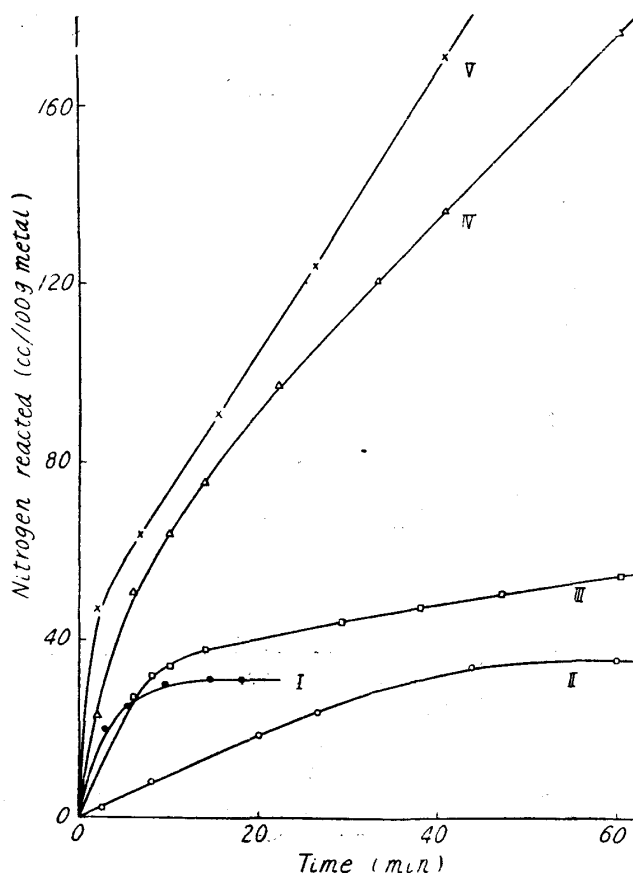

Fig. 1.   Reaction curves of molten Fe-Al alloys.
I : Electrolytic iron, 1560°   II : 1.2% Al, 1630°
III : 7.2% Al, 1670°   IV : 19.9% Al, 1650°
V : 38.1% Al, 1490°

samples containing higher content of non-metalic inclusions under microscope show higher values of nitrogen content upon analysis, as may be seen by comparing the points marked • and -•- in Fig. 2, the reason of the apparent irregularity of the plotting may be inferred to be as follows. As past literatures[3] show, the melting point of AlN is higher than 2,000°C, though authorities vary upon the exact value, and its dissociation pressure attains one atmospheric pressure at around 2,000°C, so it is a stable solid within the temperature range used in the experiments. The specific gravity of AlN is about

(3) Gmelin's Handbuch d. Anorg. Chem., 8 Aufl. (1934), system No. 35, Teil B, 132.

3.03 and much smaller than that of molten iron, but as it exists in the melt in the temperature prevailing during the experiments in the form of minute particles of solid, the particles hardly coagulate, and are thus only imperfectly separated and floated out of the iron melt, the larger part remaining suspended in the melt and being thus retained in solid sample made thereof in an undefinite quantity. As the result, the quantity of the total nitrogen content fails to show a definite value against varying percentage of aluminium content.

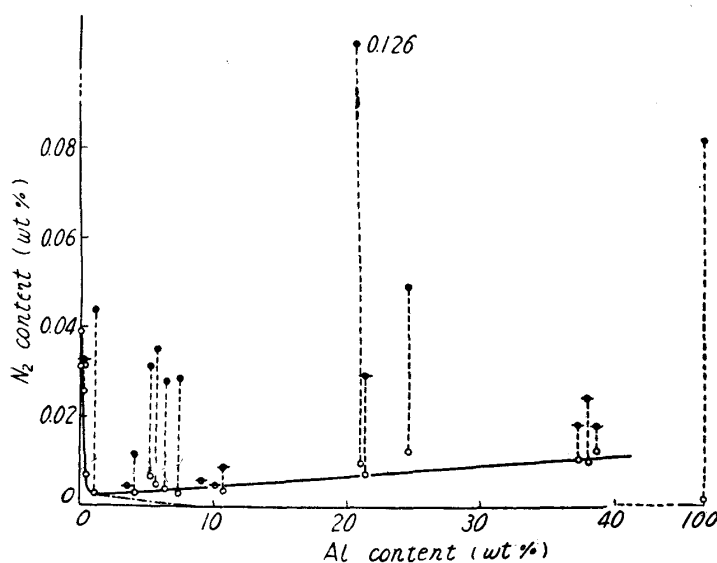In order to ascertain the validity of such an assumption, it becomes necessary to analyse the samples for the respective quantities of the nitrogen solved interstitially in the quasi-crystal of the iron melt and the nitrogen contained in the form of AlN paricles retained in the solidified sample. As shown below, there exists a theoretically explicable fixed relation between the dissolved nitrogen and the aluminium percentage in the sample, but the quantity of the AlN particles retained in the sample could not be controled in these experiments.

Fig. 2. Relation between total nitrogen, interstitial nitrogen, AlN nitrogen vs. aluminium content of Fe-Al Alloys.

• Total nitrogen content
○ Interstitial nitrogen content
Samples considerably clean under microscope are designated by -•- or -○-
•----○ Content of AlN nitrogen
————— Curve which corresponds to the equation
[Al] [N] = const.

In the present report, Kjeldahl's method, as prescribed by the Foundation for Promotion of Science and Industry of Japan, was used in quantitative analysis for the determination of the total quantity of nitrogen in the samples, and an ester-halogen method for analytic determination of the net quantity of the AlN nitrogen. The difference of these two quantities were used to determine the quantity of the nitrogen solved interstitially in the quasi-crystal of the molten iron, and the result was studied statistico-thermodynamically as described below.

2. Study of Methods of Determining AlN Content

Beeghly's method[4] was adopted in analysing the AlN nitrogen in the samples. In this method, the sample is dissolved in a solvent which readily reacts upon the iron matrix, but does not affect AlN, which is separated from the solution

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(4) H. F. Beeghly, An. Chem., 21 (1949), 1513.

as residue and quantitatively analysed according to micro-Kjeldahl's method. Eeeghly, upon testing several ester-halogen solvents filling the above conditions, recommends the use of bromine-methylacetate. In our tests, this solvent was used, as well as an apparatus similar to that used by Beeghly. The process is hereinunder referred to as ester-halogen method. Prior to tests, we first proceeded to conduct preliminary tests to ascertain, a), the utility and accuracy of this method and b), whether the Kjeldahl's method we commonly use is efficient enough in determining the total content of nitrogen (AlN nitrogen plus dissolved nitrogen), that is, in completely analysing the AlN retained in the sample, using specially prepared pure AlN powder. The aluminium powder used in the preparation of the AlN sample contained the following impurities upon analysis:

Table 1. Result of chemical analysis of aluminium powder.

SiO_2 (wt.%)	Al_2O_3 (wt.%)	Fe (wt.%)
1.67	0.31	0.36

where the entire quantity of Si present in the powder is listed as existing in the form of SiO_2.

This powder was put in a porcelain boat and a flow of nitrogen gas purified by passing through a net of reduced copper, ammoniacopper solution, soda lime, calcium chloride and phosphorus pentoxide was passed thereover, and the temperture of 1,100°C was maintained for about 2 hours, to prepare the AlN powder. As it is possible that the SiO_2 in the porcelain might oxidize the aluminium powder under the reaction:

$$3SiO_2 + 4Al \rightarrow 2Al_2O_3 + 3Si , \qquad (1)$$

the porcelain boat was filled with aluminium powder and heated at a temperature of 1,100°C for a long time before the testing, to prevent the reaction (1) as far as possible.

The data obtained upon checking the ester-halogen method with AlN powder prepared thus are given in Table 2.

The column II in the Table shows the weight increment of the sample upon completion of the above described nitriding treatment, which values are about 1% higher than the weight percent of the total nitrogen content determined by Kjeldahl's method given in column III (Confer the percentage of oxidation increment given in column IV). The oxidation by equation (1) above could not have been entirely excluded, though every care was taken to purify the nitrogen gas and the interior of the nitridizing furnace. As past literatures[5] show it, it was impossible to prevent oxidizing reactions occurring to this extent. The column V gives the percentage of Al_2O_3 equivalent to the percentage of oxidation given in column IV. Also, it must be noted that the sample AlN contain Al_2O_3 which was originally contained in the aluminium

(5) For example, P. Röntgen, H. Braun, Metallwirts., 11 (1932), 471.

powder used in the preparation and that which was formed by the reaction between the aluminium powder and oxides of Si and Fe existing in the aluminium powder. These impurities account for the existing of some Al_2O_3 generated in the AlN powder. The calculated total weight percent values of such Al_2O_3 content are given in VI, which are in tolerable agreement with the quantity determined by the analysis given in column VII, though the former are slightly smaller than the latter. As it has been known,[3] AlN is reacted upon by water to a certain extent, and is readily dissolved in a warm solution of NaOH, so it may be expected that AlN will almost completely decompose by distillation with NaOH in the Kjeldahl's method. This fact, coupled with the result of the collation of the value of the quantitative analysis of AlN plus Al_2O_3 and the increment in weight of the sample, was sufficient in the effectiveness of the Kjeldahl's method for analysing AlN nitrogen. As it is almost impossible to obtain chemically pure AlN, these data, though imperfect in minute details, must suffice for our purpose. The column VIII shows the results of Kjeldahl's analysis applied to the AlN samples treated in bromine-methylacetate solution for 90 min., a duration adequate for complete solution of

Table 2. Accuracy of the ester-halogen method (the analysing
method of AlN nitrogen in steel)
Sample: Synthetic AlN powder which contain Al_2O_3
as impurity.

I	II	III	IV	V	VI	VII	VIII	IX
1	28.46	27.72	0.74	1.57	4.11	9.73	27.26 27.21	98.34 98.16
2	27.50	26.12	1.38	2.93	5.47	6.92	25.30 25.43 25.55	96.86 97.36 97.82
3	24.97	23.47	1.50	3.19	5.73	7.51	22.69 22.90 22.94	96.68 97.57 97.74

mean　　97.57±0.37

Columns
　　I　:　No. of samples.
　　II　:　Increment of weight of sample by the nitriding treatment (wt.%).
　　III　:　Analytical value of total nitrogen content determined by Kjeldahl's method (wt.%).
　　IV　:　Increment by oxidation through nitriding treatment; the difference of II and III.
　　V　:　Calculated values of Al_2O_3 percent corresponding to the increment of oxygen in column IV.
　　VI　:　Total of Al_2O_3 percent in AlN sample estimated from
　　　　　a) Al_2O_3 content originally contained in the aluminium powder,
　　　　　b) Al_2O_3 which was formed from the aluminium powder and SiO_2, FeO, and
　　　　　c) oxygen that entered from the atmosphere during the nitriding treatment.
　　VII:　Analytical value of Al_2O_3 in AlN sample (wt.%);
　　　　　Collate with the estimated values in VI.
　　VIII:　Analytical values of nitrogen in AlN samples after treating them with bromine-methylacetate solvent; determined with the same procedure as in III.
　　IX　:　Values of VIII/III, by which the accuracy of ester-halogen method may be estimated.

steel samples. The differences of these values from the analytic values obtained on samples without prior bromine-methylacetate treatment are less than 3%, as shown in column IX.

By the above preliminary experiments, it was ascertained that the total nitrogen content can be measured by the Kjeldahl's method and the AlN compound retained in the sample by the ester-halogen method. The difference of the results of the analysis by the two methods can be taken as representing the quantity of dissolved nitrogen.

III. Results of Experiments

The results obtained by the experiments as described above were as follows:

A) The relation between the quantity of solved nitrogen and aluminium percentage in the Fe-Al melt

The results of the measurement of total nitrogen, obtained by separate quantitative analyses of dissolved (interstitially solved) nitrogen and AlN nitrogen respectively, are plotted and shown in Fig. 2., which indicates the existence of a fixed quantitative relation between the percentage of solved nitrogen (marked o) and that of the aluminium content in the sample. Namely, the dissolved nitrogen sharply decreases at the outset, when the percentage of aluminium content is increased, reaching a minimum at 1~5 wt.% of aluminium, but rises gradually with further increase of aluminium. A similar result has been reported by Wentrup and Hieber[6] of a test of deoxidation of iron melt with titanium, except that elimination of oxygen was the object here, instead of nitrogen. Their tests showed that the oxgen content decreased sharply till Ti reached 0.05 wt.% (0.006 wt.% O_2) and continued to decrease at a slower rate up to 1 wt.% Ti, but began to rise therafter, the equilibriuum of oxygen content in the range of 2.4~4 wt.% Ti being reached at 0.01 wt.%. They do not touch on a theoretical explanation of this phenomenon in their report. Takeuchi,[7] in his recent report, succeeded in deducing theoretically that similar relations should exist between [C] and [O], and [Si] and [O] in iron melts.

B) Relation between quantity of AlN nitrogen and aluminium percentage in the Fe-Al melt

The quantity of AlN particles retained in the specimen is indefinite, even in specimens of the same aluminium percentage, as shown in Fig. 2. There is also no apparent fixed relation of the quantity with aluminium content, in going over all specimens of varying aluminium contents. That is, alloys rich in aluminium are not necessarily high in AlN inclusion. But, specimens found rich in particles of non-metallic inclusion under microscope were found to contain a large quantity of AlN upon analyses. A part of the inclusions consists of Al_2O_3 particles, as evidenced in experiment II, but it is impossible to

(6) H. Wentrup, G. Hieber, Arch. Eisenhüttenwes., **13** (1939/40), 69.

(7) S. Takeuchi, J. Japan Inst. Metals (N. K. G.), **15** (1951), 24 ; 36.

distinguish AlN and Al_2O_3 under a microscope.

C) Absorption of nitrogen by pure aluminium

There have been many studies reported on the subject, e. g., by Czochralski,[8] Hessenbruch,[9] Claus,[10] Roentgen, Braun,[5] etc., but the data of all these studies are somewhat old, and we have no knowlege of such studies made more recently. Smithells[11] cited the results of experiments by Roentgen and Braun as follows:

   i) Absorption of nitrogen in solid aluminum is slight.

   ii) That in molten aluminium rises with temperature.

   iii) AlN is formed, which become unstable at high temperature.

As to the instability of AlN cited in iii), as has been touched upon in the foregoing, we believe that it is not applicable within the range of temperature up to 1,600~1,700°C. The stability is exemplified by the fact that the AlN nitrogen content does not appreciably decrease by the vacuum melting for degasification of the samples we always go through prior to the experiments. (The results of the tests on the subject are omitted). As to ii), we beileve that Smithells misinterpreted the data given by Roentgen and Braun. The latter have not sought for the effect of temperature on the saturation value of nitrogen in molten aluminium, but only the relation between the temperature and the quantity of nitrogen reacted upon aluminium in the case when molten aluminium is subject to reaction upon nitrogen under a fixed temperature (700 ~1,000°C) and one atmospheric pressure, for 3 hours. The results of the experiments on the absorption of nitrogen by pure aluminium, obtained by Roentgen and Braun, as well as by the authors of this report, may be summarized as follows:

   i) When heated to the vicinity of its melting point, aluminium speedily reacts with nitrogen and forms a film of AlN on its surface, which is nearly impervious to gas.

   ii) This formation of AlN is expedited by rise in temperature, so that the quantity of generated AlN sharply increases at around 900°C, but as the film is impervious to gas, it is difficult to turn the entire specimen into AlN. Accordingly, nitrogen is not dissolved in aluminium, strictly speaking, but only AlN is formed by chemical reaction between aluminium and nitrogen. Upon analysis by the ester-halogen method of the pure aluminium samples subjected to the absorption tests, all the nitrogen content is found in the form of AlN, as shown in Fig. 2, and nothing is left for interstitial solution. In short, when nitrogen is reacted upon molten aluminium under one atmospheric pressure, the reacting nitrogen will all combine into AlN, so that equilibrium is reached when the entire quantity of aluminium transforms into the compound AlN.

(8) J. Czochralski, Z. Metallk., 14 (1922), 278.

(9) W. Hessenbruch, ibid., 21 (1929), 54.

(10) W. Claus, ibid., 21 (1929), 268.

(11) C. J. Smithells, "Gases and Metals" (1937), 174, Chapman & Hall, London.

In this state, it is impossible to speak of nitrogen dissolved interstitially in the aluminium crystal lattice.

This fact may be theoretically derived from the phase rule as well. In this case, as the system consists of two components, the degree of freedom will be one if the number of phases is taken at three (i. e., nitrogen gas, AlN and molten aluminium containing dissolved nitrogen), but according to our experiments, where the pressure of nitrogen is fixed at one atmosphere and the temperature at about 1,600°C, only two phases, namely, AlN and nitrogen gas can coexist at such a high temperature. The above three phases can coexist at fixed temperature only under a certain vapor pressure, and when the vapor pressure is fixed, only under a certain temperature.

In view of the above results of experiments and discussion thereof, it has been ascertained that the statement by Smithells that "the saturation value of absorption of nitrogen in molten aluminium rises logarithmically with the rise in temperature" is erroneous.

IV. Discussion

The conclusions derived from the results of the experiments on denitrification of iron melt by aluminium as shown in Fig. 2 may be summarised as follows.

While the quantity of added aluminium is small, the relation $[Al][N]$ = constant holds good and the quantity of solved nitrogen is reduced with the increase of aluminium reaching a minimum at 1~5 wt.% aluminium, but a further increase of aluminium content accompanies an increase of dissolved nitrogen. Such a relation cannot be completely explained thermodynamically, by treating the ternary melt Fe-Al-N as an ideal solution and applying the law of mass action thereto. Only by applying statistico-thermodynamical consideration to the equilibrium between the ternary melt and AlN-slag, adequate clarification may be given for these results of the above experiments.

The authors have expressed our views as follows on several occasions:

i) Metals and alloys retain in their molten state a quasi-crystalline structure, and in an alloy melt AB, a gas atom α dissolved therein occupies an interstitial position in the lattice of AB quasi-crystal.

ii) The distribution of such gas atoms in the holes is not at random, but they are arranged in the holes around the metal atoms, according to the strength of the affinity with metal atoms.

The thermodynamical functions of the atom assembly arranged in such a manner may be introduced as shown in the equation hereunder, taking $\Delta\psi_\alpha \equiv \psi_{A\alpha} - \psi_{B\alpha}$ or $\kappa = \exp(-\Delta\psi_\alpha/z'kT)$ as parameter, where $\psi_{A\alpha}$ is the interaction energy between A and α-atoms, and $\psi_{B\alpha}$ is that between B and α-atoms respectively. The signs used in the following have similar meanings as in our previous reports.

The free energy G^m of such an atom assembly $(AB\alpha)$ is derived as follows: (Please refer to the equation (19) in the previous report.)

$$G^m = \frac{1}{2}(N_A\psi_A + N_B\psi_B) + N_\alpha\psi_{B\alpha}$$

$$-kT\left[N_A \ln \frac{J_A}{N_A} + N_B \ln \frac{J_R}{N_B} + N_\alpha \ln \frac{J_\alpha}{N_\alpha} + N\ln N + N'\ln N' - N_0\ln N_0\right]$$

$$+z'kTN_\alpha \ln \frac{N}{\kappa N_A + N_B} \tag{2}$$

Accordingly, the chemical potential μ_α^m of a α-atom dissolved in alloy melt AB (refer to the equation (20) in the previous report), becomes,

$$\mu_\alpha^m = \partial G^m/\partial N_\alpha = \psi_{B\alpha} - kT \ln\frac{N'J_\alpha}{N_\alpha} + z'kT\ln\frac{N}{\kappa N_A + N_B} \tag{3}$$

Similarly, the chemical potential μ_B^m of a B-atom in the melt is expressed in

$$\mu_B^m = \partial G^m/\partial N_B = \frac{1}{2}\psi_B - kT \ln\frac{N'I_R}{N_B} + z'kT\frac{N_\alpha N_A(\kappa-1)}{N(\kappa N_A + N_B)} \tag{4}$$

In this report, μ_α^m and μ_B^m express the chemical potentials of a N-atom and an Al-atom in the molten Fe-Al alloy.

The condition of equilibrium in this research may be expressed by the following equation:

$$\mu_\alpha^m + \mu_B^m = \mu_{B\alpha} \equiv F_1(T) \tag{5}$$

That is, the sum of the chemical potentials of the nitrogen and aluminium atoms in a Fe-Al alloy melt is equal to the chemical potential of generated AlN compound, and since AlN does not form solid solution, its chemical potential $\mu_{B\alpha}$ may be expressed as a function $F_1(T)$ depending on temperature alone.

When equations (3) and (4) are substituted in equation (5), the next equation is given for the case in equilibrium:

$$\frac{N_\alpha}{N}\frac{N_B}{N} = \exp\left\{\frac{\mu_{B\alpha} - (1/2\psi_B + \psi_{B\alpha})}{kT}\right\}J_\alpha J_B\left(\frac{\kappa N_A + N_B}{N}\right)^{z'}\exp\left\{-\frac{z'N_\alpha N_A(\kappa-1)}{N(\kappa N_A + N_B)}\right\} \tag{6}$$

Now, let x stand for the concentration of aluminium in the Fe-Al alloy, or the atomic fraction N_B/N, and y for the atomic fraction N_α/N of the dissolved nitrogen atom, then the equation (6) becomes as follows:

$$xy = F_2(T)\left\{\kappa(1-x) + x\right\}^6\exp\left\{\frac{6(1-x)(1-\kappa)y}{\kappa(1-x) + x}\right\} \tag{7}$$

When, in equation (7), $\kappa = 1$ or $\psi_{A\alpha} - \psi_{B\alpha} = 0$, an ideal solution wherein α-atoms are distributed at random in the holes in Fe-Al crystal lattice is represented. In this case, $xy = F_2(T)$, which agrees with the accepted relation [Al][N] = constant derived from the law of mass action with the premise of ideal solution. In treating Fe-Al-N alloy systems in a general way, as is done in this report, the general equation (7) holds in correspondence of values of κ, wherein the hitherto accepted relation is included to express a special case.

Now, transform the relation (7) into (8) below, to discuss the case where the Fe-Al-N alloy system is in equilibrium with AlN slag under a fixed temperature:

$$y = F_3(T)\left(1 - x + \frac{x}{\kappa}\right)^6\exp\left\{\frac{6(1-x)(1-\kappa)y}{\kappa(1-x+\frac{x}{\kappa})}\right\}\frac{1}{x} \tag{8}$$

The first term $F_3(T)$ is a constant at a fixed temperature, the second an

equation treated in the previous report (see Fig. 2 of the previous report), and it follows that with a larger value for x, the value of the second-term grows larger, as in a Fe-Al-N alloy system $|\psi_{Ax}|<\psi_{Bx}|$ or $\kappa<1$; while the 3rd and 4th terms diminish with the rise in the value of x. Accordingly, the saturation value of nitrogen y at the first stage diminishes with the increase of aluminium content x, through the influence of the 3rd and 4th terms, and upon reaching a minimum, y begins to rise apace with the rise in the value of the 2nd term.
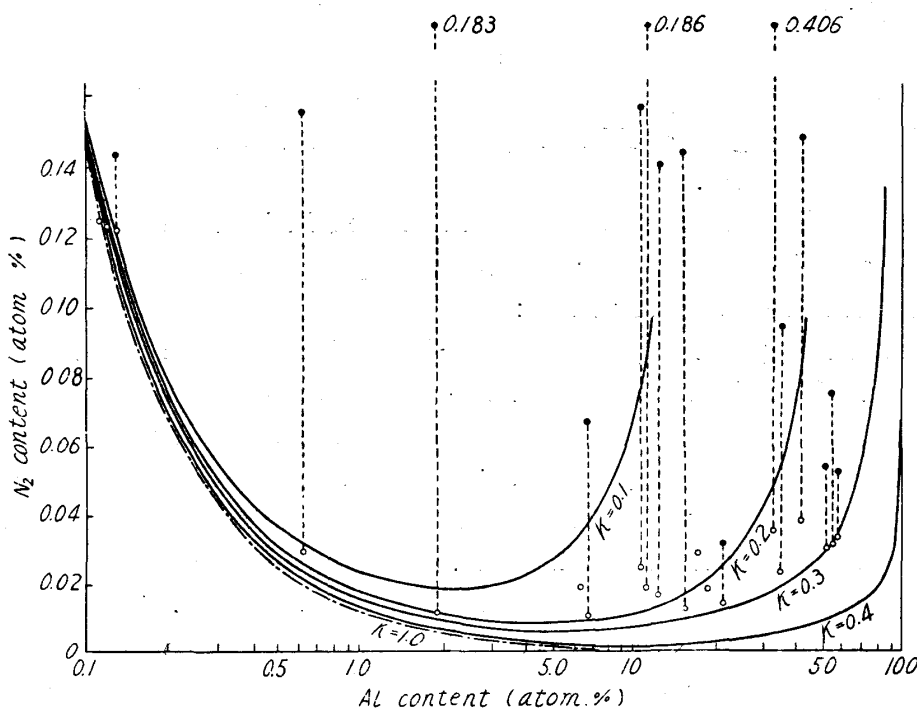

Fig. 3. Comparison between theoretical curves and experimental points.
- Total nitrogen content
- Interstitial nitrogen content

Fig. 3 shows the relation of equations (7) or (8), that is, the relation between x and y, for various values of κ. The measured values shown in Fig. 2 have been superposed for comparison with the curves deduced from this theoretical equation. The experimental values may be seen as represented by the curve lying in the range of $\kappa=0.2\sim0.3$.

From the above discussions, it may be considered that when nitrogen gas is dissolved in Fe-Al alloys, the nitrogen atoms do not situate interstitially in the lattice holes statistically at random, but cluster around Al atoms, to which they have stronger affinity than to Fe-atoms. Only when the behavior of absorbed nitrogen atoms is assumed as such, in general treatment, the curve of denitrification as shown in Fig. 2 can be deduced as the natural theoretical consequence.

Summary

No thoroughgoing experiment and discussion on the denitrification of iron melt have been known to date, and the rough conclusion that the more nitrogen

in the iron melt is eliminated, the larger the percentage of the added aluminium, has been accepted, in simple application of a relation $[Al][N]=$ constant, similar to that in the elementary study of deoxidation. Our research has elucidated the following basic phase of the reactions in denitrification. That is, a part of the nitrogen in a Fe-Al alloy is included in the form of AlN compound, while the other part is dissolved interstitially in the holes of Fe-Al quasi-crystal lattice in an atomic state. The reaction of denitrification cannot be adequately understood, unless the nitrogen in both the above forms is separately analysed for the respective quantity.

The ester-halogen methods were tried for the separate analysis, and found effective in measuring the AlN nitrogen and dissolved nitrogen contents to the accuracy of around 97%.

By the ester-halogen method, the total nitrogen in iron melts was separately measured for AlN nitrogen and dissolved nitrogen respectively and the following conclusion have been arrived at:

1)　There is no apparent quantitative relation between the total content of nitrogen and aluminium content in Fe-Al alloy, but between the dissolved nitrogen and aluminium contents, there exists a fixed relation explicable in the terms of statistical thermodynamics. The quantity of dissolved nitrogen at first sharply diminishes with the addition of aluminium, reaching a minimum in the range of approximately 1~5 wt.% aluminium, beyond that point, it increases gradually with the increase of aluminium content. This fact shows that the dissolved nitrogen atoms are not distributed statistically at random in the holes of the Fe-Al crystal lattice, but cluster around the aluminium atoms, with which nitrogen atoms has stronger affinity than with iron atoms.

2)　There was no quantitative relation between the AlN nitrogen contents and those of aluminium in Fe-Al alloys.

3)　Nitrogen gas reacts upon pure aluminium in molten state under one atmospheric pressure to form AlN compound, but no nitrogen was found dissolved therein in an atomic state. It has been hitherto believed that the saturation value of nitrogen in molten aluminium rises rapidly with a rise in temperature, but in fact, only the quantity of formed AlN increases with the rise in temperature, no interstitially dissolved nitrogen as mentioned above existing at higher temperatures under this pressure.