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The Behavior of Some Delta-Stabilized Plutonium Alloys at High Pressures*

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(Talk to be presented by R. O. Elliott)

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Place: Plutonium Physical Metallurgy Session.
Italian Room, St. Francis Hotel
2nd paper; 20 minutes

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Delta-phase plutonium can easily be retained at room temperature by the proper combination of alloying and heat treating. Alloys of this type are frequently referred to as delta-stabilized alloys. Actually, the delta phase in many such alloys is metastable and it will transform to a more stable state when heated or compressed. We have prepared a number of delta-stabilized plutonium alloys containing Al, Zn, and Ce, and have observed their behaviors at pressures up to 11,000 atmospheres. Many of these alloys transformed under compression, and their transformation pressures, transformation volume changes, and compressibilities were determined as a function of composition.

We made our alloys by melting in an induction furnace under vacua and casting into ingots about 1/2" O.D. x 1-3/4" long. These ingots were homogenized in the delta region at 450°C for at least 200 hours, air-quenched to room temperature, and, finally, machined to right cylinders 0.434" O.D. x 1.5" to 1.7" long.

The compression measurements were made in the simple piston and cylinder device shown in Slide 1. The stationary piston is placed in one end of the high-pressure cylinder, the specimen and glycerine are then inserted at the opposite end, followed by the insertion of the movable piston. Dial indicators are attached to the movable piston holder and guide and these indicators measure the linear displacement of the movable piston.

The apparatus is shown assembled in Slide 2. The force is applied to the movable piston with a 50 ton Lombard press. A Baldwin SR-4 load cell is used to measure the pressure, which is transmitted hydraulically to the specimen with the glycerine. Volume changes are not obtained directly from the dial indicator readings because as the pressure is generated the glycerine compresses, the cylinder expands and the pistons shorten. We corrected the data for the compression of glycerine with the values published by Bridgman, and for the expansion of the cylinder and shortening of the pistons by calculations using elastic theory.

The compression curves of seven Pu-Al delta-stabilized alloys, containing between 1.7 and 12.5 a/o Al, are shown in Slide 3. Irreversible transformations occur at 2040, 4480, and 7020 atms. in the 1.7, 2.5, and 3.4 a/o Al alloys, respectively. These transformations are due to changes of delta into either alpha phase or a phase mixture of mostly alpha and some beta. In general there remains some untransformed delta phase after compression. No transformations were found in the alloys containing from 4.0 to 12.5 a/o Al when compressed at pressures up to 10,600 atms. (The transformation pressures were obtained by extrapolating the pressure-volume curves at the transformation breaks; volumes of transformation are the percentage volume changes at the pressure of transformation.)

The transformation pressures and volumes of transformation of these Pu-Al alloys are shown in Slide 4 plotted against atomic per cent aluminum. They are seen to vary linearly with aluminum content over the range of the experimental measurements. Alloys containing 0.8 and 1.2 a/o Al were heat treated and examined. The 0.8 a/o Al alloy was mostly alpha phase plus some beta and gamma phases and had a density of 17.7 gm/cc. The 1.2 a/o Al alloy was delta phase and had a density of 15.7 gm/cc. The linear extrapolation to zero transformation pressure is interpreted, therefore, as determining the minimum amount of aluminum, 1 atomic per cent, which is required to retain delta phase at room temperature with the heat treatment being used.

The extrapolation to zero volume of transformation occurs at 4.7 a/o Al. There might be some tendency to interpret this composition as the Pu-rich boundary of the delta phase region on the Pu-Al phase diagram. This conclusion is not justified, however, because these data were at high pressures and the effect of pressure on this phase boundary is unknown.

The compression curves of four Pu-Zn alloys containing between 1.7 and 5.0 a/o Zn are shown in Slide 5. Our Pu-Zn alloy compositions are nominal and may be high by as much as 1.5 a/o. All of these Pu-Zn alloys transformed under compression. Their microstructures after compression showed predominantly alpha phase with some untransformed delta and traces of beta phases. Also, $PuZn_2$ was seen in the microstructure of the 5 a/o Zn alloy after compression. The transformations occur at 950, 2520, 4210, 5290, and 6820 atms. in the alloys containing 1.7, 2.5, 3.4, 4.0, and 5.0 a/o Zn, respectively.

The transformation pressures and volumes of transformation of the Pu-Zn alloys is shown in Slide 6. Again, these are seen to vary linearly with zinc content over the range of experimental measurements and extrapolation to zero transformation pressure occurs at 1 a/o Zn.

Slide 7 shows compression curves of Pu-Ce alloys containing between 3, 4 and 6.0 a/o Ce. All of these alloys undergo double, irreversible transformations under compression. The first, lower-pressure transformations occur at 60 or less, 500, 1490, and 1840 atms. in alloys containing 3.4, 4.0, 5.0 and 6.0 a/o Ce, respectively. The second, higher-pressure transformations occur at 3000, 4380, 6470, and 8220 atms., respectively.

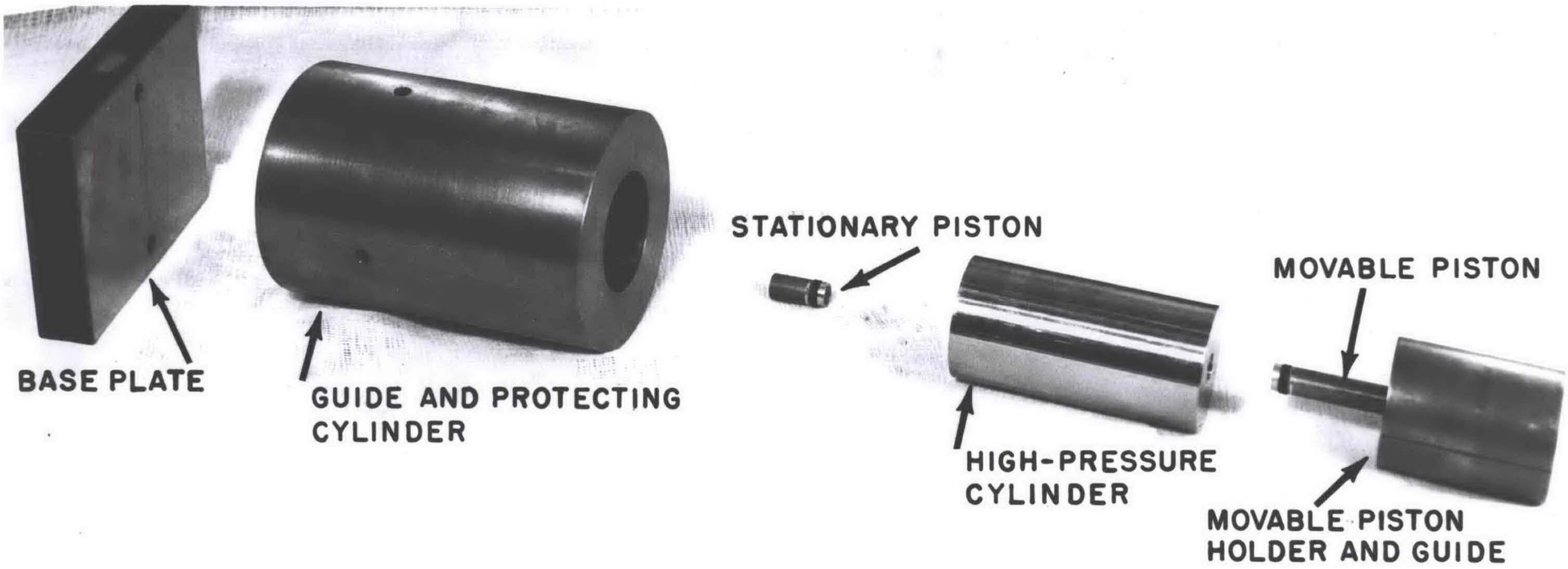
We have determined the nature of these double transformations in Pu-Ce alloys. Slide 8 shows the delta phase microstructure in a 4 a/o Ce alloy after heat treatment. Slide 9 shows a transformation structure in the same alloy after compression through the first, lower-pressure transformation, which was found to be irreversible delta-to-beta. This method can be used to retain beta phase plutonium at room temperature and to make beta phase alloys that are free of voids and micro-cracks, since the denser beta phase is caused to form under compression. Slide 10 shows the final transformation structure in the 4 a/o Ce alloy after compression through the second, higher-pressure transformation, which was found to be irreversible beta-to-alpha. This alloy is also free of voids and microcracks since the denser alpha phase was formed under compression.

The volumes of transformation for both the first and second transformations are shown in Slide 11 plotted against atomic per cent cerium. In both cases, the volumes of transformations vary linearly with cerium content. Slide 12 shows that the transformation pressures also vary linearly with atomic per cent cerium for both transformations. The extrapolations to zero transformation pressure occur at 3.4 a/o Ce for the first, lower- pressure transformation and at 1.9 a/o Ce for the second, higher-pressure transformation.

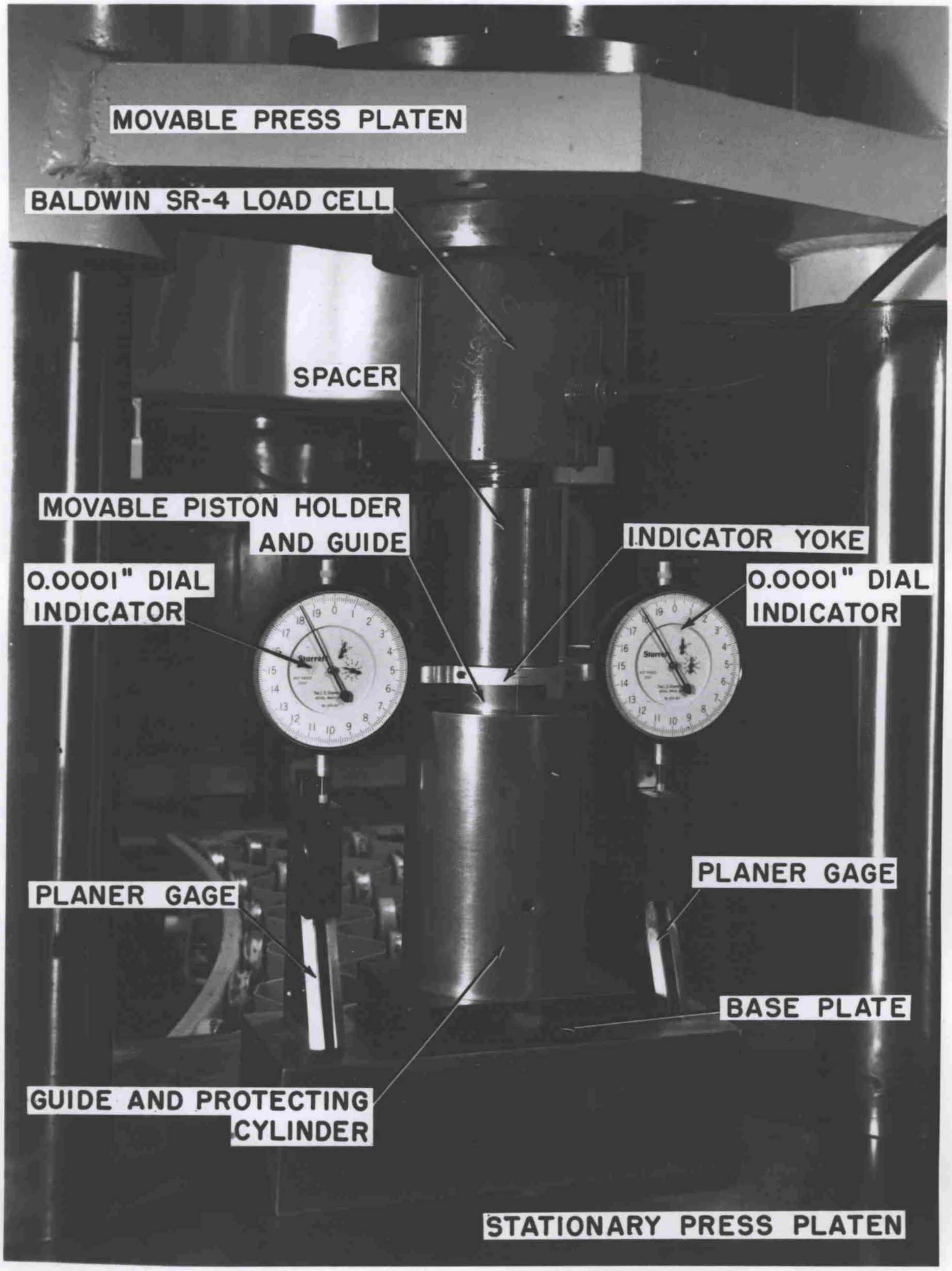
Slide 13 shows densities plotted against atomic per cent zinc, cerium, and aluminum. The lower curve on each figure shows the densities of the heat-treated alloys and the upper curve on each figure shows the densities of the same alloys after compression. The Pu-Al figure at the bottom of the slide shows that delta-stabilized alloys in this system will not transform at pressures below 10,600 atms., provided that they contain at least 4.5 a/o Al. The Pu-Zn and Pu-Ce figures, in the upper part of the slide, show that all of the alloys studied in these systems transform below 11,000 atms. Extrapolation of these data shows that delta phase containing at least 10.8 a/o Zn or 14.8 a/o Ce in solid solution would not be expected to transform.

The compressibilities of the Pu-Zn and Pu-Ce alloys increase linearly with increasing zinc or cerium contents at the approximate rate of 0.5×10^{-6} atm.$^{-1}$ per atomic per cent. The compressibilities of the Pu-Al alloys decrease linearly with increasing aluminum content at the approximate rate of 0.1×10^{-6} atm.$^{-1}$ per atomic per cent. We do not have a good value for the compressibility of pure delta plutonium at room temperature but extrapolation of our data gives a value near 3×10^{-6} atm.$^{-1}$

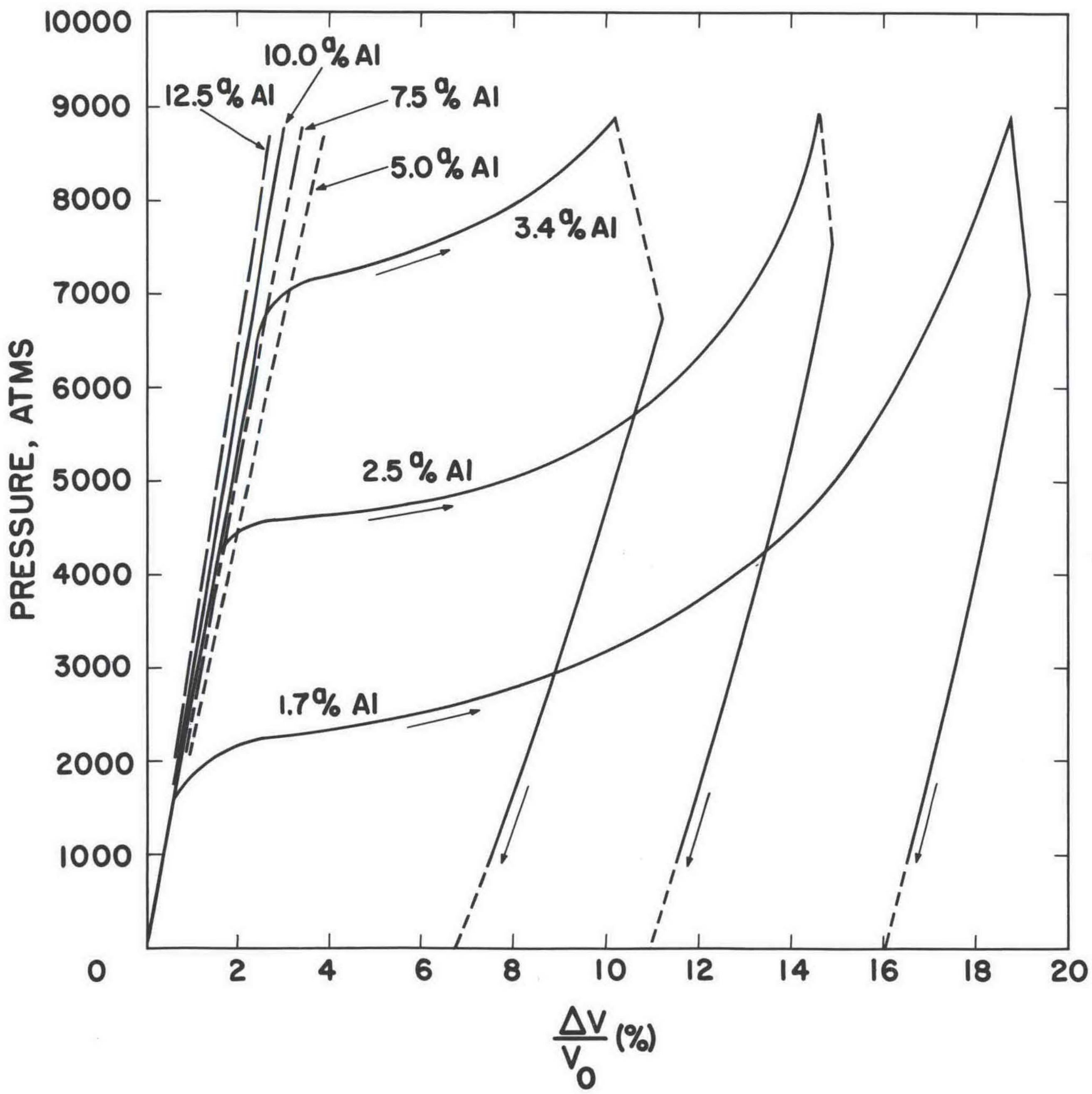
We have also made some experiments with delta-stabilized Pu-In alloys at high pressures but the results, so far, have not been meaningful because the alloys have not been homogeneous.



EXPLODED VIEW OF 10 KILOBAR HIGH-PRESSURE CHAMBER



PHOTOGRAPH OF 10 KILOBAR HIGH-PRESSURE APPARATUS



10000
9000
8000
7000
6000
5000
4000
3000
2000
1000
0
PRESSURE, ATMS
12.5% Al
10.0% Al
7.5 % Al
5.0% Al
3.4% Al
2.5 % Al
1.7% Al
2
4
6
8
10
12
14
16
18
20
$\frac{\Delta V}{V_0}$ (%)

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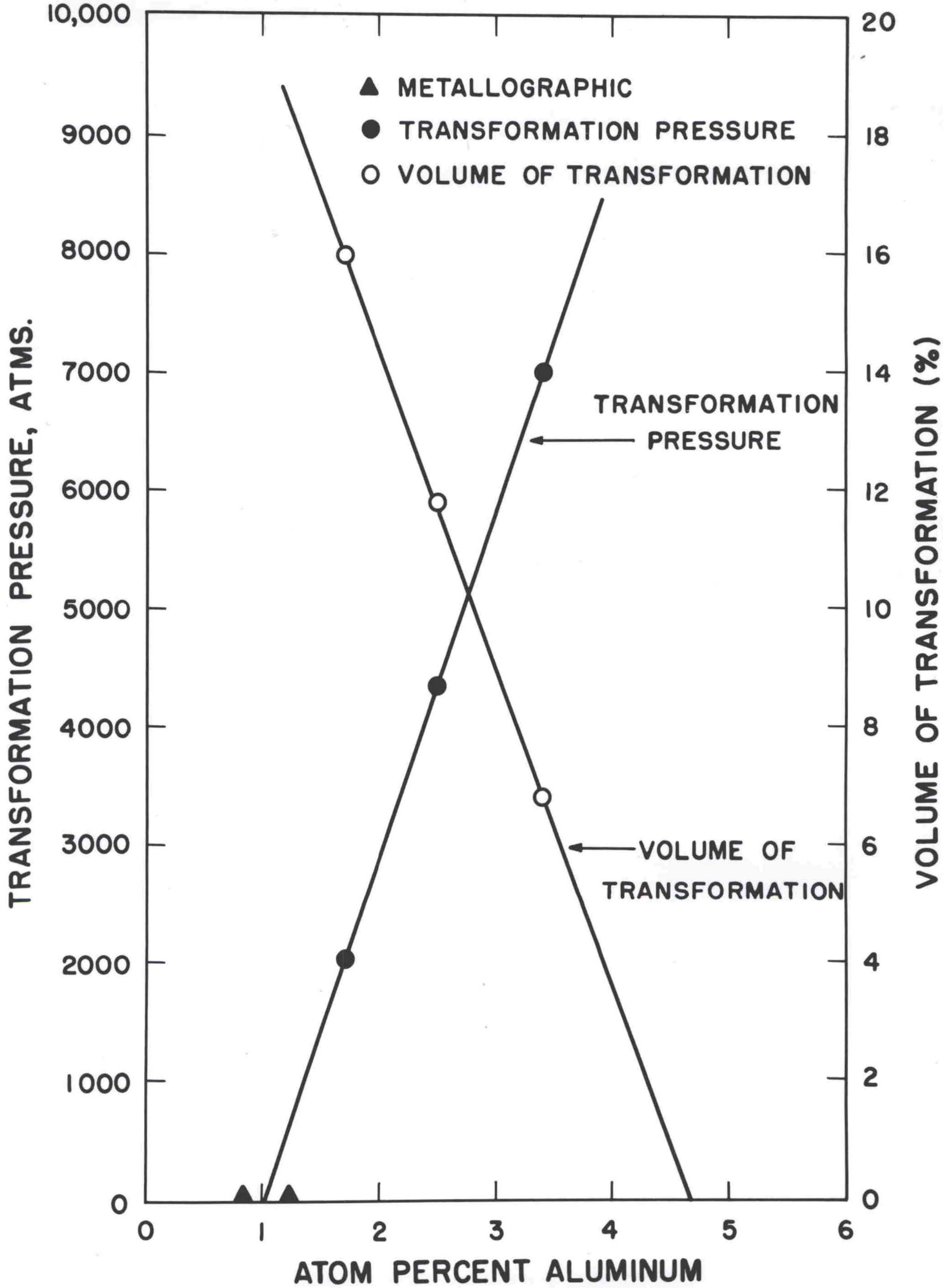
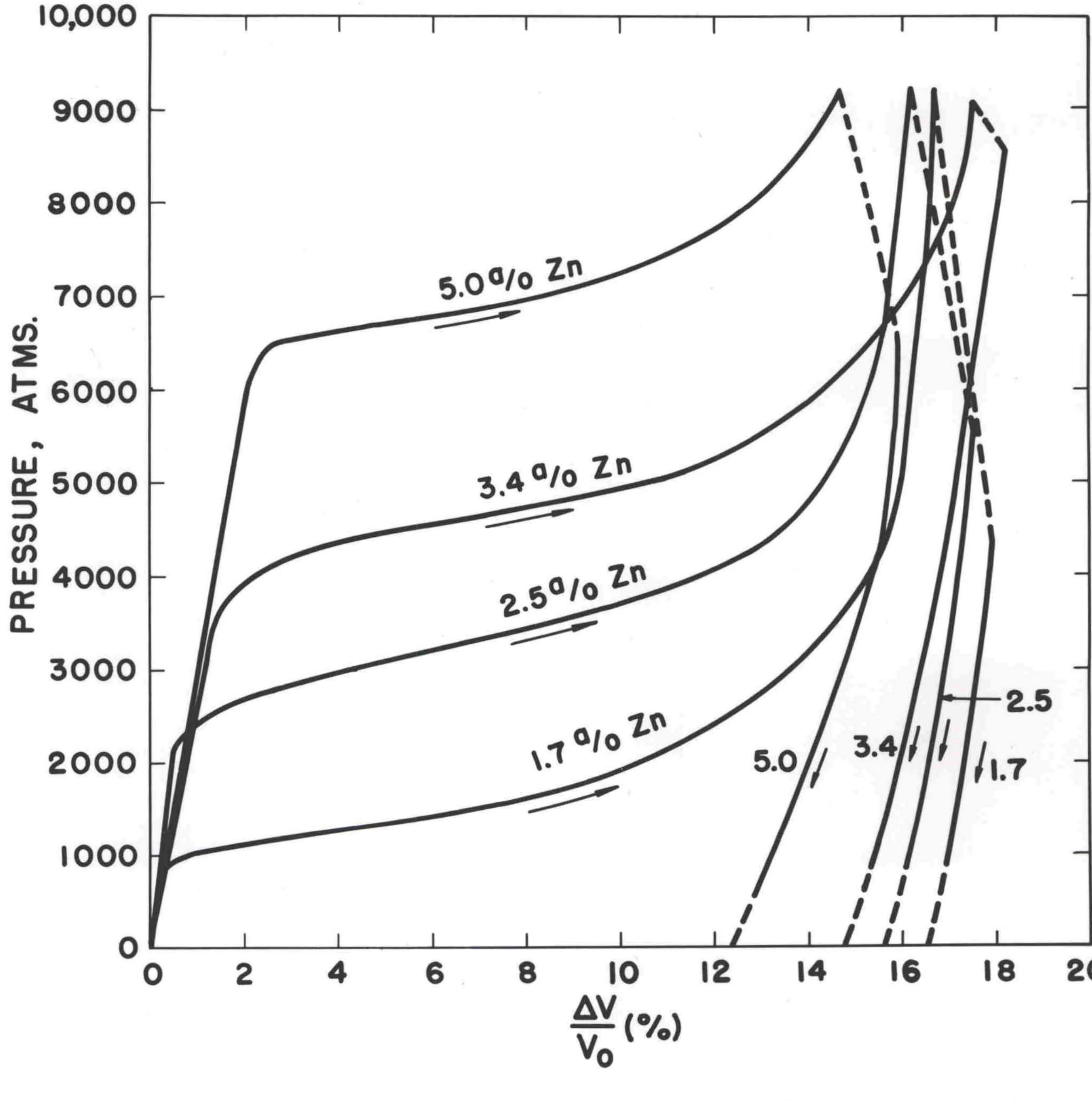
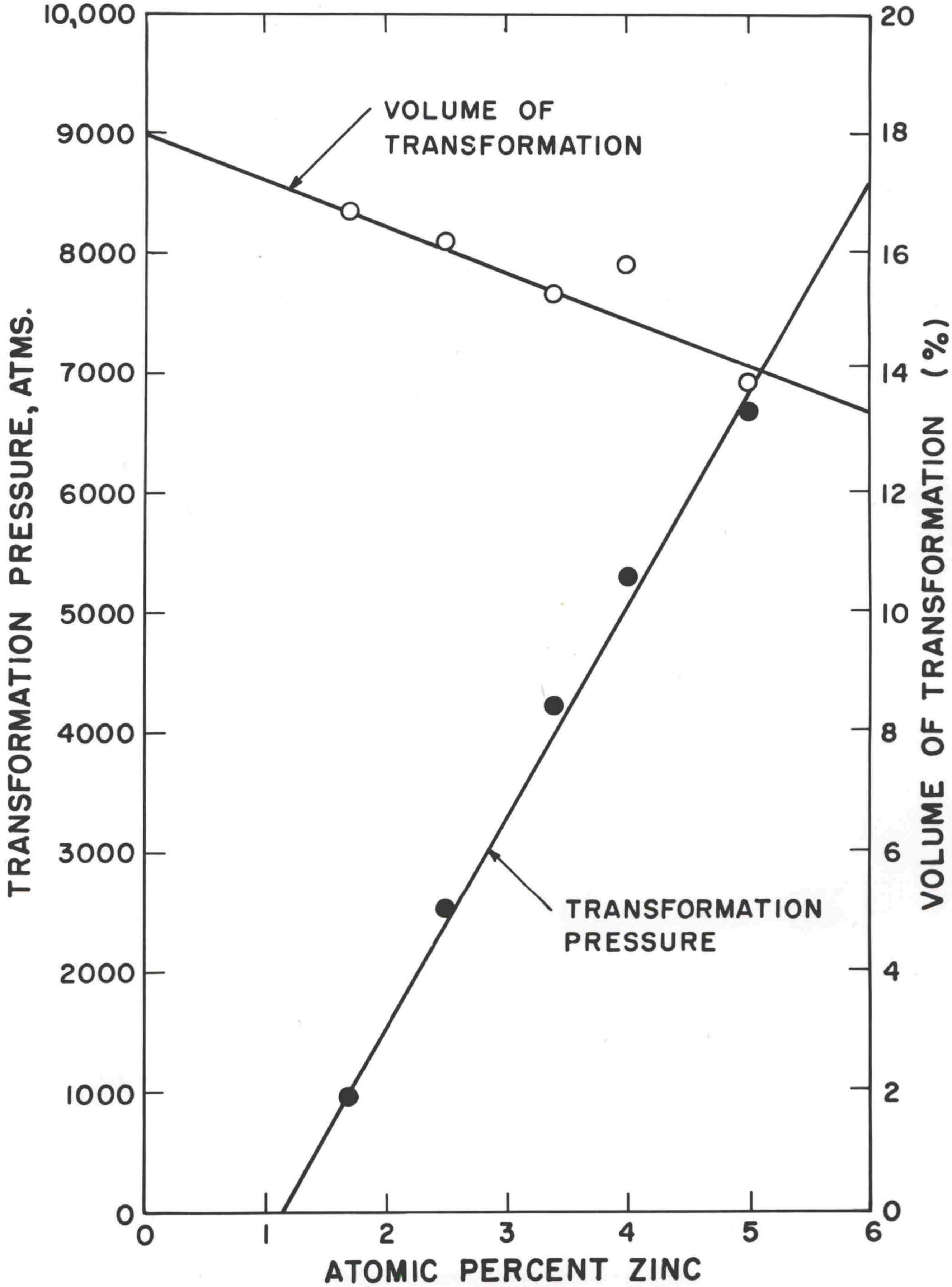
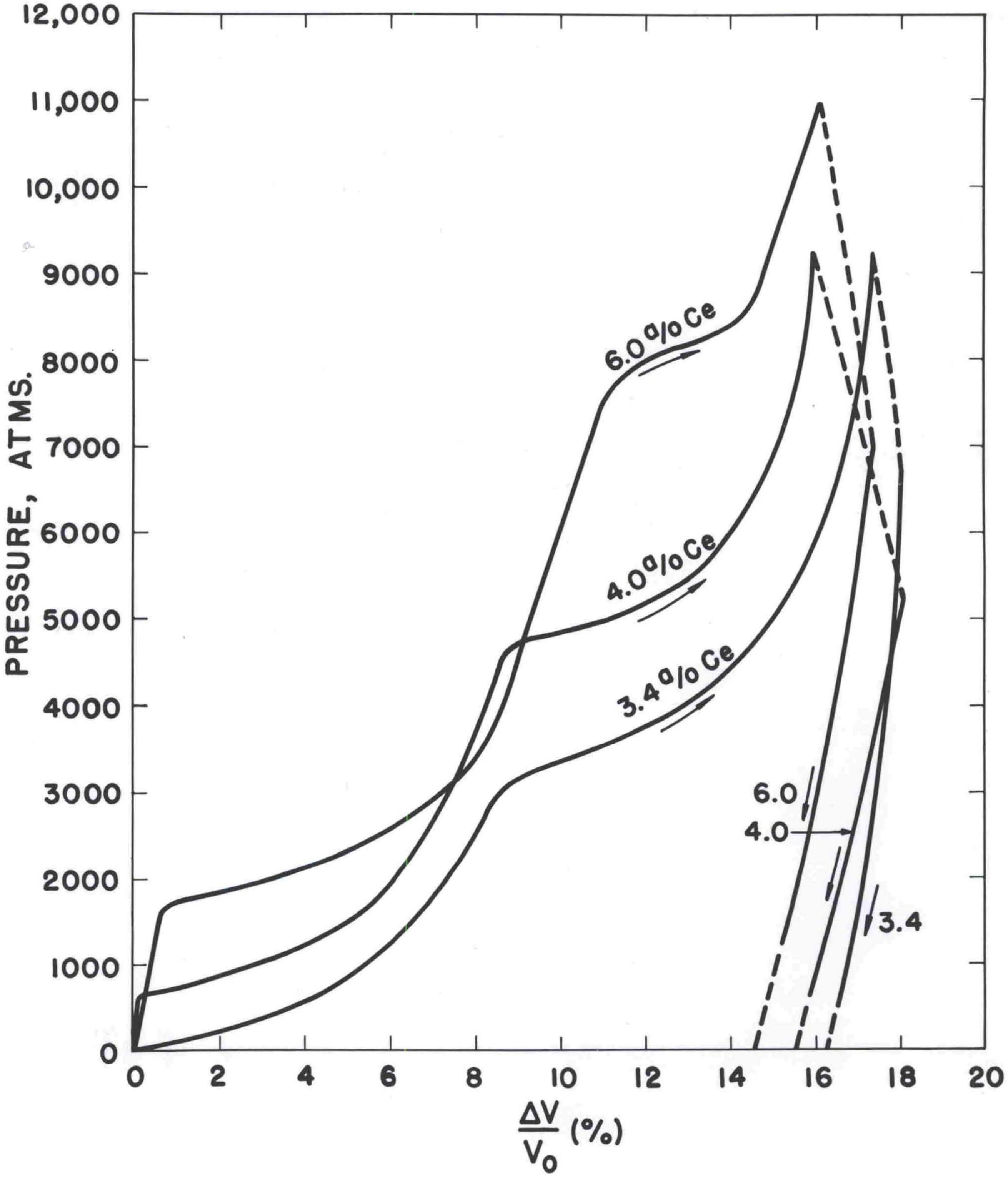


METALLOGRAPHIC
TRANSFORMATION PRESSURE
VOLUME OF TRANSFORMATION
TRANSFORMATION PRESSURE
VOLUME OF TRANSFORMATION
TRANSFORMATION PRESSURE, ATMS.
VOLUME OF TRANSFORMATION (%)
ATOM PERCENT ALUMINUM
10,000
9000
8000
7000
6000
5000
4000
3000
2000
1000
0
20
18
16
14
12
10
8
6
4
2
0
0
1
2
3
4
5
6

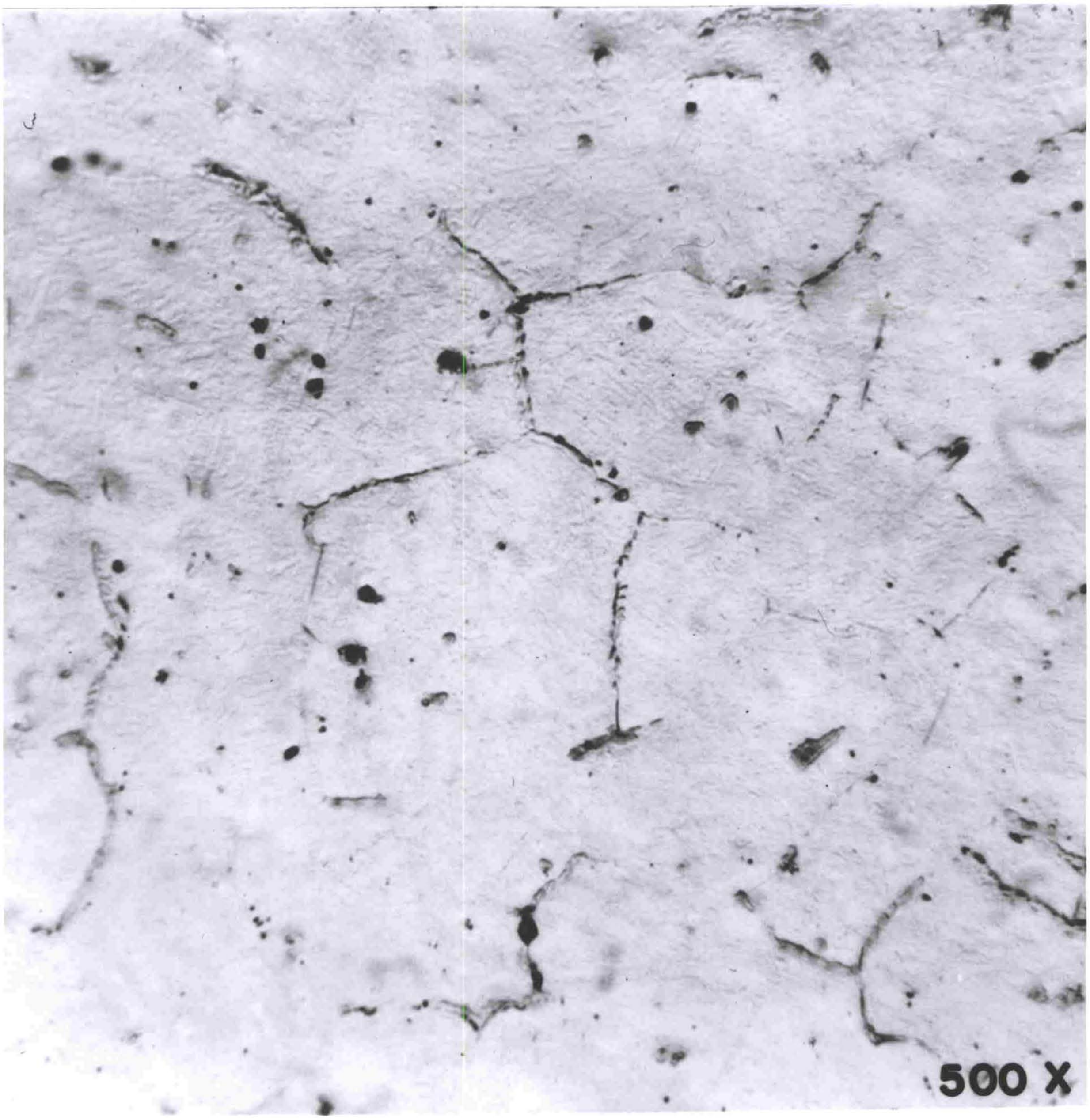

10,000
9000
8000
7000
6000
5000
4000
3000
2000
1000
0
PRESSURE, ATMS.
5.0 a/o Zn
3.4 a/o Zn
2.5 a/o Zn
1.7 a/o Zn
5.0
3.4
2.5
1.7
0
2
4
6
8
10
12
14
16
18
20
$\frac{\Delta V}{V_0}$ (%)



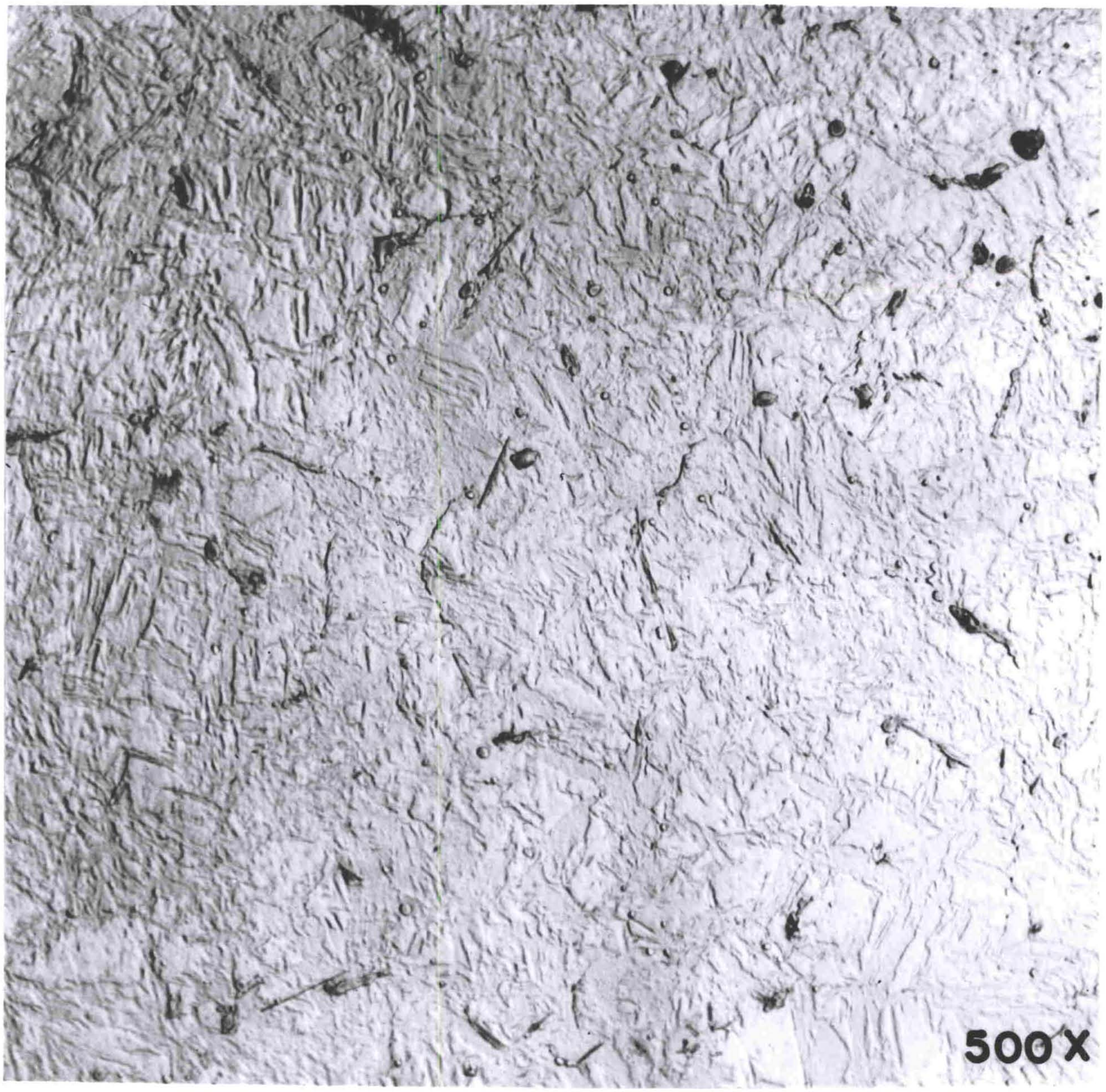
VOLUME OF
TRANSFORMATION
TRANSFORMATION
PRESSURE
TRANSFORMATION PRESSURE, ATMS.
VOLUME OF TRANSFORMATION (%)
ATOMIC PERCENT ZINC
10,000
9000
8000
7000
6000
5000
4000
3000
2000
1000
0
20
18
16
14
12
10
8
6
4
2
0
0
1
2
3
4
5
6



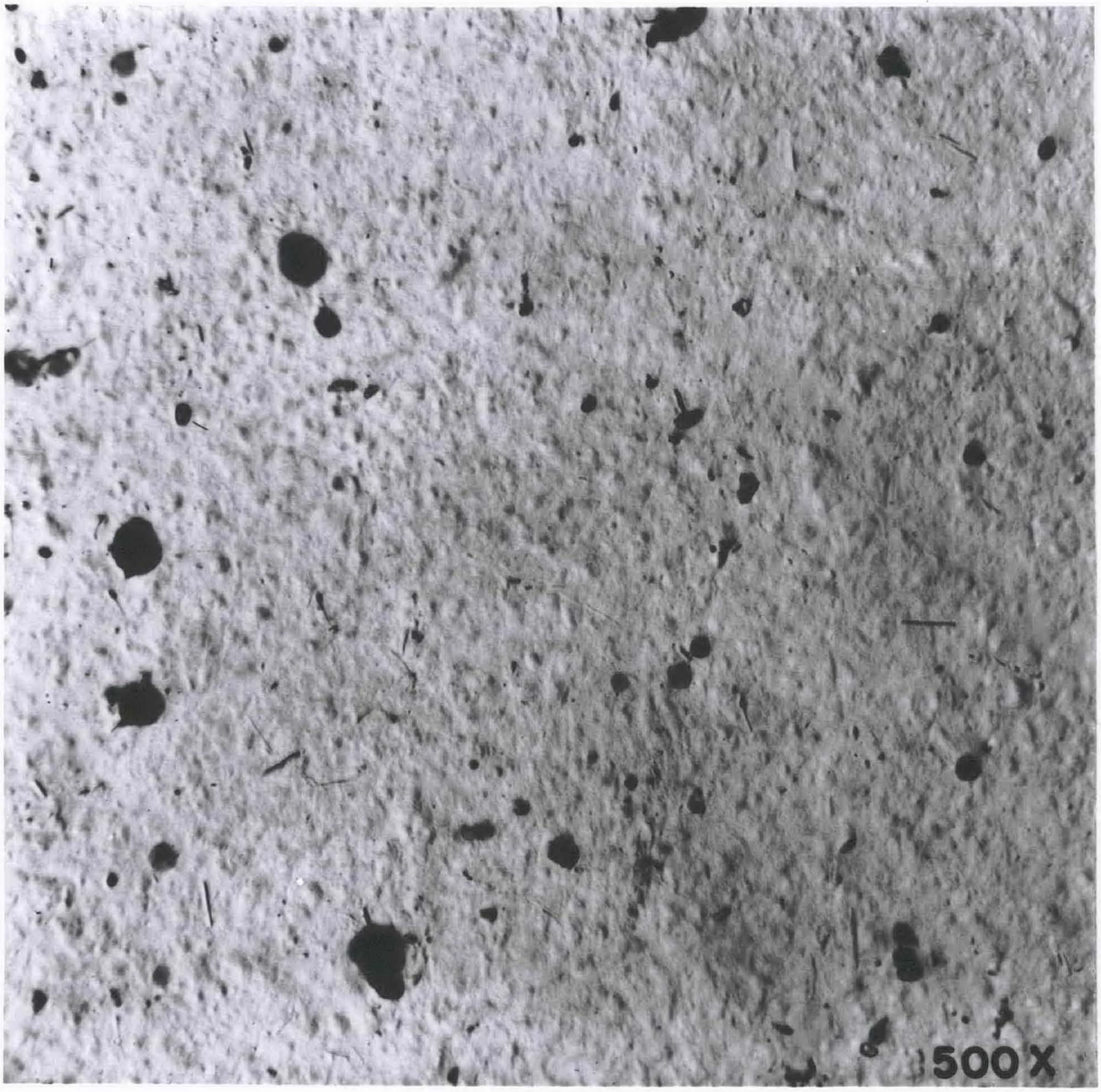
PRESSURE, ATMS.
12,000
11,000
10,000
9000
8000
7000
6000
5000
4000
3000
2000
1000
0
0
2
4
6
8
10
12
14
16
18
20
ΔV/V0 (%)
6.0 a/o Ce
4.0 a/o Ce
3.4 a/o Ce
6.0
4.0
3.4



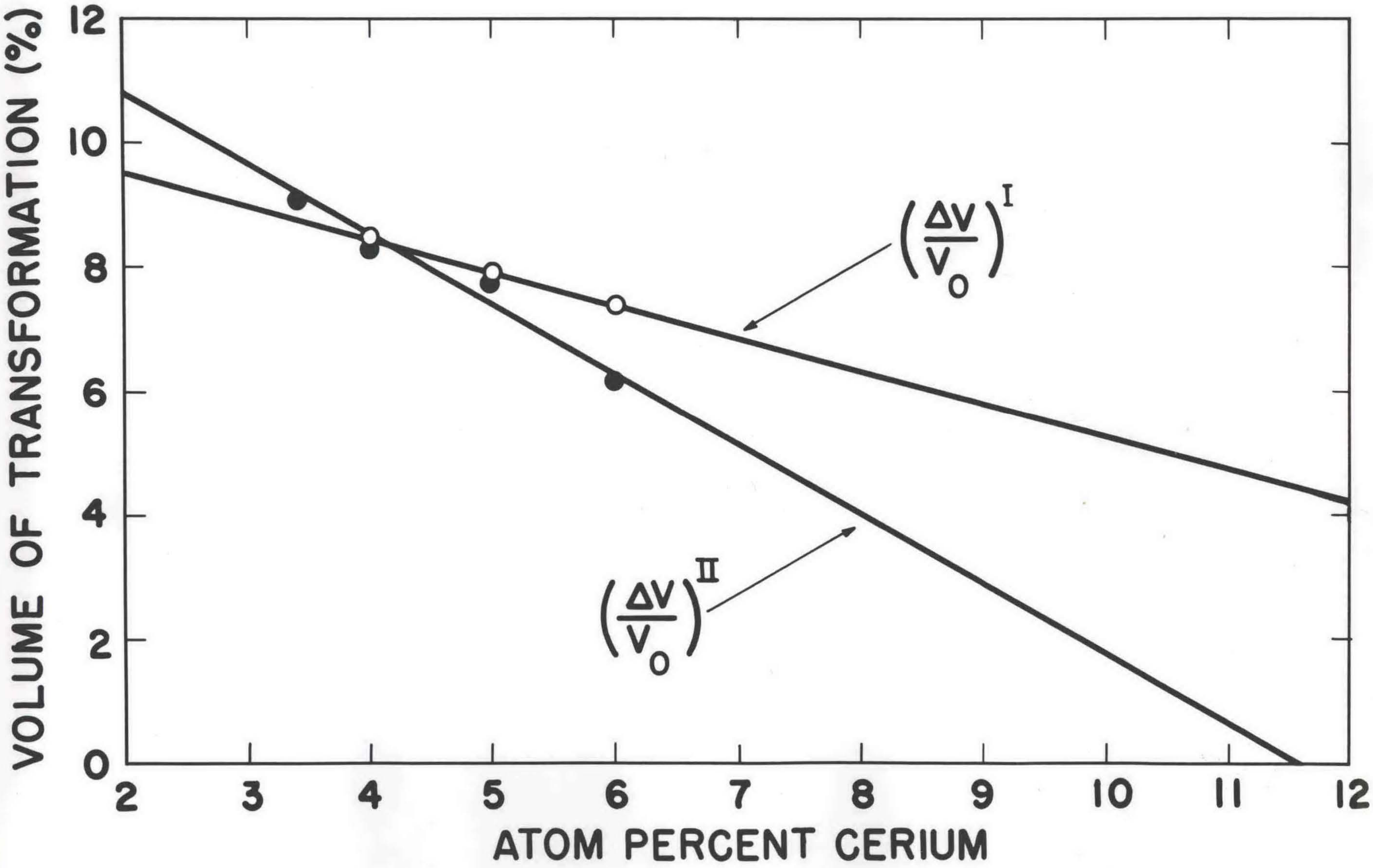
COMPOSITION : 4 a/o Ce — 96 a/o Pu
HEAT TREATMENT : 213 HRS / 450 °C / A.Q.
DENSITY : 15.56 gm/cc.
PHASES : 100 % DELTA



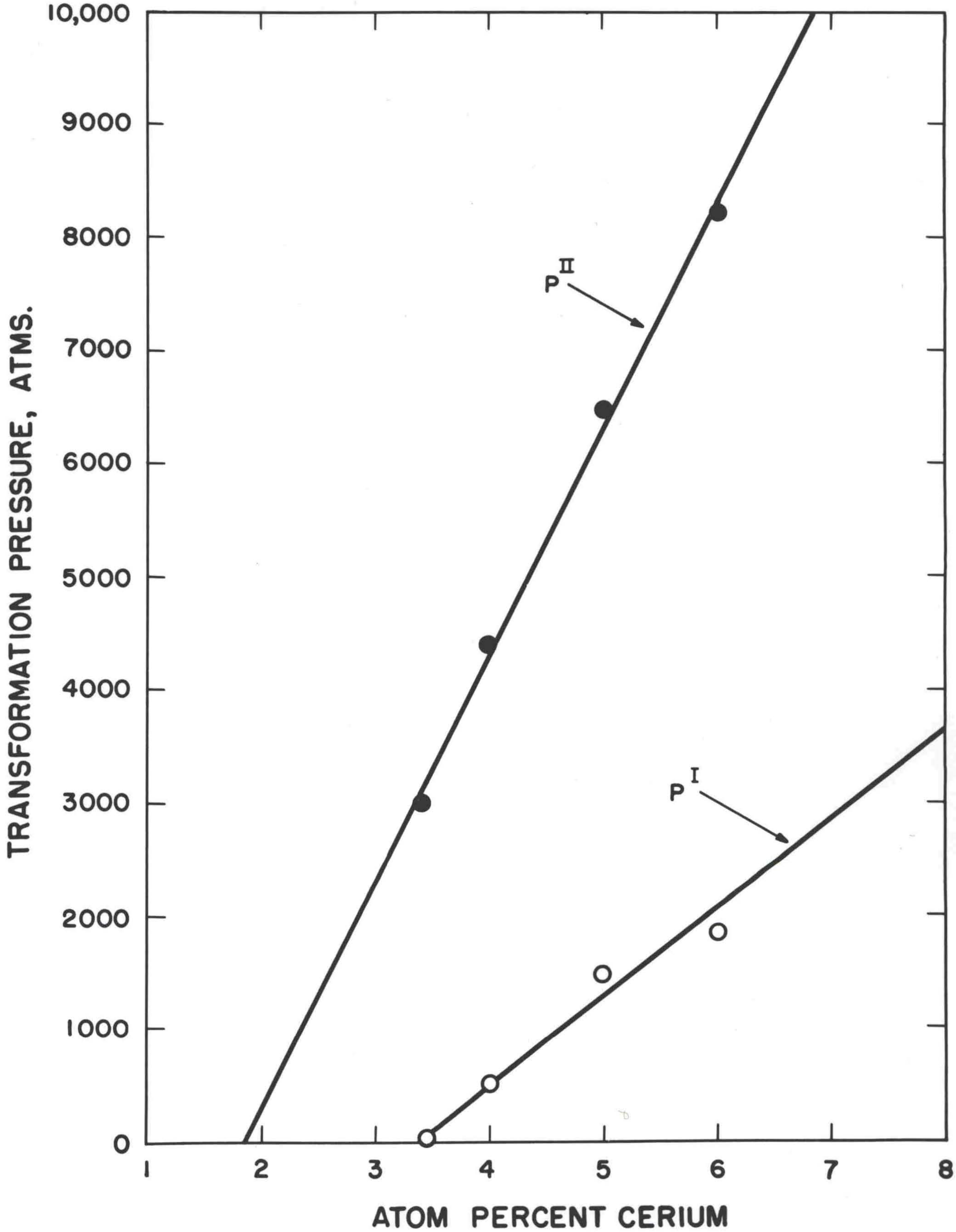
COMPOSITION: 4 a/o Ce – 96 a/o Pu
HEAT TREATMENT: 213 HRS/450°C/A.Q.
HISTORY: AFTER COMPRESSION TO 3,600 ATMS.
DENSITY: 16.97 gm/cc.
PHASES: 77% BETA + 14% DELTA + 9% GAMMA



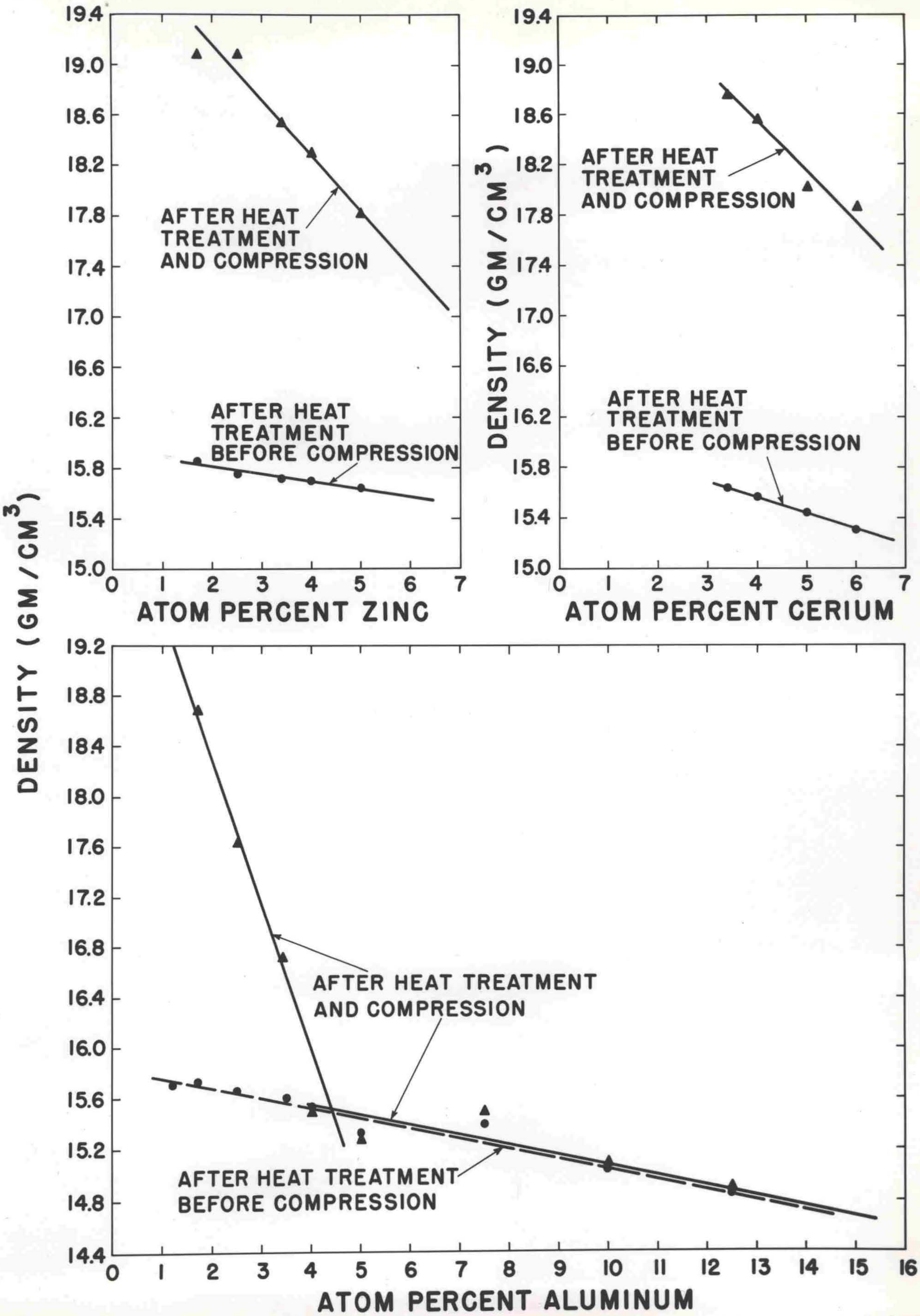
COMPOSITION: 4% Ce-96% Pu
HEAT TREATMENT: 213 HRS/450°C/A.Q.
HISTORY: AFTER COMPRESSION TO 9,250 ATMS.
DENSITY: 18.55 gm/cc.
PHASES: 77% ALPHA + 14% DELTA + 9% GAMMA



VOLUME OF TRANSFORMATION (%)
12
10
8
6
4
2
0
2
3
4
5
6
7
8
9
10
11
12
ATOM PERCENT CERIUM
$\left(\frac{\Delta V}{V_0}\right)^{I}$
$\left(\frac{\Delta V}{V_0}\right)^{II}$



10,000
9000
8000
7000
6000
5000
4000
3000
2000
1000
0
1
2
3
4
5
6
7
8
TRANSFORMATION PRESSURE, ATMS.
ATOM PERCENT CERIUM
P^II
P^I


DENSITY (GM/CM^3)
AFTER HEAT TREATMENT AND COMPRESSION
AFTER HEAT TREATMENT BEFORE COMPRESSION
ATOM PERCENT ZINC
AFTER HEAT TREATMENT AND COMPRESSION
AFTER HEAT TREATMENT BEFORE COMPRESSION
ATOM PERCENT CERIUM
AFTER HEAT TREATMENT AND COMPRESSION
AFTER HEAT TREATMENT BEFORE COMPRESSION
ATOM PERCENT ALUMINUM