Effect of Gravity on the Macro-Segregation of Larger Steel Ingots

S. Onodera
Y. Arakida

UPON the problem involved in the study of solidification and segregation of larger steel ingots numerous papers have hitherto been published. Nevertheless their nature is still vague from both the scientific and practical points of view.

Previous studies have been devoted to the measurement of specific physical values of molten steel, the detailed observation of the sections of solidified ingots, simulation experiments using different materials, experiments on the formation and change of non-metallic inclusions, etc. Isn't there any room for re-examination in these methods of study ? Even if these items could be clarified in details, it would not give rise to a satisfactory solution for the phenomenon of "differential freezing" and to the reduction of segregation and other accompanied defects. We feel much necessity of thorough studies on the solidification and segregation of large steel ingots, before we rush into the epoch of large scale vacuum casting of steel ingots.

Apart from those audacious trials of the centrifugal process of heavy ingot making[1], the solidification of steel ingots, of large size in particular, has essentially been left as a "natural phenomenon"; ingots solidify layer by layer from the inner surface of iron mould set vertically in quiet air.

The effect of gravity on the solidification of ingots has been, therefore, tacitly taken into consideration in the previous knowledges, of which the sedimentation theory of negative segregation and upward movement of non-metallic inclusions are the main interpretations. Recently B. Gray[2] also studied the effect of gravity in which opinions much alike to the previous publications were repeated.

The present paper is a new step to the study of gravitational effect. Through observations by means of the bar-test method of measuring solidification rate and experiments of sand cast ingot inclined during solidification, a new finding was proposed to the effect of gravity on the formation of macro-segregation. Three major segregations, i.e. inverse V-, V- and negative segregations, are considered here.

Messrs S. Onodera and Y. Arakida, Research Dept., Muroran Plant, The Japan Steel Works Ltd., Japan.

1 K. Kurokawa: "Tetsu to Hagane" (J. Iron and Steel Inst., Japan), vol. 30, No. 1-2 (1944), pp. 9–18.

B. Gray: J. Iron and Steel Inst., vol. 182, Pt. 4 (April 1956). pp. 366–374.

Observations on the vertical segregation proceeding in the melt of ingot core by bar-test. Effect of gravity on the negative segregation

Bar-test

Intensive works research was carried out in the

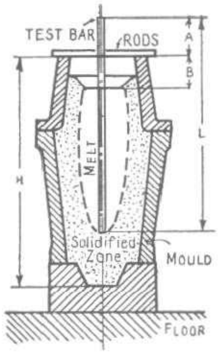


Fig. 1.
Layout of bar-test.

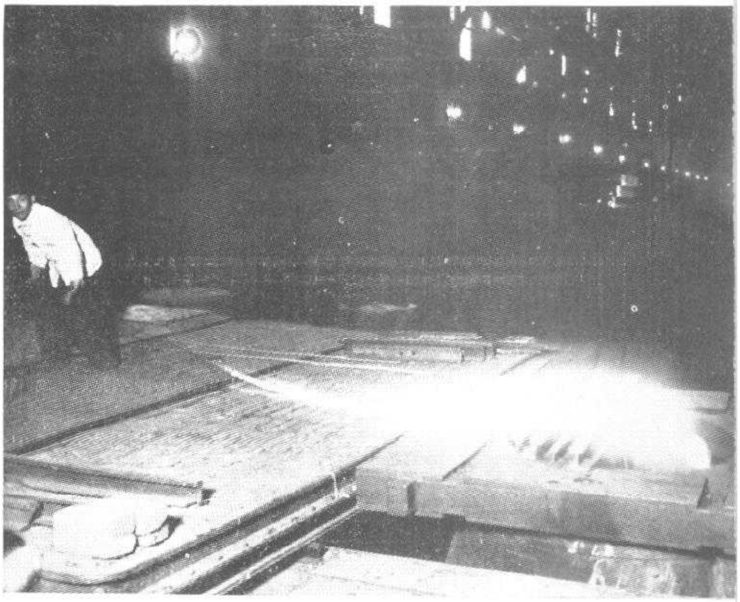


Fig. 2.
A scene of bar-test (Ingot: 60t).

authors' plant[3] in which the vertical solidification rate along the axial core of steel ingots weighing from 3^t to 140^t was measured non-destructively by the so-called "bar-test" method. The method prescribes itself that an ingot mould is set in a pit and several steel rods are put across the top of the mould to use them as a scaffold and standard plane for measuring (Figs. 1 and 2) : one of the two measurers drops a mild steel bar (9 mm ϕ, ground surface) into the ingot along the core, and when the tip of the bar gets against the liquid-solid interface, it is pushed in slightly at a force of 2 kg or less to confirm the solid shell : then the other puts a mark to the bar at the bottom of the standard rods and the bar is again pulled out rapidly. The height of solidification is calculated by the formula Fig. 1) :

$$\text{Height of solidification} = H - (L - A)$$

Some of the results are shown in Fig. 3[3] comparing with the previous works[4, 5].

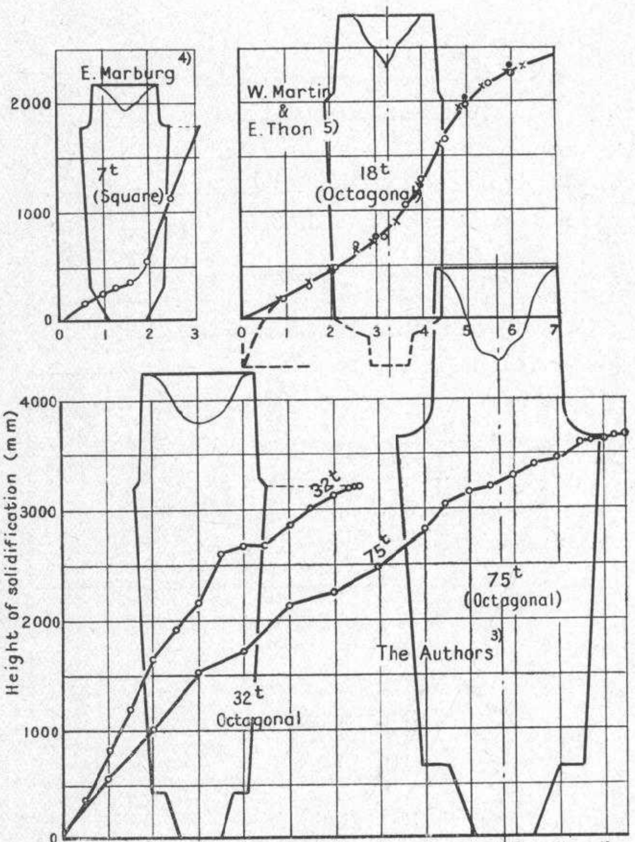


Fig. 3.
Typical solidification curves : variation with increasing ingot size.

[3] S. Onodera, Y. Arakida et al. : "Tetsu to Hagane", vol. 44, No. 1 (1958), pp. 9–14.

[4] E. Marburg : J. Metals, Feb. 1953, pp. 157–172.

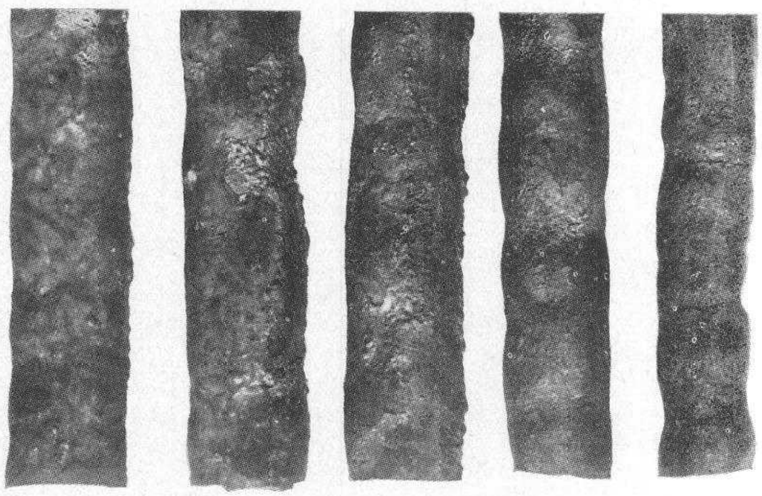
[5] W. Martin and E. Thon : Stahl u. Eisen, 75 (1955), S. 1,765–1,774.

Melt adhered to the test-bars[6]

During a short time of immersion which is usually 10 seconds or under, the diameter of the test-bars becomes about 20 mm or so due to the adhesion of solidified steel as shown in Fig. 4. The temperature rise of test-bars is estimated by a calculation as shown in Fig. 5. It is considered from Fig. 5 that the metal 1·5–2·5 mm deep after removing surface oxidised zone coincides nearly with the melt of the depth at which the bar has been stopped for marking for several seconds. This was approximately confirmed by a comparison of adhered metal with the sample taken from the longitudinal section of a split ingot.

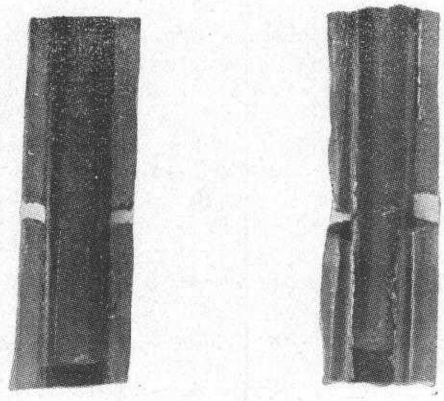
Results of chemical analysis of adhered metal

Table I is the result of analysis of adhered metal in the course of solidification of 47^t and 20^t ingots.



Bottom-side Top-side

(*a*) Surface undulation of adhered metal.



(*b*) Longitudinal section showing loose adhesion.

Fig. 4. Solidified melt adhered to the test bar.

[6] S. Onodera, Y. Arakida et al. : "Tetsu to Hagane", vol. 44, No. 8 (1958), pp. 872–880.

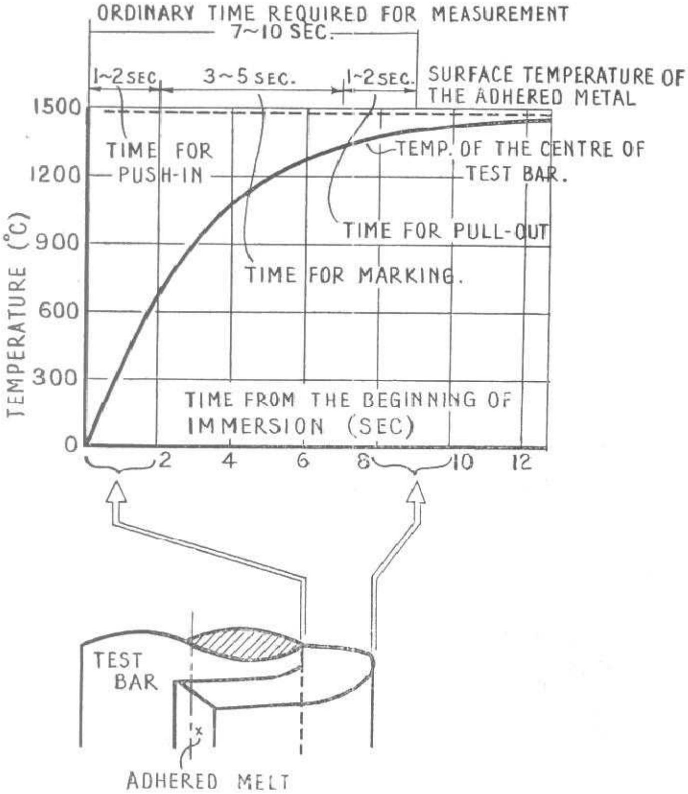


Fig. 5.

Estimated temperature rise of test bars during immersion.

Samples were taken from the above mentioned layer at every 400–600 mm. C, Si, Mn, P, and S for Ex. 1 of Table I, and C, Mn and S for Ex. 2 are plotted against the depth of immersion in melt, as in Figs. 6 and 7. From Table I and Figs. 6 and 7 the following is known :

(1) Soon after the pour, difference is hardly found in the chemical composition of the adhered melt between the top-side and bottom-side ends. But as the depth of immersion reduces due to the advance of solidification, the difference between the top-side and bottom-side ends increases. At the same time, the level of alloying elements becomes higher on the whole.

(2) This phenomenon is most remarkable for C, P and S, next for Si and Mn, while Ni, Cr, Cu, Mo and V remain almost unchanged.

(3) It has been a well known fact that the axial segregation along the ingot core takes a shape of curve A–B in Fig. 8. Comparing the curve A–B with Figs. 6 and 7, curve A–B is considered to be an envelope which connects the left ends of successive distribution curves of elements.

Discussion of the result

Fig. 9 shows schematically the iso-solidification lines and horizontal and vertical segregations. As time passes from t_2 to t_3, for instance, the alloying elements would be enriched into the melt. If the enriching is the sole action, the concentration of molten core must be continuously richer with nearly no gradient of concentration. The above observations indicate, however, some concentration gradient.

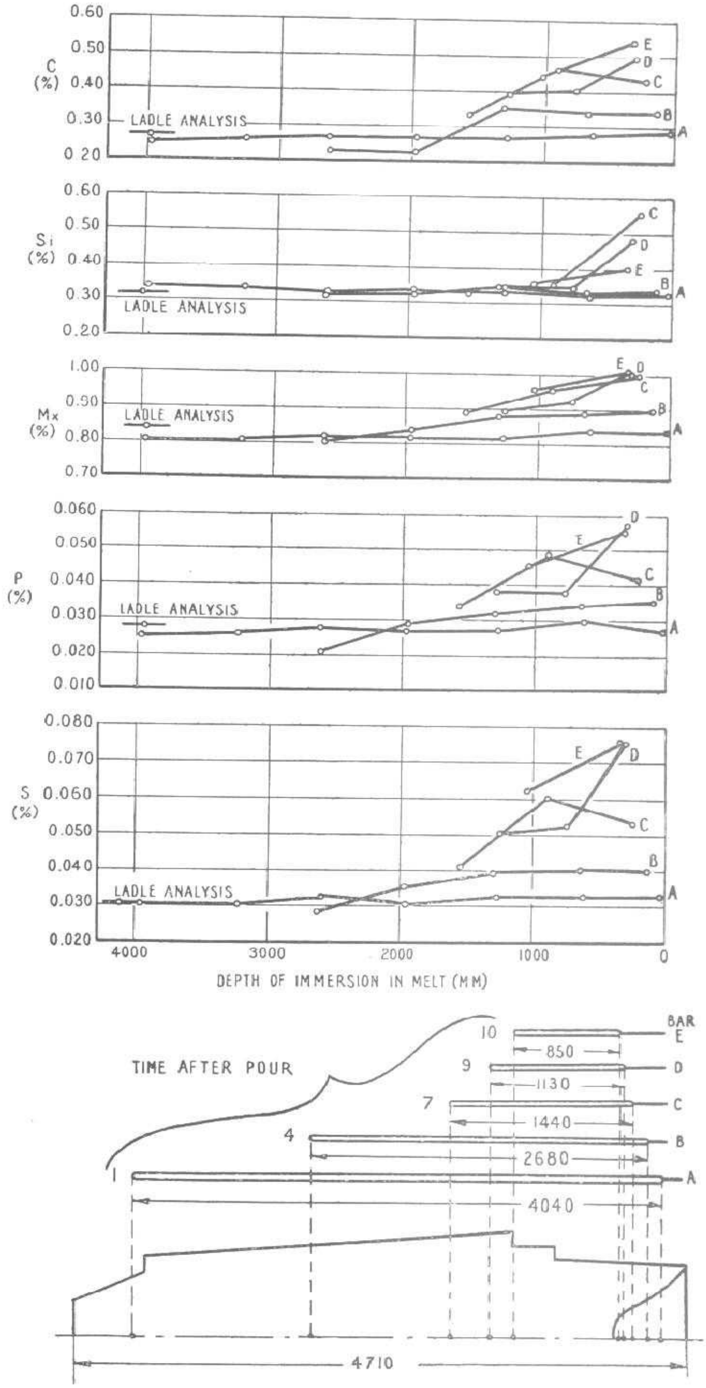


Fig. 6.

Analyses of melt adhered to the test bars ; samples taken at several points along their axis. (Ingot : 47t).

Table I

Results of chemical analysis of the adhered metal.

Example 1. 47ᵗ ingot.

Time after pour (hr)	No. of sample	Chemical composition (%)								
		C	Si	Mn	P	S	Ni	Cr	Cu	Mo
(Ladle)		0·27	0·32	0·84	0·028	0·030	0·11	0·08	0·17	0·01
	A_{1}	0·25	0·34	0·81	0·025	0·030	0·10	0·05	0·16	tr.
	$A_{4\text{-}5}$	0·26	0·34	0·81	0·026	0·030	0·10	0·07	0·17	,,
	$A_{8\text{-}9}$	0·26	0·33	0·82	0·028	0·032	0·11	0·05	0·17	,,
1	$A_{12\text{-}13}$	0·27	0·34	0·82	0·027	0·030	0·11	0·07	0·17	,,
	$A_{16\text{-}17}$	0·27	0·34	0·82	0·027	0·032	0·13	0·07	0·17	,,
	$A_{20\text{-}21}$	0·28	0·33	0·84	0·030	0·032	0·13	0·10	0·18	,,
	A_{24}	0·29	0·33	0·84	0·027	0·033	0·11	0·08	0·18	,,
	B_{1}	0·23	0·32	0·81	0·021	0·028	0·11	0·07	0·17	tr.
	$B_{4\text{-}5}$	0·23	0·33	0·84	0·029	0·035	0·13	0·07	0·18	,,
4	$B_{8\text{-}9}$	0·36	0·35	0·88	0·032	0·039	0·11	0·08	0·19	,,
	$B_{12\text{-}13}$	0·35	0·33	0·89	0·034	0·040	0·11	0·08	0·19	,,
	B_{16}	0·35	0·34	0·90	0·035	0·040	0·11	0·08	0·19	,,
	C_{1}	0·34	0·33	0·89	0·034	0·041	0·11	0·08	0·19	tr.
7	$C_{4\text{-}5}$	0·47	0·36	0·96	0·048	0·060	0·13	0·09	0·19	,,
	C_{8}	0·44	0·57	1·00	0·042	0·053	0·09	0·08	0·19	,,
	D_{1}	0·40	0·35	0·90	0·038	0·050	0·08	0·08	0·18	tr.
9	$D_{4\text{-}5}$	0·41	0·35	0·93	0·038	0·052	0·09	0·07	0·19	,,
	D_{8}	0·50	0·48	1·01	0·057	0·075	0·13	0·09	0·19	,,
10	E_{1}	0·45	0·36	0·96	0·045	0·062	0·11	0·08	0·19	tr.
	E_{4}	0·55	0·40	1·01	0·055	0·075	0·11	0·08	0·19	,,

Example 2. 20ᵗ ingot.

Time after pour (hr)	No. of sample	C	Si	Mn	P	S	Ni	Cr	Cu	Mo
(Ladle)		0·39	0·26	0·55	0·028	0·031	0·14	0·07	0.17	0·02
	K_{1}	0·34	0 28	0·50	0·020	0·025	0·17	0·08	0·17	0·02
	$K_{4\text{-}5}$	0·37	0·28	0·54	0·025	0·025				
	$K_{8\text{-}9}$	0·40	0·28	0·55	0·024	0·031				
1	$K_{12\text{-}13}$	0·41	0·29	0·55	0·023	0·033				
	$K_{16\text{-}17}$	0·41			0·026	0·038				
	$K_{20\text{-}21}$	0·41			0·027	0·031				
	K_{24}	0·42	0·29	0·54	0·024	0·034	0·17	0·09	0·18	0·02
	L_{1}	0·40	0·28	0·51	0·021	0·027	0·17	0·08	0·18	0·02
	$L_{4\text{-}5}$	0·40		0·52	0·021	0·030				
2	$L_{8\text{-}9}$	0·45	0·30	0·52	0·025	0·034				
	$L_{12\text{-}13}$	0·45		0·52	0·025	0·036				
	$L_{16\text{-}17}$	0·47	0·29	0·52	0·024	0·034				
	L_{20}	0·46	0·29	0·52	0·032	0·034	0·17	0·08	0·18	0·02
	M_{1}	0·42	0·29	0·51	0·026	0·035	0·17	0·08	0·17	0·02
	$M_{4\ 5}$	0·45		0·52	0·024	0·034				
3	$M_{8\text{-}9}$	0·47	0·30	0·54	0·031	0·040				
	$M_{12\text{-}13}$	0·50		0·55	0·029	0·038				
	M_{16}	0·55	0·35	0·56	0·035	0·042	0·17	0·08	0·18	0·02
	N_{1}	0·48	0·27	0·54	0·029	0·035	0·17	0·08	0·18	0·02
4	$N_{4\text{-}5}$	0·51	0·30	0·55	0·034	0·039				
	$N_{8\text{-}9}$	0·56		0·56	0·036	0·045				
	N_{12}	0·61	0·34	0·58	0·039	0·051	0·17	0·09	0·18	0·02
	O_{1}	0·52		0·54	0·033	0·040	0·16	0·08	0·18	0·02
5	$O_{4\text{-}5}$	0·66		0·57	0·040	0·055				
	O_{8}	0·68	0·30	0·57	0·043	0·058	0·17	0·09	0·17	0·02

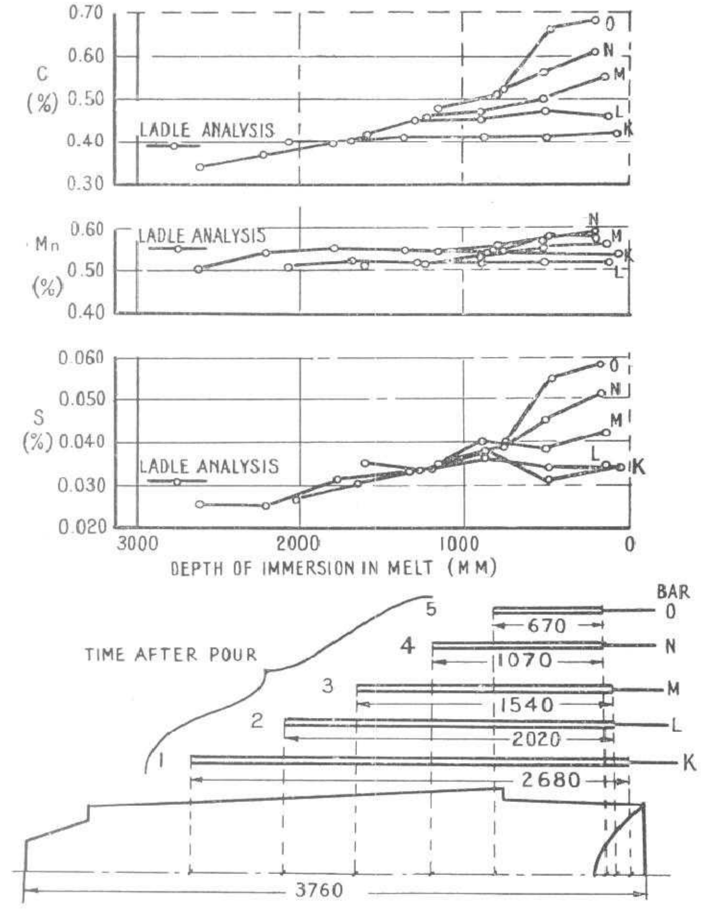


Fig. 7.

Analyses of melt adhered to test bars; samples taken at several points along their axis (ingot; 20t).

Thus the obtained result is supposed to have originated from

(Uniform enriching at the solidification front)
+(Proceeding of vertical segregation in the molten core).

In other words, it is evident that the vertical segregation proceeding in the molten core is captured.

This segregation in molten state is explained by the upward movement of light elements. Several previous studies or experiences support the view, as follows:

(1) *Experiment of Professor J. H. Andrew et al.*[7]

In a vertical crucible of 3/4″$\phi \times$10″, a melt of length 6″ was kept quietly for 24 hours, then frozen. The result of chemical analysis was as below:

	C	Si	Mn	P	S
Top	0·48	0·83	2·00	1·04	0·59
Bottom	0·33	0·51	1·10	0·21	0·20

Figures: %

Ingots stirred just before solidification, on the contrary, revealed no segregation.

(2) *Theory of solution in the gravitational field.*[8]

Assume that the melt of ingot core is kept at uniform temperature and is in equilibrium state. Then, from the relation between pressure P and composition N, the change of composition by height is calculated as follows:

$$\frac{dN_1}{dh} = \left(\frac{wv_1}{v} - w_1\right) g \Big/ \frac{\delta F_1}{\delta N_1}$$

where

w : mass of 1 mol of the solution
v : volume of 1 mol of the solution
w_1 : mass of 1 mol of a solute X_1
v_1 : partial molal volume of a solute X_1
N_1 : mol fraction of a solute X_1
F_1 : partial molal free energy of a solute X_1
g : gravitational acceleration
h : height

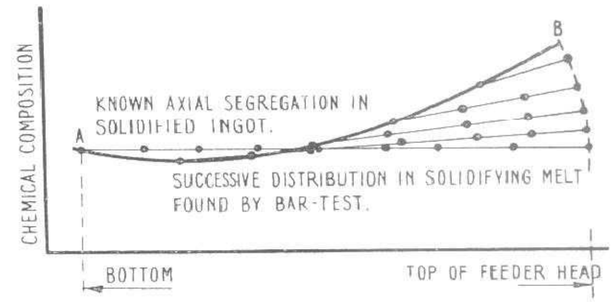


FIG. 8 AXIAL SEGREGATION AS RELATED TO THE RESULT OF ANALYSES OF ADHERED METAL.

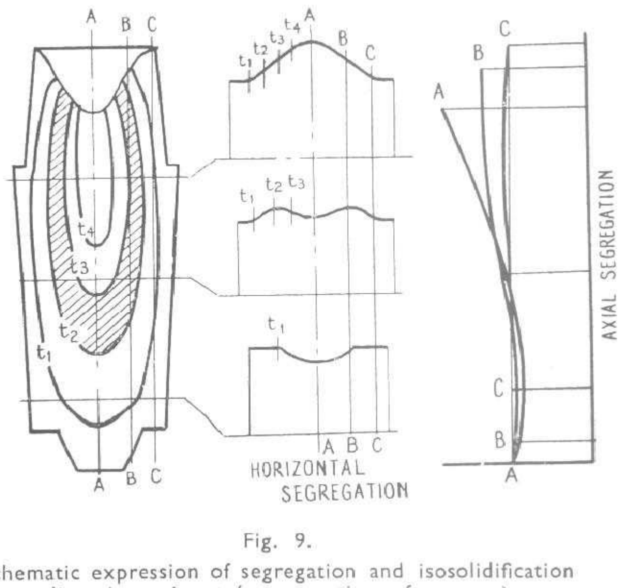


Fig. 9.

Schematic expression of segregation and isosolidification lines in an ingot ($t_1 \sim t_4$: time after pour).

[7] 8th Rep. Heterogeneity Committee, Iron and Steel Inst., Special Report No. 25 (1939), pp. 1–9.

[8] G. N. Lewis and M. Randall: "Thermodynamics", McGraw-Hill (1923), pp. 242–245.

This equation is, of course, not directly applied to such a rate process as solidification of ingot. But it is a strong clue for this phenomenon.

(3) *Experience of centrifugal casting of heavy ingot.*[1]

In the study of centrifugal casting process that was previously applied to ingots up to 80^t of weight, light elements and their compounds were forced to move towards smaller radius. Gravitational and centrifugal fields are of quite analogous character.

Mechanism of formation of negative segregation

Two theories have been proposed for the mechanism of formation of negative segregation; the sedimentation theory and the upward movement theory[9]. Neither has evidence that is confirmed in the actual ingots, but the former received wider support.

The observations by bar-test provide against the sedimentation theory with definite evidence. The above discussion needs, however, to be supplemented by considering a balance with the enriching at the solid-liquid interface, further discussion of which is given.

Behaviours of segregates in sand cast ingots solidified with their axes inclined. Effect of gravity on the V- and inverse V-segregation[10]

Experiments with sand cast ingots

To obtain further knowledge on the effect of gravity, an experiment was designed in which four sand mould ingots of $4{\cdot}9^t$ were held either vertically or in an inclined position, otherwise alternately inclined and re-erected, during solidification. The mould was constructed in two flasks for cast steel rolls as in Fig. 10.

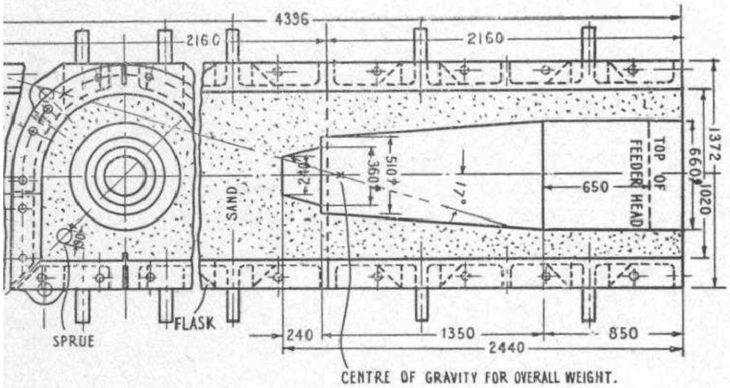
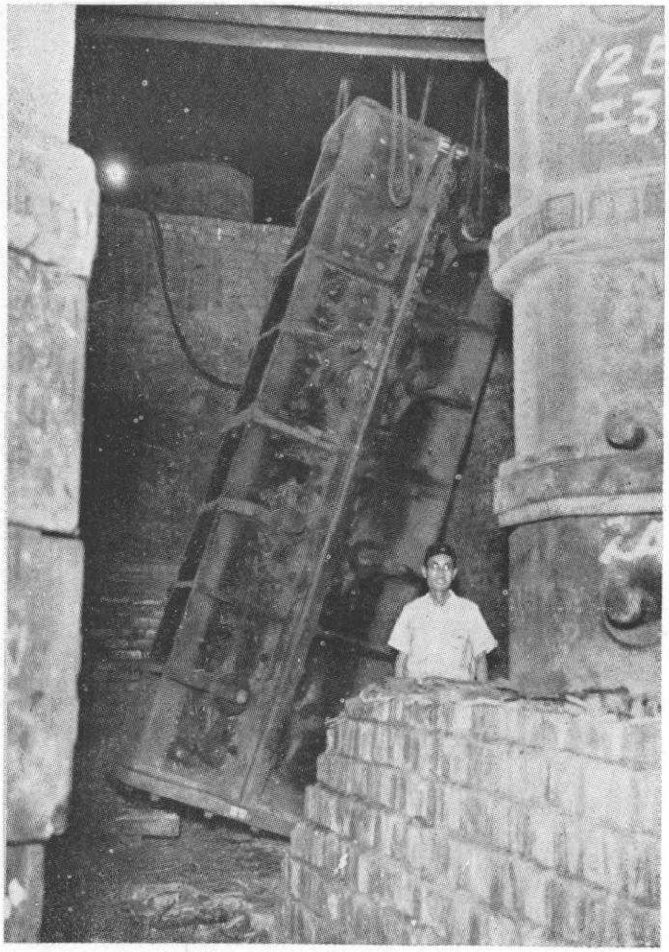


Fig. 10.
Sand mould for test ingots.



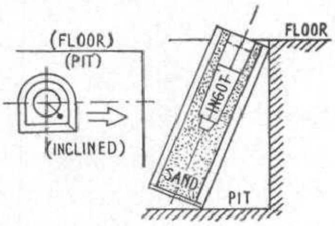


Fig. 11.
Mould inclined in the pit with ingot poured in it.

The keypoints of this experimental design are:

(1) Three macro-segregations appear very conspicuously in sand mould ingots even for smaller sizes, due to the slower speed of solidification. Preliminary measurement by bar-test for ingot No. 1* that was solidified vertically determined the vertical solidification rate to be five hours and a quarter which is equal to that of ordinary ingots of $22\text{–}25^t$.

(2) The inclining procedure can be carried out with sufficient safety, certainty and ease. In the

9 7th Rep. Heterogeneity Committee, Iron and Steel Inst., Special report No. 16 (1937), pp, 1-14.
10 S. Onodera and Y. Arakida et al.: "Tetsu to Hagane", vol. 44, No. 11 (1958), pp. 1,259-1,265.

* vide Table II.

case of an iron mould which usually consists of three pieces, the procedure is very dangerous or impossible.

(3) With comparatively a small angle of inclination, the whole mould plus ingot can be settled in a stable position.

The casting record for the four ingots is arranged in Table II. Fig. 11 is one of the moulds inclined in the pit.

Results of experiment

Figs. 12 and 13 are the sulphur print and macro-etching (cupic ammoneous chloride aq. method) for the four ingots. Figs. 14, 15 and 16 are the details of sulphur prints of ingots No. 2, 3 and 4.

1. *Observation of ingot No. 2*

From Figs. 12 and 14 the following can be said:

(*a*) Between the second inclination and the second re-erection, the V-segregates move to the right side of ingot, and both left and right branches of them make a clockwise turn slightly to get symmetry with the perpendicular during inclining.

(*b*) After the second re-erection, the V-segregates move leftwards to the axial zone, and left and right branches of them return to be symmetrical as to the axis of ingot.

(*c*) After the third inclination, the V-segregates behave just as the second inclination.

(*d*) Between the inclination and the movement

TABLE II

Casting record of test ingots

Items		Number of ingots			
		No. 1	No. 2	No. 3	No. 4
Chemical composition (%)	C	0·38	0·20	0·30	0·23
	Si	0·27	0·22	0·35	0·37
	Mn	0·55	0·45	0·68	0·62
	P	0·028	0·025	0·024	0·023
	S	0·032	0·028	0·037	0·011
	Ni	0·11	0·20	0·09	0·18
	Cr	0·14	0·06	0·05	0·14
	Cu	0·15	0·19	0·25	0·22
	Mo	0·02	tr.	0·02	0·05
Tapping temperature (°C)		1,630*	1,630*	1,630*	1,668*
Casting temperature (feeder head) (°C) ...		1,529*	1,525*	‡	1,530†
Casting rate (mn) ...		4′–00″	3′–05″	‡	‡
Time of solidification for ingot body (hr) ...		5°–15′	4°–30′	‡	‡
Condition of solidification		Cast and solidified vertically	Cast vertically. After 44 mn, Inclined. (1st time) After 38 mn more, Re-erected. (1st time) After 31 mn more, Inclined. (2nd time) After 38 mn more, Re-erected. (2nd time) After 37 mn more, Inclined. (3rd time) Then left to complete solidification	Cast vertically. After 3 hours passed, inclined and left to complete solidification	Cast vertically. After 34 mn passed, inclined and left to complete solidification

* Measured by immersion pyrometer. † Measured by optical pyrometer. ‡ Not measured.

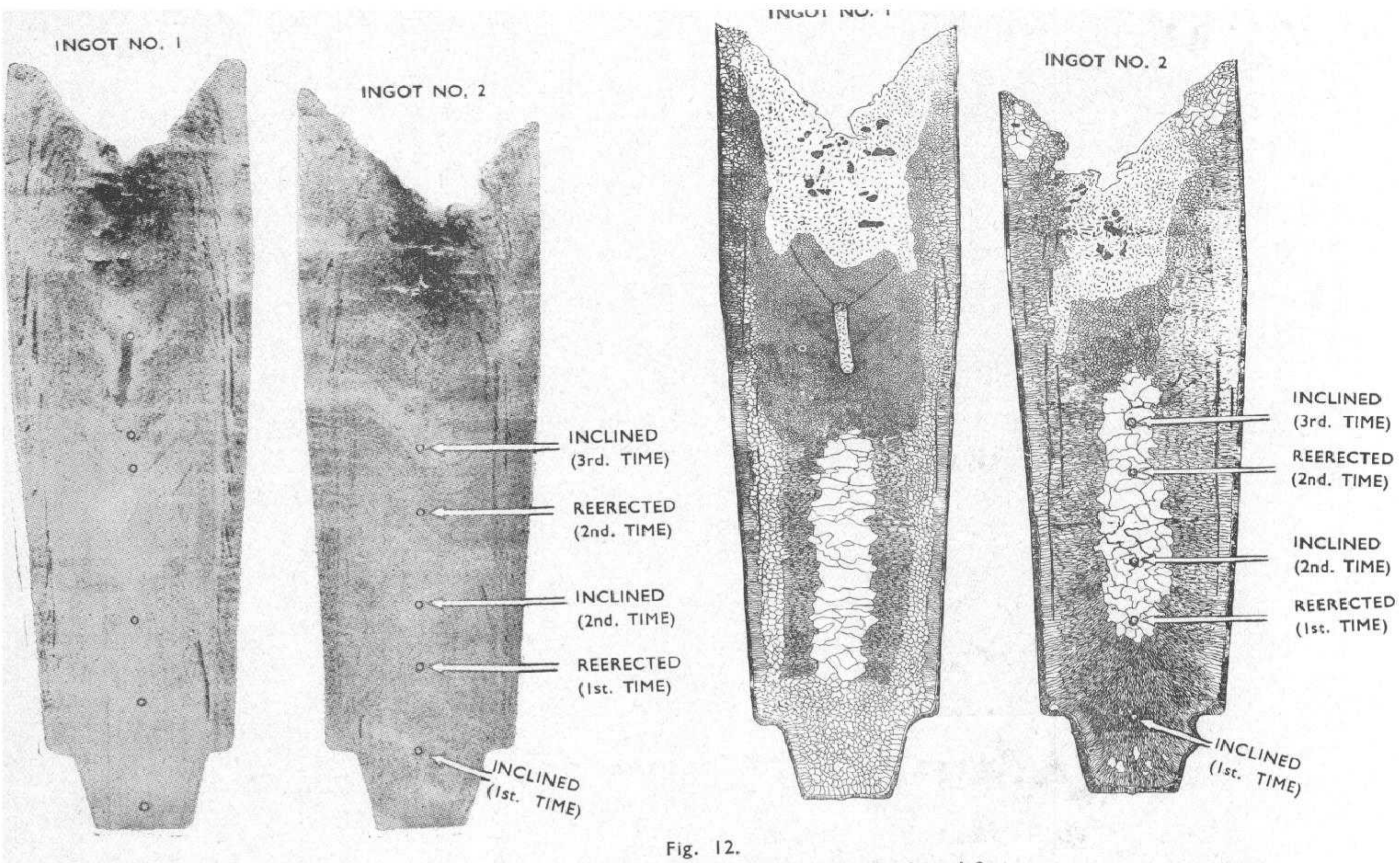


Fig. 12.
Sulphur print and macro-etching of ingots No. 1 and 2.

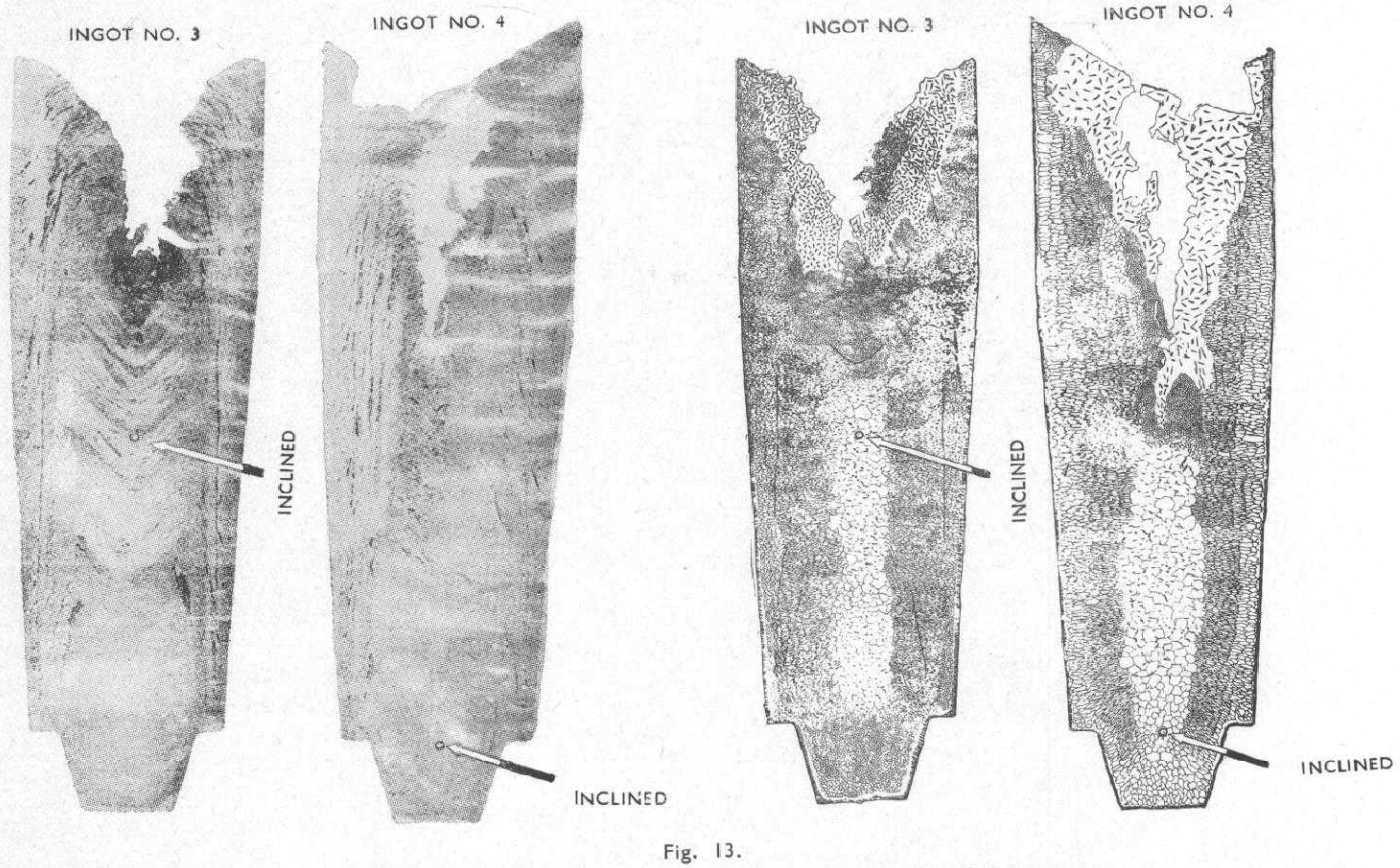


Fig. 13.
Sulphur print and macro-etching of ingots No. 3 and 4.

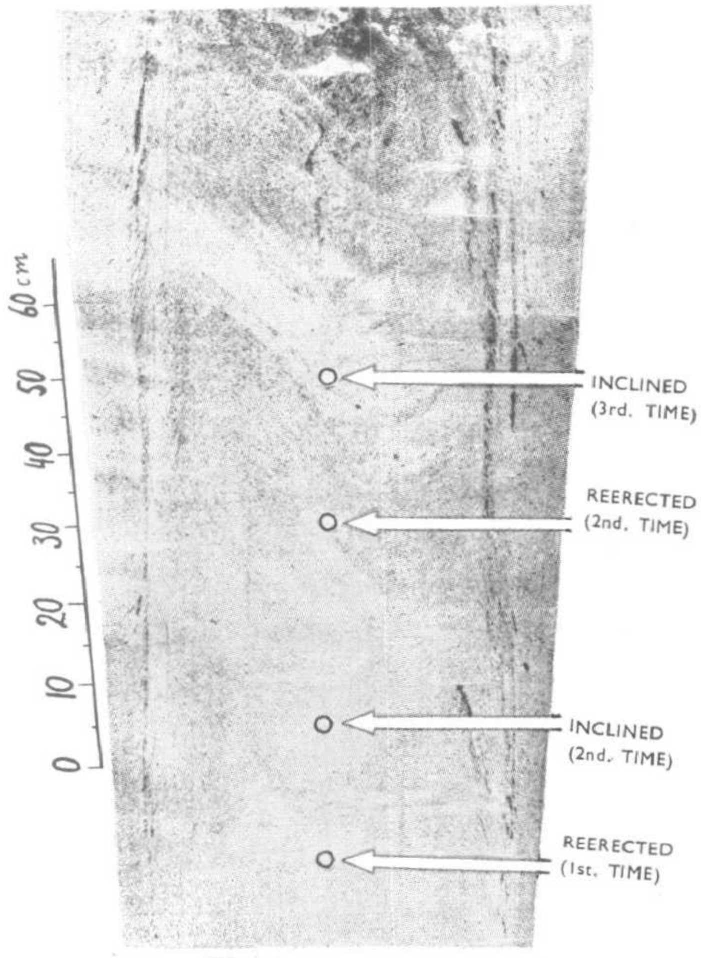


Fig. 14.
Sulphur print of middle part of ingot No. 2.

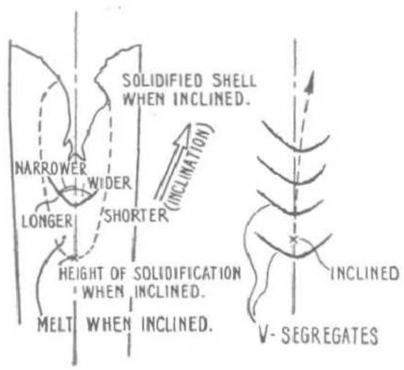


Fig. 15.
Sulphur print of ingot No. 3.
(Schematically drawn)

of V-segregates is some timely "gap", and the behaviours of V-segregates at the top-side of ingot are influenced accumulatively by the inclination of the lower side.

(*e*) By repeating the inclination and re-erection, the inverse V-segregate strings are blurred slightly. The zone of max. segregation at the top of ingot is also deformed by inclination.

(*f*) During the interval between the pour and a little after the third inclination, the solidification keeps nearly axial symmetry and is independent of the movement of the V-segregates.

2. *Observation of ingot No. 3.*

As shown in Figs. 13 and 15, the V-segregates after being inclined are behaving just like those of ingot No. 2.

3. *Observation of ingot No. 4*

From Figs. 13 and 16, it is observed that the V-segregates at the right half of ingot which lay

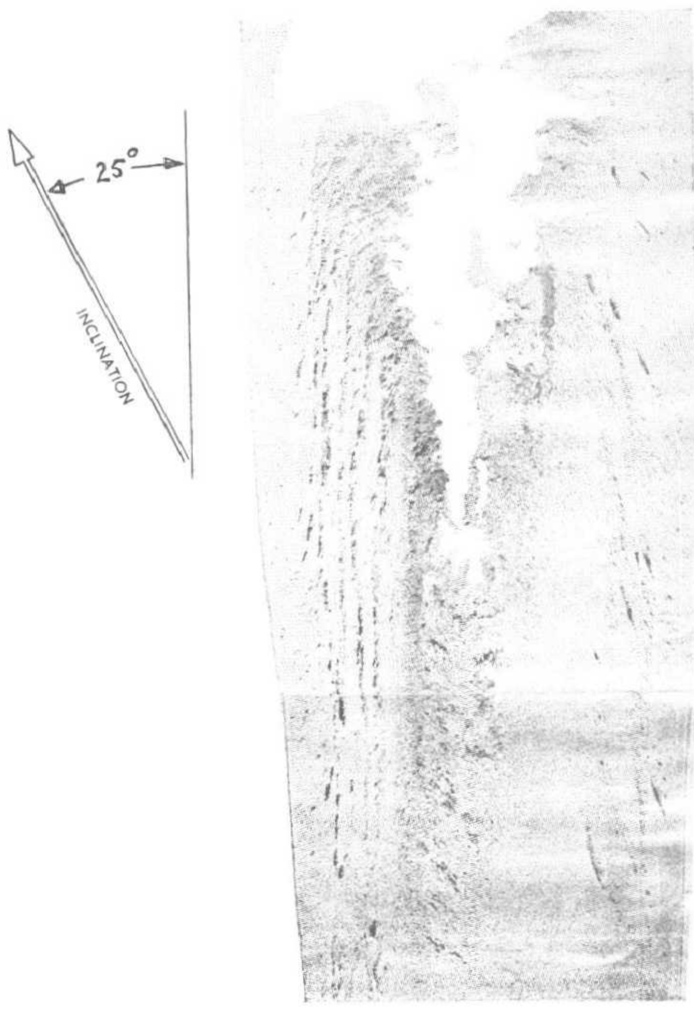


Fig. 16.
Detail of sulphur print of ingot No. 4.

in the lower level of gravity nearly disappear while in the left half of higher level of gravity they are much deeper to the extent of inverse V-segregates. The influence of inclination to the inverse V-segregates is in the same manner as for the V-segregates. It is also indicated that the convection of the solidifying melt seems negligible.

Discussion. Mechanism of formation of V- and inverse V-segregates.

Getting the above observations together, the results of this experiment are concluded as below:

(*a*) Convection in the solidifying melt seems to be negligible.

(*b*) The "germs" of V-segregates are already formed in the solidifying melt, which are fixed as V-segregates when the solid-liquid interface passes them. At some stage of solidification, they can move independently of the growing crystals.

(*c*) When a solidifying ingot is inclined (22–25° in this experiment), the germs of V-segregates in the melt move towards the direction of the new perpendicular. While, when the ingot is solidified in an inclined position from right after the pour, the V-segregates disappear within the half of the lower level of gravity. These show that the effect of gravity on the formation of V-segregates is very conspicuous and delicate.

(*d*) The effect of inclination is also notable for inverse V-segregates.

The forming mechanism of V-segregates indicated here is of particular importance. The close correlation that has been supposed between the solid-liquid interface and the V-segregates should be re-examined in the light of these results.

Unified interpretation for the mechanism of formation of the three major segregations[11]

Previous views

The individual cause of three macro-segregations, schematically shown in Fig. 17, has been proposed by many researchers. They are expressed in a brief manner:

Inverse V-segregation—strings of enriched metal the direction of which is a resultant of enriching and upward movement.

V-segregation—traces of enriched metal periodically remained corresponding to solid-liquid interfaces.

Negative segregation—sedimentation theory.

The new findings mentioned above suggest some modifications, especially to the theory of V- and negative segregations.

[11] S. Onodera and Y. Arakida: ibid., now in the press.

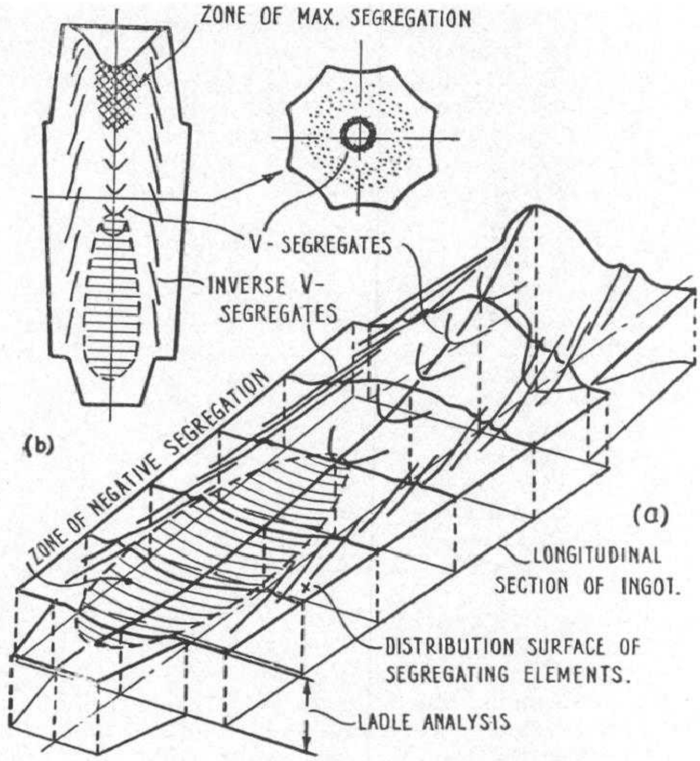


Fig. 17.

Three major segregations in a large ingot, perspectively drawn as peaks and valleys on the distribution surface along the longitudinal section.

Unified explanation for V- and negative segregations

The negative segregation has earlier been reasonably explained by the upward movement of segregating elements. Further, it has been indicated that the germs of V-segregates would presumably be formed in the solidifying core.

Combining these facts, a mechanism of formation of nagative and V-segregations can be derived. Along the core of ingot, upward concentrating action into the melt and upward transfer of segregating elements within the melt are the primary causes of segregation (Fig. 18*c*). Dilution of enriched elements or Soret effect at the solid-liquid interface is of minor influence. At the time when the solidification front reaches up to the point B, a peak as BCD due to enriching should be formed[18b]. In the zone of negative segregation, however, the upward movement covers the upward enriching fully, consequently no trace of enriching appears in the lower half of the solidified ingot. As the vertical concentration gradient in the melt increases and exceeds a certain value, the balance of upward movement with upward enriching is presumably broken, thus, some peaks are formed in the molten core which are fixed as V-segregates when the solidification front passes them.

In other words, the forming mechanism of V- and negative segregations is supposed to be essentially indentical. The difference in the balance between the upward concentration and upward movement in the molten core merely produces either of them, accordingly, a V-segregate can naturally lie in the zone of negative segregation.

Unified and systematic explanation for the three segregations

Joining the above theory to the existent view for the mechanism of inverse V-segregation, a systematic explanation for the forming mechanism of three major segregations is obtained. In Fig. 19,

Part A: Negative segregation in the lower half, but it gradually has V-segregates with the rise of the solidified shell.

Part C: Inverse V-segregation comes out as the solidification proceeds.

Part B: Intermediate zone of Parts A and C. Segregates are sprinkled on the whole.

Summary and conclusions

Observations by means of bar-test and experiments with sand mould ingots revealed some new findings concerning the effect of gravity on the formation of macro-segregation of large steel ingots.

The results are summarised as follows :

1. The negative segregation is formed by the upward movement of segregating elements.
2. Germs of V–segregates are already formed in the solidifying melt, which are fixed as V-segregates when the solid-liquid interface passes them. At some stage of solidification, they can move independently of the growing crystals.
3. Convection in the solidifying melt seems to be negligible.
4. The forming mechanism of V- and negative segregations is essentially identical. The difference in the balance between the upward movement in the melt merely produces V-or negative segregation.
5. Combining 4 with the existent theory for inverse V-segregation, an unified and systematic explanation for the mechanism of formation of three major segregations can be derived.

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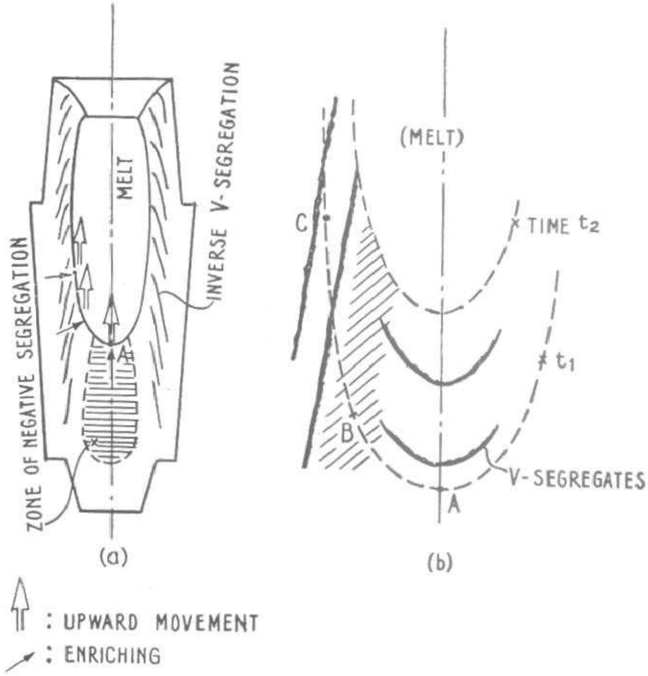

Fig. 19.
Segregations being formed near the solid-liquid interface.

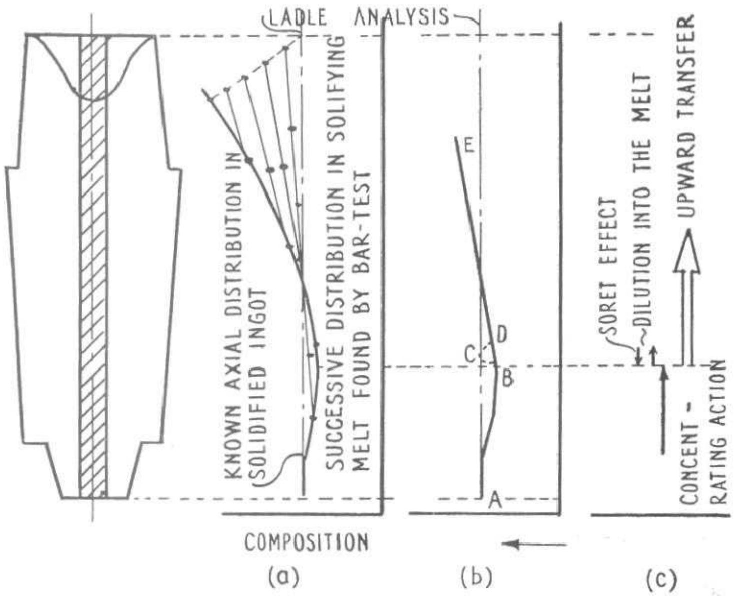

FIG. 18 MECHANISM OF VERTICAL SEGREGATION.