ATOMIC ENERGY OF CANADA LIMITED

Research and Development

EFFECT OF CARBON ON THE GRAIN REFINEMENT OF URANIUM

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by

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Chalk River, Ontario
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ABSTRACT

A study was undertaken on small uranium samples to investigate a suitable method for grain refinement and to determine the effect of carbon on the grain refinement of uranium.

It was found that considerable grain refinement may be achieved in samples which have been quenched from the beta phase and annealed at high alpha-phase temperatures. A study of the microstructures obtained in this report support a recrystallization mechanism for grain refinement by this process.

The effect of carbon content on the magnitude of grain refinement in the surface layers is small whereas the effect on the interior of the sample is large; the degree of refinement increased with increasing carbon content within the range 10 ppm to 1000 ppm.

I INTRODUCTION

It is now well established that uranium fuel elements which have a certain strong preferred orientation suffer gross deformation upon irradiation. This dimensional instability can essentially be eliminated by heating into the beta phase and thereby producing an almost random structure. There is evidence[1] that there is a weak (100) texture after beta heat treatment which agrees with the findings[2][3] that beta heat treated NRX rods always shorten slightly during irradiation.

Beta heat-treatment, however, produces a marked enlargement of the grain size which is considered responsible for surface roughening or "wrinkling". Fine grained as-rolled plates do not show this behaviour upon irradiation. Although "wrinkling" has not been observed on NRX rods, it has been seen on NRU flats[4].

One trouble which has been experienced with NRX rods is that of the sheath rupturing during irradiation; this has been correlated with the formation of longitudinal cracks in the uranium[5]. In addition, transverse cracks have also been observed although these cracks have caused less trouble than longitudinal cracks. This cracking phenomenon is presumably a result of the combination of high thermal stresses and lack of ductility in irradiated uranium. Because the stress required to propogate a crack is inversely proportion to the square root of the grain size, it is considered that grain refinement may help alleviate this cracking problem.

The work to be described was therefore initiated to find a suitable method for grain refinement and to determine the effect of carbon on the grain refinement of uranium. Gardner and Riches[6] have studied the kinetics of the beta to alpha transformation and have found that rapid cooling from beta-phase temperatures followed by an anneal at high alpha-phase temperatures effects a marked refinement of the grain size of the transformed metal. Using the above investigation as a guide, a study was undertaken to obtain optimum grain refinement, on small samples of uranium with varying carbon contents, by quenching from the beta region and annealing for various time intervals in the alpha phase at 650°C.

II EXPERIMENTAL PROCEDURE

(a) Materials

The uranium metal used in these experiments was taken from NRX rod material of low, medium and high carbon contents. These samples were 1/8 inch thick and 1.36 inches diameter. Some tests were also done on dingot uranium obtained from Fernald Material Processing Centre. The dingot samples were 1/8 inch thick and 1 inch diameter. The uranium analyses are given in Table 1.

TABLE 1

Uranium Analyses

Material	Impurities - p.p.m.		
	C	Fe	Al
High Carbon	1000	30	5
Medium Carbon	370	31	3
Low Carbon	50	35	5
Dingot	10	28	8

(b) Heat Treatment

All heat treatments were carried out in a salt bath containing a eutectic mixture of lithium and potassium carbonates, the temperature being controlled to $\pm$ 3°C. The specimens were suspended in the centre of the salt bath by use of an Inconel wire cage.

The samples were heat treated in the beta range (750°C) for four minutes and quenched by immediate immersion in cold water. Moderate stirring of the samples in the water was carried out during the quench in order to ensure that a steam envelope was not formed around the samples. The majority of specimens were then given an alpha anneal at 650°C for various time intervals followed by water quenching. Alpha annealing times listed in this report refer to times at temperature, allowances being made for the time required to come to temperature as determined by preliminary measurements. A few samples were just heated into the beta region and air cooled.

Heating and cooling curves were determined for a dummy sample

to follow the beta quench portion of the test; for this a D.C. amplifier and a Brush Oscillograph were used. In this test thermocouples were placed in 1/8 inch O.D. Inconel tubes which were either attached to the outside of the sample or placed in a hole extending from the top surface to the middle of the sample.

(c) Metallographic Examination

Heat treated specimens were cut in half (into two semi-circular discs) and both surface and interior regions were polished, examined and photographed. The polishing procedure consisted of mechanically polishing the samples on silicon carbide papers with kerosene as the lubricant followed by diamond polishing. The samples were then electrolytically polished using the following electrolyte: 50 ml of a solution of 100 gm chromic oxide in 118 ml water, and 200 ml of acetic acid.

III RESULTS

(a) Heating and Cooling Curves

The heating and cooling curves for the outside and inside of a dummy sample heated into the beta range (750°C), held there for four minutes, and then quenched into cold water are shown in Figures 1 and 2 respectively. From the heating curves, it can be seen that both the exterior and the interior of the samples heated up reasonably quickly. The outside and inside of the sample reached the required temperature of 750°C in approximately 15 and 22 seconds respectively.

In the determination of the cooling curve for the surface of the sample, there was a two second delay between taking the sample out of the salt bath and immersing it in water as compared with a three second delay in determining the corresponding curve for the interior region. Consequently, the temperature of the exterior and interior regions immediately before insertion into the water bath was 720°C and 710°C respectively.

Both the interior and exterior regions reached the α - β transition temperature (668°C) in approximately 0.05 seconds after being immersed in water. The cooling curves are reasonably linear down to the α - β transition temperature giving cooling rates of approximately 1040°C/sec. and 840°C/sec. for the exterior and interior regions respectively.

(b) Discussion of Microstructures

The microstructures of the interior and exterior regions of specimens of various carbon contents and for various heat treatments are shown in Figures 3 to 15 inclusive. All the fields shown were examined and photographed using polarized light and are at a magnification of X75.

Figures 3 and 4 show the structures of the surface and interior regions for dingot uranium (10 p.p.m.c) specimens which have been air cooled and water quenched from the beta phase. The grains are quite large and irregular for both the air-cooled and water quenched specimens. It appears that the grains are slightly smaller in the water quenched specimens. It is difficult to

ascertain for certain the magnitude of this effect due to the irregular and poorly defined grains.

Maximum grain refinement in dingot uranium occurred for a five minute alpha anneal after beta-quenching and the corresponding microstructures are shown in Figure 5. The surface of the sample exhibits some grain refinement as characterized by smaller, better defined and equi-axed grains. The grain refinement appears to extend for one or possibly two grains into the interior of the specimen beyond which the grains are rather large and irregular. Measured grain size for the surface region is 120 microns (μ) and for the interior region is 160 μ (values tabulated in Table 2).

TABLE 2

Summary of Grain Sizes Obtained in Uranium Samples Containing Different Carbon Contents by Water Quenching from the Beta Phase followed by Alpha Annealing at 650°C

Material	Alpha Annealing Time (Minutes)	Mean Grain Size (Microns) Surface	Interior
Dingot	5	120	160
Low Carbon	10	115	140
Medium Carbon	10	95	110
High Carbon	10	90	100

Alpha annealing uranium samples for 10 and 20 minutes produced virtually no change in the microstructure of either the surface or interior regions.

Corresponding microstructures for low carbon (50 p.p.m) specimens appear in Figures 6, 7 and 8. Similar to dingot uranium,

samples air cooled or water quenched from the beta region exhibit large and irregular grains. A sample alpha annealed for 10 minutes at 650°C after beta quenching (Figure 8) exhibits considerable refinement on the surface but very little in the interior. The surface grain refinement extends for one or two grains into the interior. Measured grain size for the surface and interior regions are 115 μ and 140 μ respectively. Alpha annealing a sample for 15 minutes left the microstructures of surface and interior regions virtually unchanged.

The microstructures for medium carbon (370 p.p.m.) material are shown in Figures 9, 10, 11 and 12. Samples air cooled or water quenched from the beta region (Figures 9 and 10) are similar to those for dingot and low carbon material. However, the effect of alpha annealing after beta quenching is now more significant, since a medium carbon sample annealed for 10 minutes at 650°C (Figure 11) exhibits good grain refinement throughout the sample in contrast to that for dingot and low carbon material. Measured grain sizes are 95 μ and 110 μ for surface and interior regions respectively. Increased annealing times gave primary grain growth, see Figure 12.

The microstructures for high carbon (1000 p.p.m.) material are shown in Figures 13, 14 and 15 and are similar to those obtained for medium carbon specimens. Alpha annealing for 10 minutes produces grain refinement throughout the specimen (Figure 15). Measured grain sizes are 90 μ and 100 μ for surface and interior regions respectively.

Due to the irregular nature of the grains for material water

quenched from the beta phase, it is rather difficult to ascertain the magnitude of the effect of carbon on the beta-quenched grain size. It appears, however, that there is a general trend for increasing carbon content to cause a decrease in the beta-quenched grain size (compare Figures 7 and 14).

IV DISCUSSION OF RESULTS

Consistent with the results of Gardner and Riches, it has been found in this study that considerable grain refinement may be achieved in small uranium samples which have been quenched from the beta phase and annealed at high alpha-phase temperatures. The effect of carbon content on the magnitude of grain refinement in the surface layers is small whereas the effect on the interior of the sample is large; the degree of refinement increasing with increasing carbon content within the range 10 p.p.m. to 1000 p.p.m.

This study was undertaken to examine the effect of carbon on the grain refinement in uranium and no real attempt was made to study the mechanism governing the grain refinement by the quench-anneal method. However, it is possible to make some comments on the mechanism by which grain refinement possibly occurs. When uranium is cooled through the α - β transformation, temperature internal stresses are set up in the material as a result of three additive effects, (1) the volume change on transforming the β phase to alpha, (2) internal stresses set up by rapid cooling and (3) micro-stresses which result from the anisotropic thermal expansion of uranium. The internal stresses set up are large enough to cause plastic deformation; evidence that this is so is given by

the fact that beta-quenched samples show profuse twinning and also well developed polygonized structures. Thus after quenching, the material is in a state equivalent to that obtained by cold working and recovery, and so if given chance will recrystallize to a lower energy state.

A study of the microstructures obtained in this report support a recrystallization mechanism in that:

(1) alpha annealing produces an equi-axed grain structure with straight grain boundaries.

(2) grain refinement occurs first in the surface of the samples which is the most severely quenched region.

(3) the grain size decreases with increasing carbon content indicating that carbide particles are acting as nucleating centres for new grains or are impeding the motion of grain boundaries or both.

(4) the surface grain size is similar to the interior grain size with a carbon content exceeding 370 p.p.m. indicating that the effect of having lower quenching stresses in the interior of the specimen than at the surface is being compensated by the increase in the number of carbide particles.

It is interesting to note that Springfields in the U.K. do not normally observe recrystallization after alpha annealing beta quenched uranium, no matter whether this is calcium or magnesium reduced metal[7]. However, with material exceptionally low in iron and aluminium, they have observed recrystallization such as that reported here. The normal aluminium and iron contents of magnesium reduced uranium prepared in the U.K. is approximately 35 p.p.m. and 90 p.p.m. respectively[8]. This is higher than the iron and aluminium contents in the material used for this work. It is apparent, therefore, that to produce grain refinement by the quench anneal method, the iron and aluminium contents must be kept low. These results give further support to the recrystallization mechanism for Springfields have also found that relatively small amounts of iron and aluminium (few hundred parts per million) will inhibit the recrystallization of uranium during hot rolling at 600°C. Thus it appears that if the iron and aluminium contents are excessive, recrystallization is impeded and grain refinement by the quench anneal method is prevented.

Consider now the problem of the cracking tendency of NRX rods upon irradiation, as outlined previously in this report. This cracking phenomenon is presumably a result of the combination of high thermal stresses and lack of ductility in irradiated uranium. As shown above, in addition to the thermal stresses, there will be quenching stresses present exceeding the yield strength of uranium. The quenching stresses within the centre core of the NRX rod will presumably be relieved during operation but

those at the outside zone will still remain. These internal quenching stresses may themselves be a factor in the cracking of NRX rods. Alpha annealing after beta quenching may, therefore, relieve the cracking problem in two ways: (1) by grain refinement and (2) by relief of the quenching stresses. It is interesting to note that both the U.K. and Sweden give a stress relief anneal at 500-550°C after beta quenching.

V ACKNOWLEDGEMENTS

The author is very grateful to Mr. G.P. Kiely and Mr. H. Boychuk who assisted in the heat treatment and metallographic preparation of the uranium samples.

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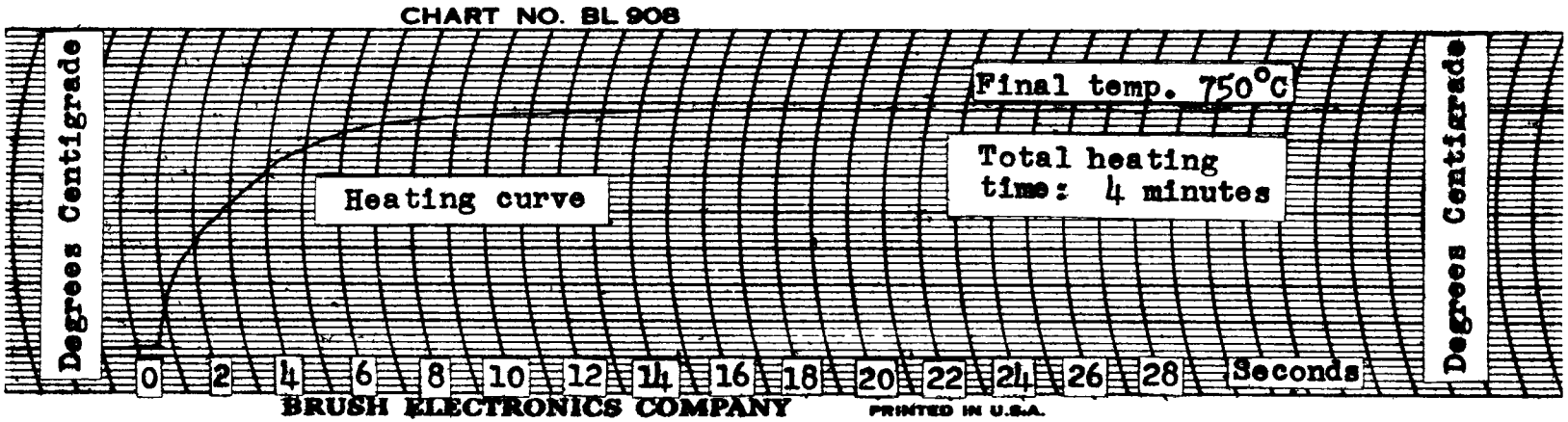


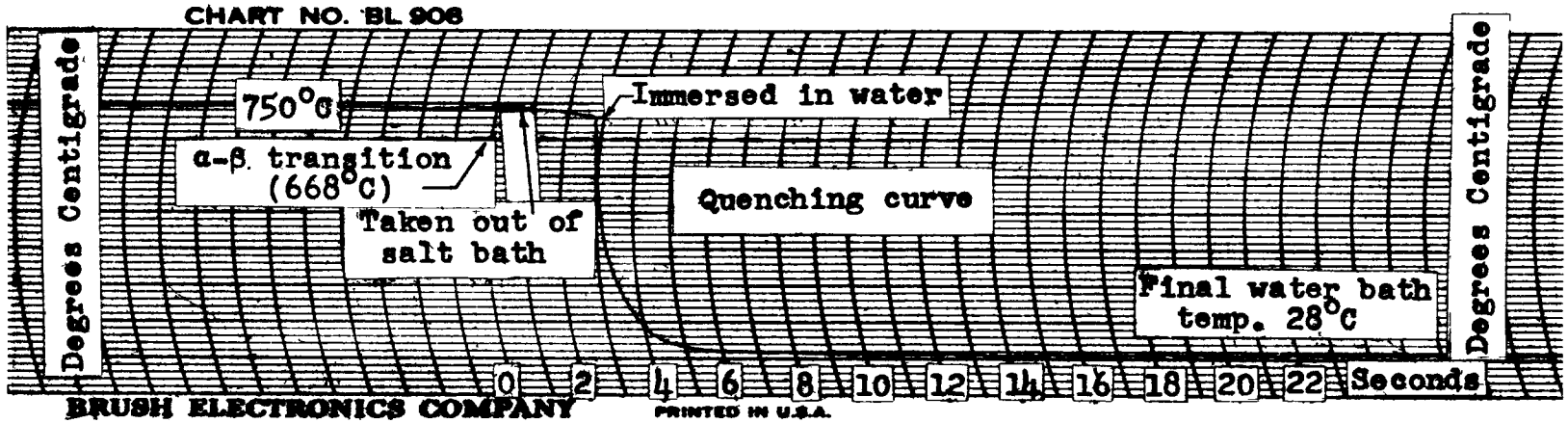
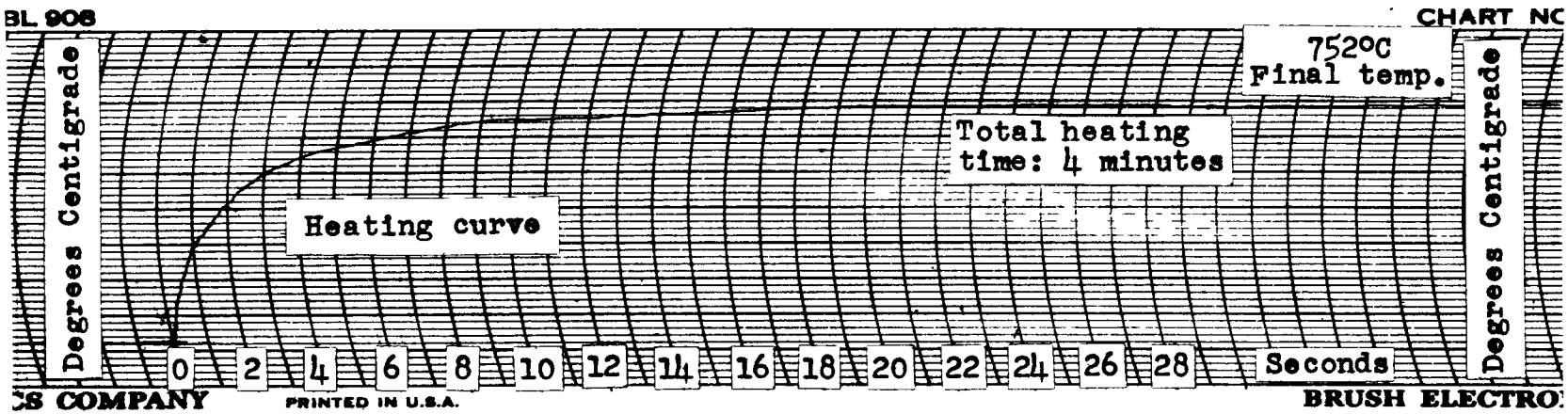


Fig. 1: Heating and cooling curves for the outside region of a 1/8 inch length of N.R.X. rod.



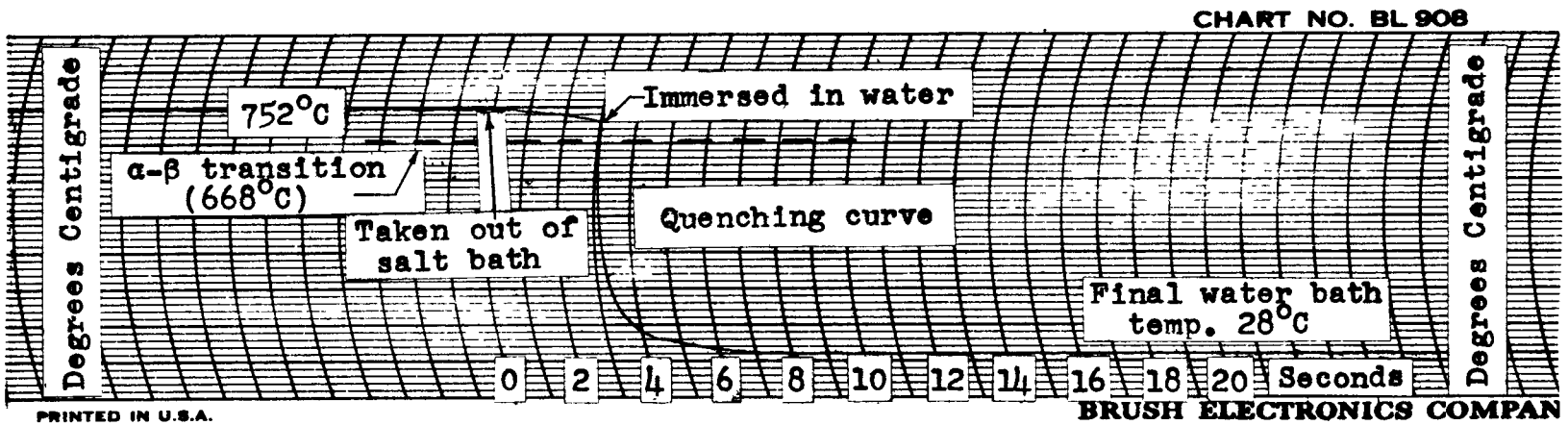


Fig. 2: Heating and cooling curves for the centre region of a 1/8 inch length of N.R.X. rod.

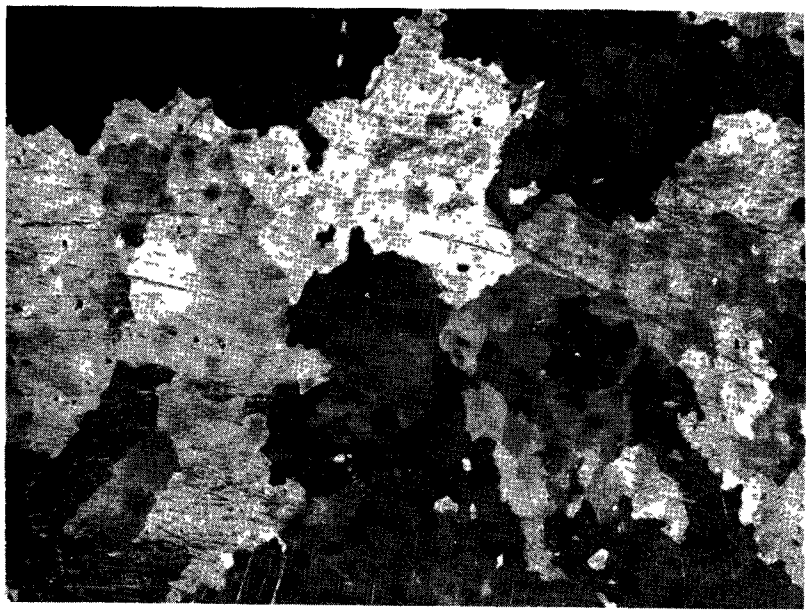


Fig. 3: Surface (top) and interior (bottom) of dingot uranium (10 p.p.m.c.) specimen air cooled from beta region (750°C.).
X75

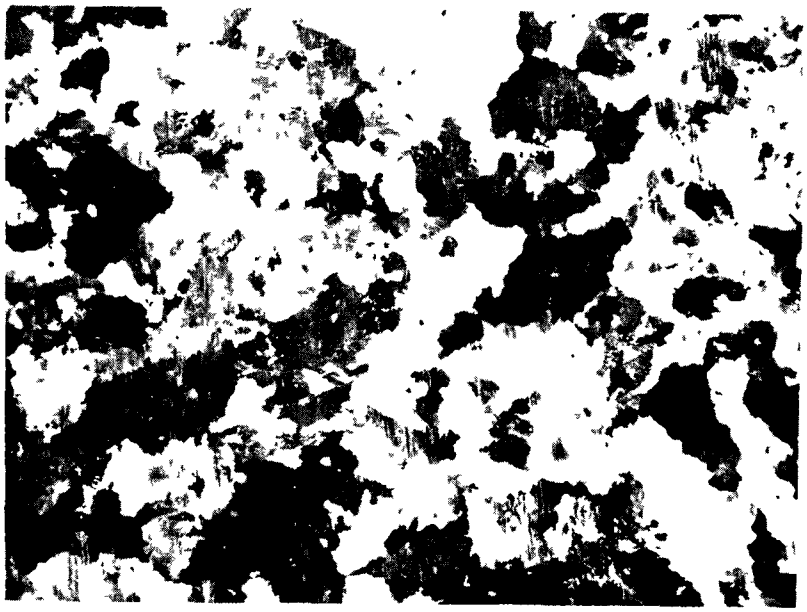
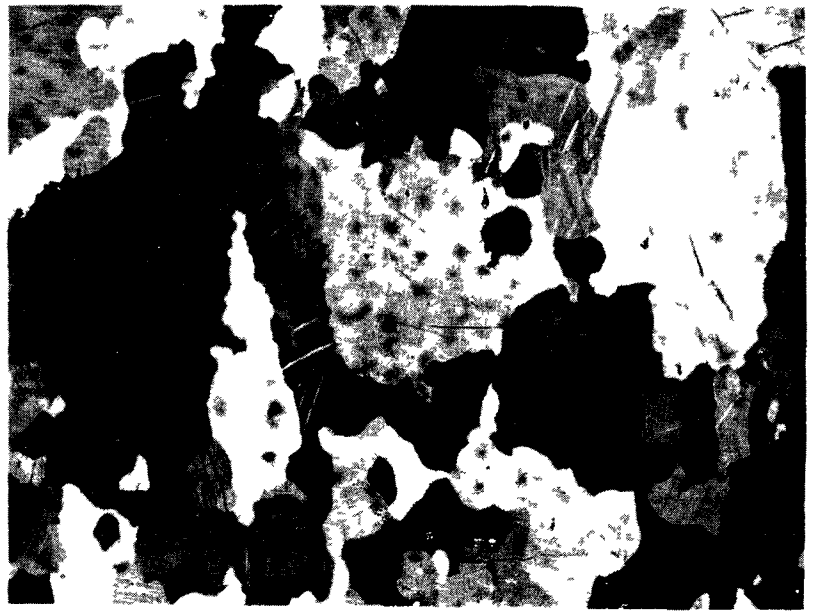


Fig. 4: Surface (top) and interior (bottom) of dingot uranium (10 p.p.m.c.) specimen water quenched from beta region (750°C.).
X75

Fig. 5: Surface (top) and interior (bottom) of dingot uranium (10 p.p.m.c.) specimen water quenched from beta region (750°C.) and annealed in alpha region (650°C.) for 5 minutes.
X75



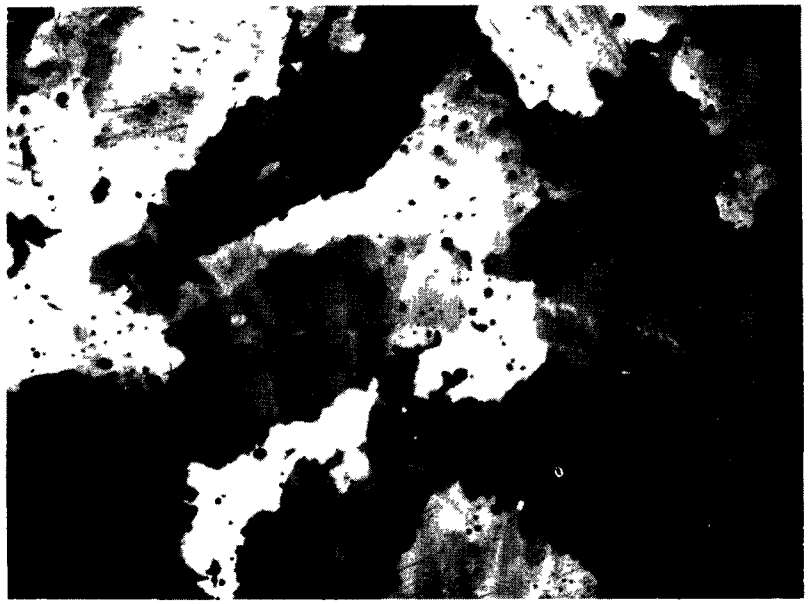


Fig. 6: Surface (top) and interior (bottom) of low carbon (50 p.p.m.) specimen air cooled from beta region (750°C.).
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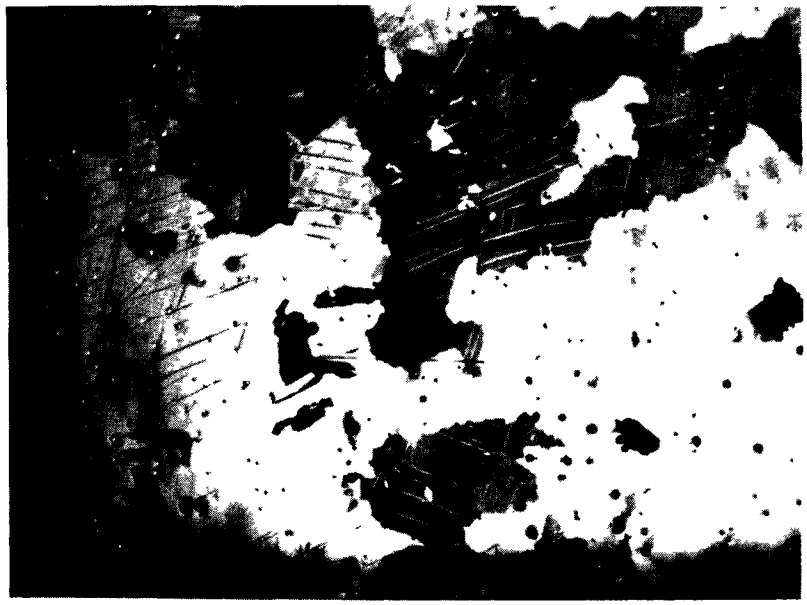


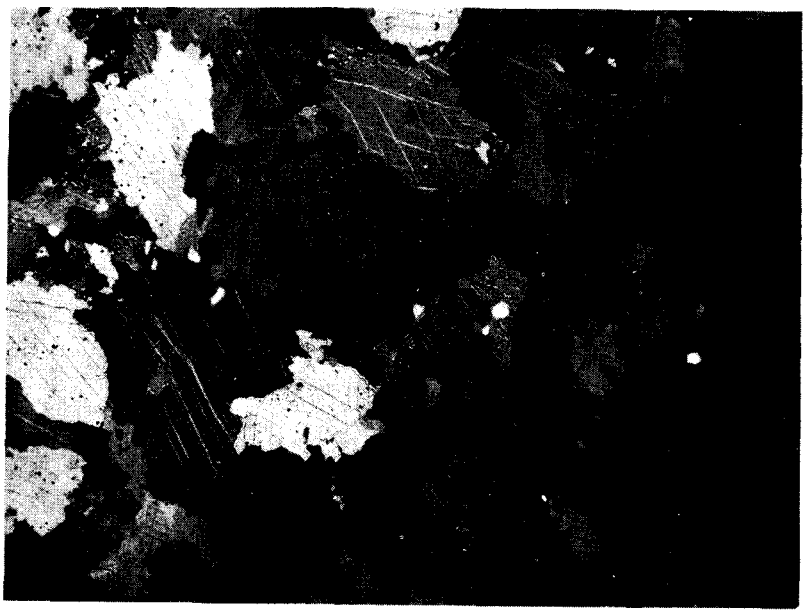


Fig. 7: Surface (top) and interior (bottom) of low carbon (50 p.p.m.) specimen water quenched from beta region (750°C.).
X75

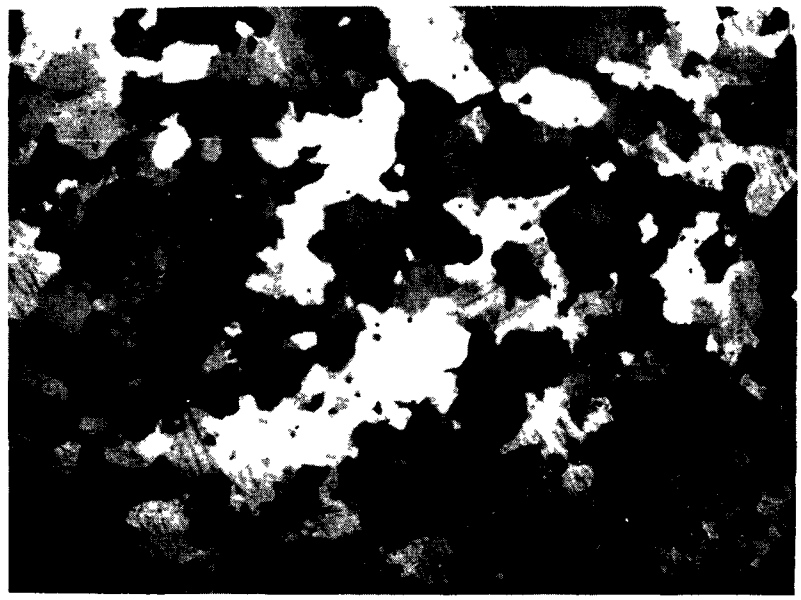
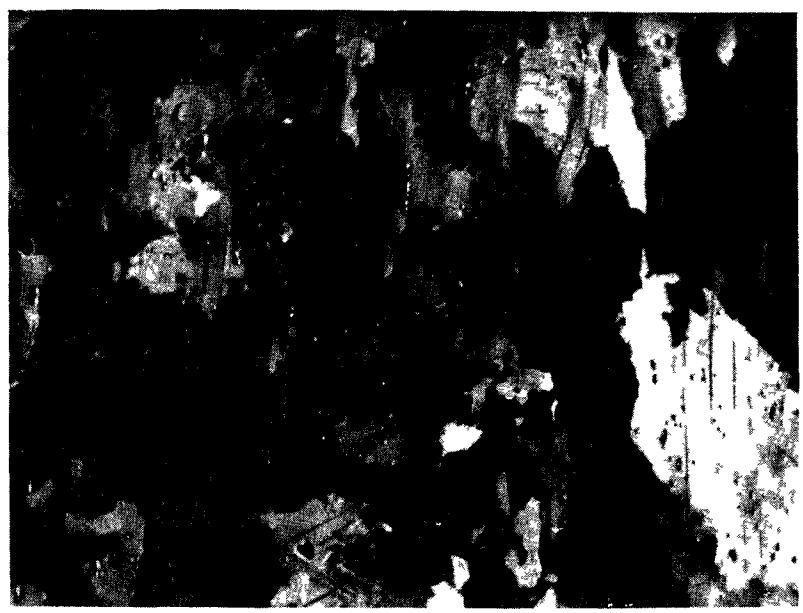


Fig. 8: Surface (top) and interior (bottom) of low carbon (50 p.p.m.) specimen water quenched from beta region (750°C.) and annealed in alpha region (650°C.) for 10 minutes. X75



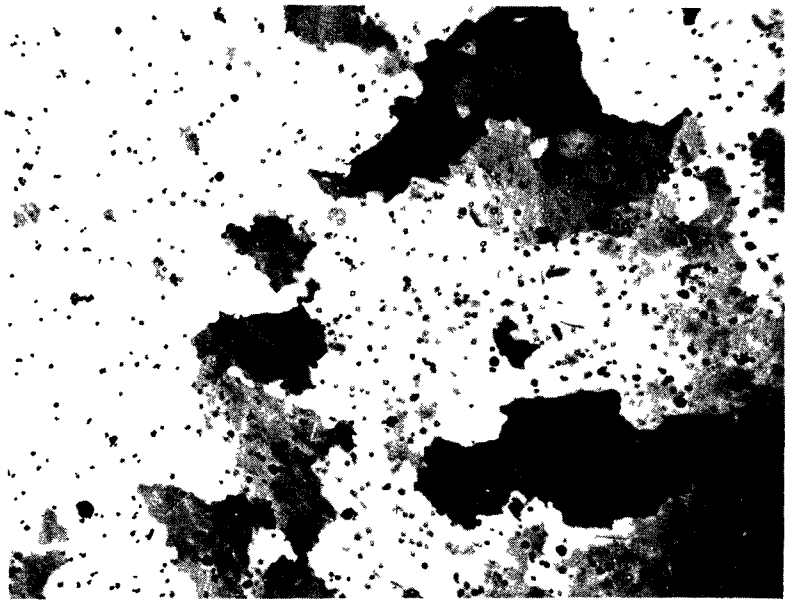
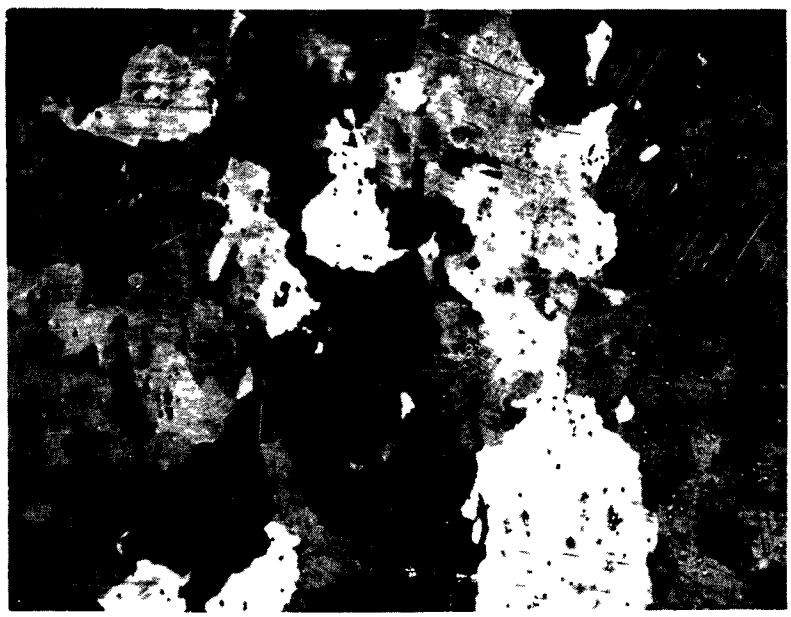


Fig. 9: Surface (top) and interior (bottom) of medium carbon (370 p.p.m.) specimen air cooled from beta region (750°C.). X75



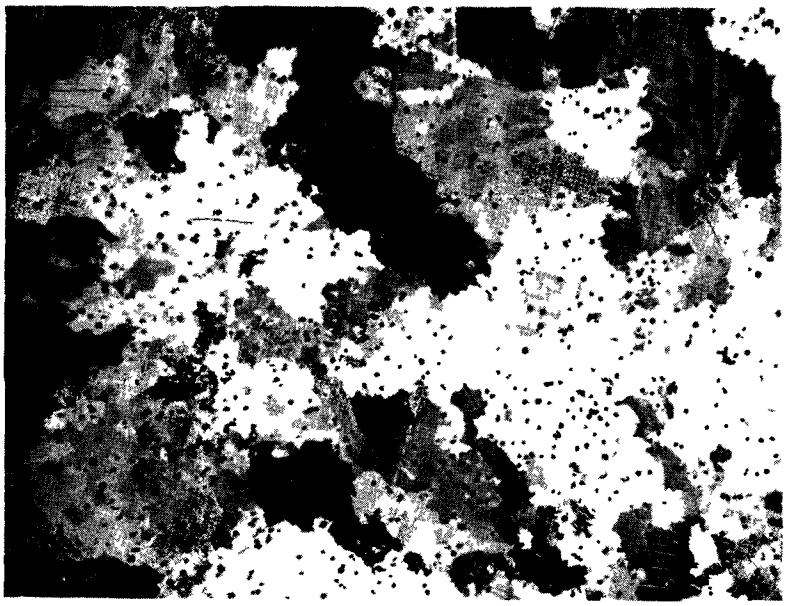
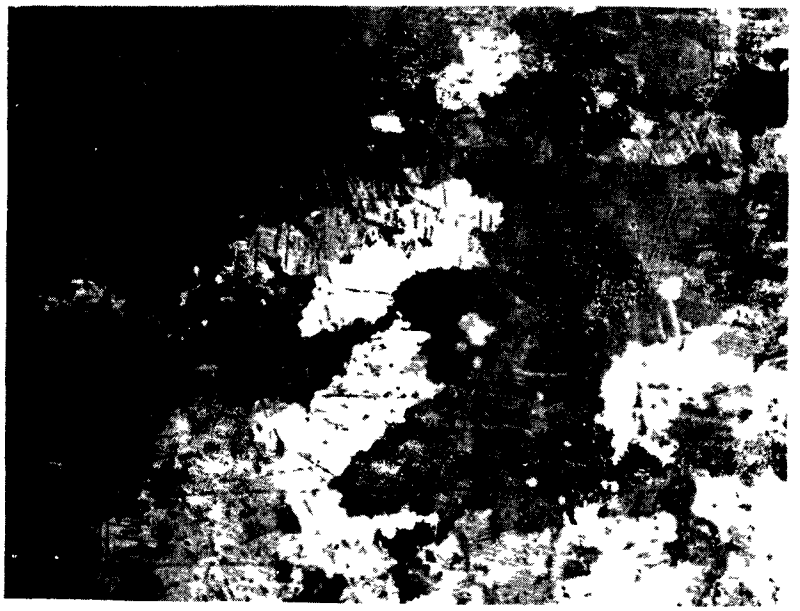


Fig. 10: Surface (top) and interior (bottom) of medium carbon (370 p.p.m.) specimen water quenched from beta region (750°C.). X75



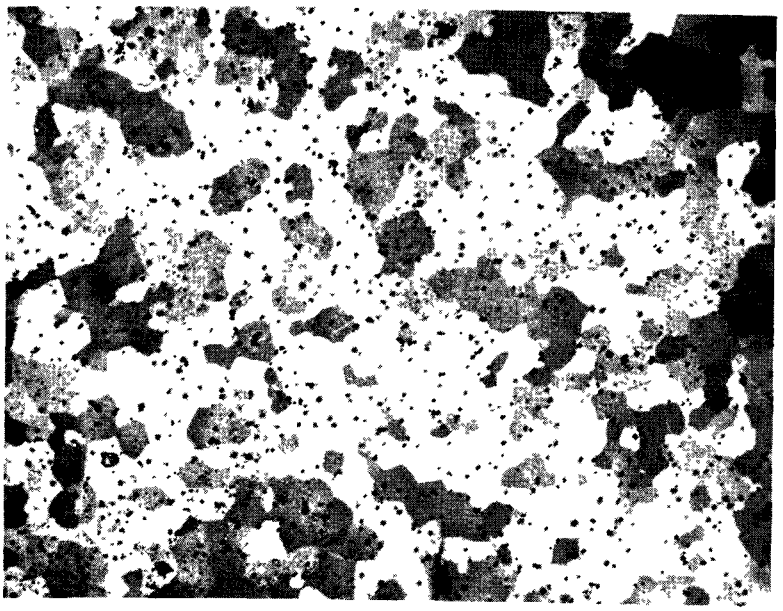
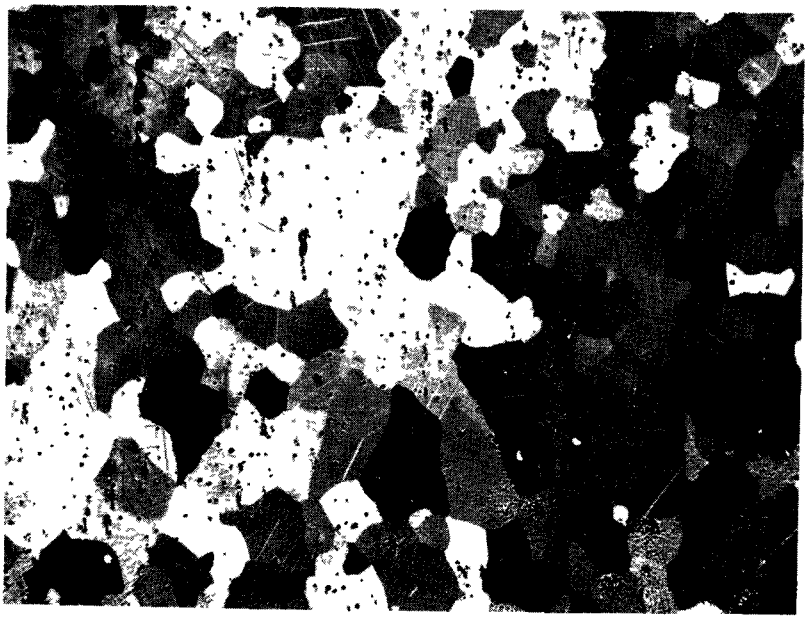


Fig. 11: Surface (top) and interior (bottom) of medium carbon (370 p.p.m.) specimen water quenched from beta region (750°C.) and annealed in alpha region (650°C) for 10 minutes.
X75



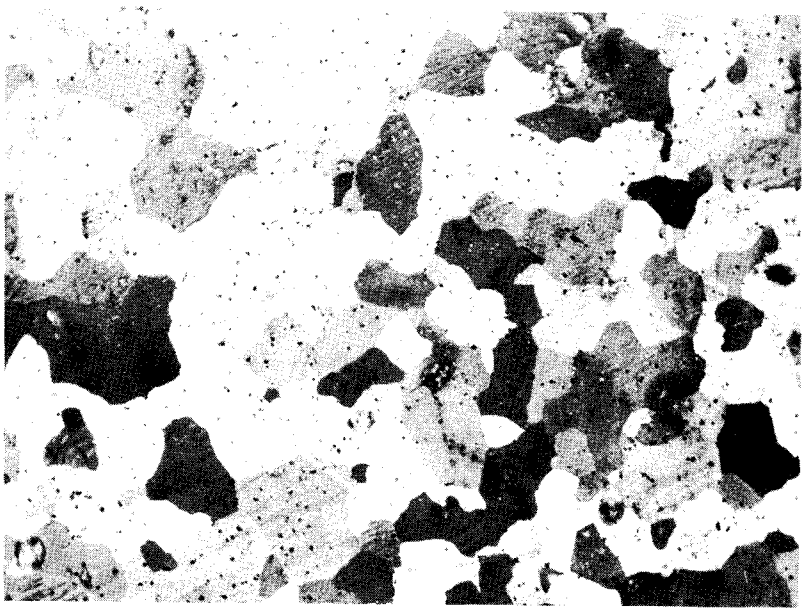


Fig. 12: Surface (top) and interior (bottom) of medium carbon (370 p.p.m.) specimen water quenched from beta region (750°C.) and annealed in alpha region (650°C.) for 20 minutes.
X75

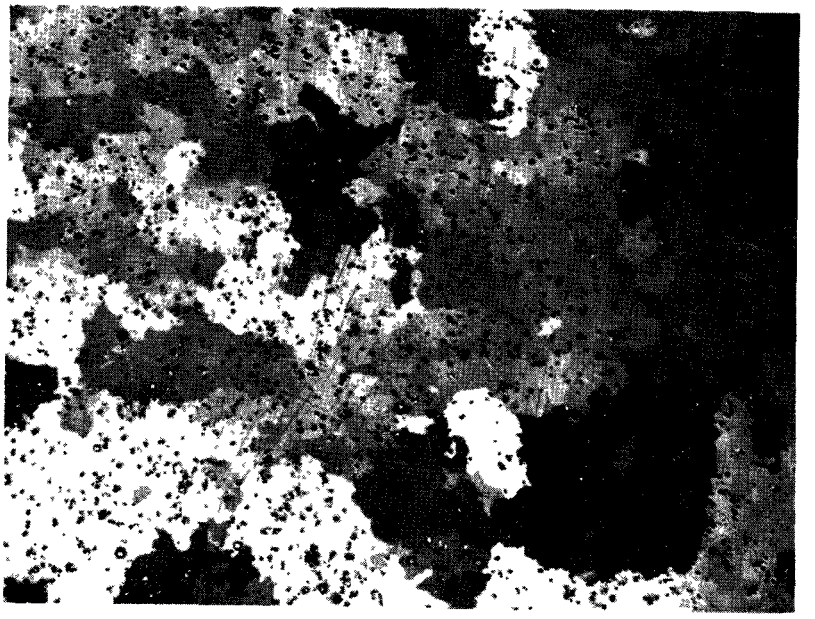
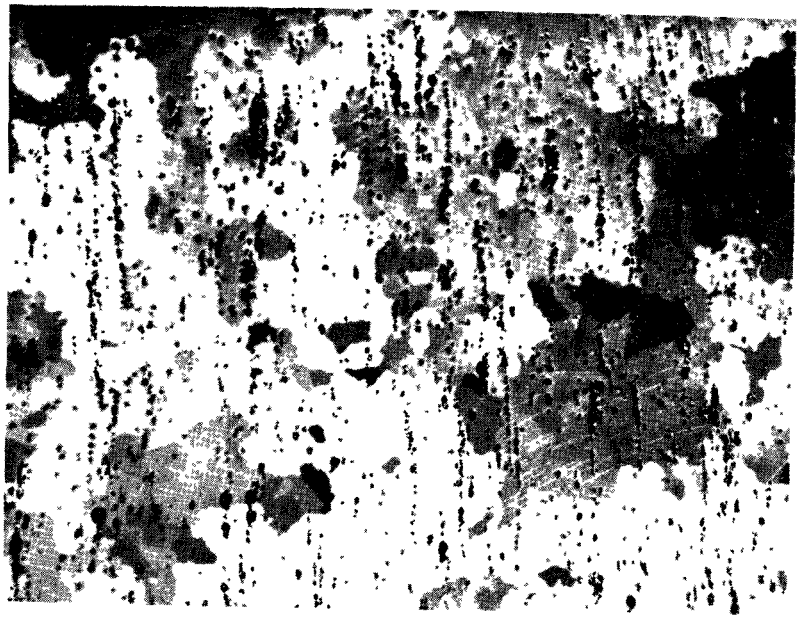


Fig. 13: Surface (top) and interior (bottom) of high carbon (1000 p.p.m.) specimen air cooled from beta region (750°C.). X75



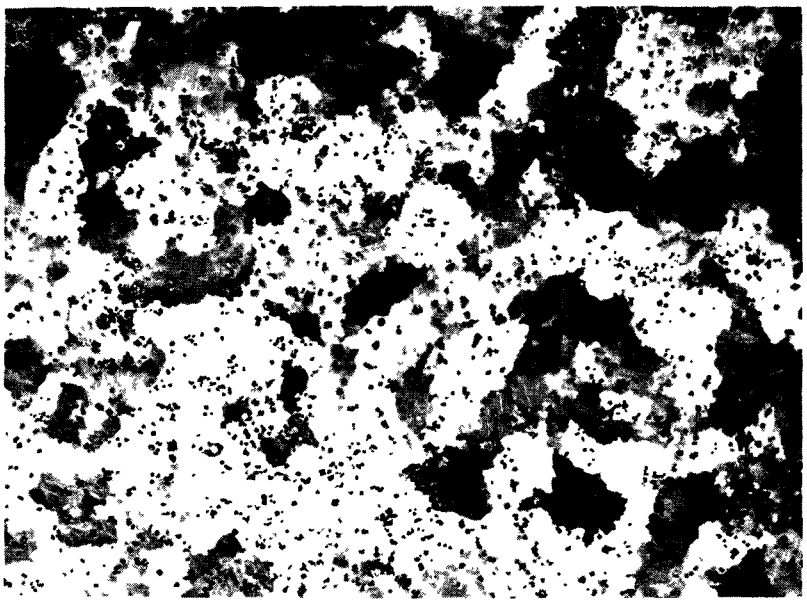


Fig. 14: Surface (top) and interior (bottom) of high carbon (1000 p.p.m.) specimen water quenched from beta region (750°C.). X75

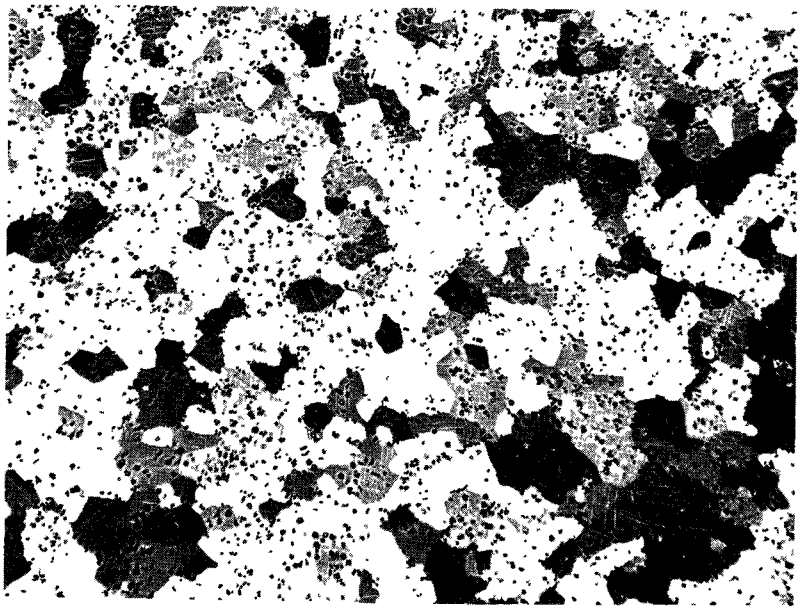


Fig. 15: Surface (top) and interior (bottom) of high carbon (1000 p.p.m.) specimen water quenched from beta region (750°C.) and annealed in alpha region (650°C.) for 10 minutes. X75