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AEC RESEARCH AND
DEVELOPMENT REPORT

DEVELOPMENT OF HIGH STRENGTH COLUMBIUM AND TANTALUM ALLOY TUBING

SECOND QUARTERLY PROGRESS REPORT

MARCH 1, 1962 TO MAY 31, 1963

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UC-25, Metallurgy and Ceramics
(TID-4500, 20th Edition)

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June 1 - August 31, 1963

March 1, 1962 - May 31, 1963

R. W. Buckman, Jr.

Contract AT(30-1)3108
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Development of High Strength Columbium and Tantalum Alloy Tubing

Second Quarterly Progress Report

by

R. W. Buckman, Jr.

ABSTRACT

The evaluation of the extrudability of B-66 (Cb-5V-5Mo-1Zr) and T-111 (Ta-8W-2Hf) at a reduction ratio of 7:1 was accomplished. An evacuated mild steel can was used as cladding for the extrusion billets. Both B-66 billets were extruded at 2200°F, using ram speeds of 275 inches per minute and 1000 inches per minute. The billet extruded at a ram speed of 275 inches per minute exhibited slight rattlesnaking for 6 inches near the nose end, with the balance of the as-extruded surface smooth and free of defects. Increasing the ram speed to 1000 inches per minute resulted in moderate rattlesnaking and circumferential cracking the entire length of the billet. The depth of the cracks was approximately .020 - .040 inches. The extrusion constant for B-66, based on the running pressure, was 78 - 84 ksi.

The initial T-111 billet was extruded at 2200°F and at a ram speed of 175 inches per minute. The as-extruded surface was smooth and free of defects. Increasing the temperature to 2300°F and the ram speed to 1000 inches per minute resulted in slight rattlesnaking over a 6-inch length approximately 6 inches from the nose end. The balance of the extrusion was free of defects. The increase in extrusion temperature and ram

speed decreased the extrusion constant for T-111 from 84 ksi at 2200°F to 80 ksi at 2300°F.

Results of the extrusion evaluation, using solid billets, indicate that the extrusion of tubular billets of B-66 and T-111 at 7:1 reduction ratios is feasible using the mild steel cladding technique.

A portion of the B-66 solid rod extrusion was conditioned and after heat treatment will be gun drilled. This starting tubular shape will be used to evaluate the hot drawing characteristics of B-66.

During the next reporting period, the tube blank extrusions will be completed and the initial processing to tubing will commence.

SUMMARY

This is the second quarterly progress report on AEC contract AT(30-1)3108, "Development of High Strength Columbium and Tantalum Alloy Tubing." The results of the extrusion evaluation of B-66 and T-111 at a reduction ratio of 7:1 are presented, and selection of the parameters for the Phase II tube blank extrusion is discussed. The results of the metallurgical study for selection of the starting microstructures for the extrusion billets are also presented.

SECOND QUARTERLY PROGRESS REPORT ON DEVELOPMENT OF HIGH STRENGTH COLUMBIUM AND TANTALUM ALLOY TUBING

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I. INTRODUCTION

During the second quarterly period, the T-111 starting material for the Phase II extrusion billets was received. This completed the procurement of all starting material required for the contract effort. Extrusion of the Phase I billets was accomplished and parameters for the Phase II tube blank extrusions were selected. The extrusion was done at Nuclear Metals, Inc., under the direction of Mr. W. Toeffin. Heat treatment studies were conducted on the "as-received" starting billet material, to permit selection of a thermal treatment which would produce an optimum metallurgical starting condition for the Phase II billet material and to approximate this microstructure in the Phase I billet material. Phase I extrusion billet B-66-1 was hot straightened and lathe conditioned. After heat treatment, this billet will be rifle drilled and used for hot tube drawing evaluation. The metallurgical evaluation of the Phase I extrusions is in progress.

II. PROGRAM STATUS

A. Extrusion Billet Preparation

T-111 starting material, heat DX-606, is being machined to the configuration[1] required for the Phase II tube blank extrusion billets. The ingot analysis for heat DX-606 is given in Table I. Heat DX-606 has a higher average total solute (W + Hf)

TABLE I - Ingot Analysis of T-111, Heat DX-606

Ingot Position	Analysis, weight per cent				
	W	Hf	C	N	O
Top - mid radius	8.3	2.2	.001	.0023	.0027
Bottom - mid radius	8.5	2.4	.001	.0018	.0025

content than heat DX-571, which was used for the Phase I extrusion evaluation. However, it is not expected that the difference in solute content 10.7 w/o vs. 9.98 w/o, will cause the processing characteristics of DX-606 to differ significantly from DX-571.

Both DX-571 and DX-606 were processed from 8-inch diameter ingots which were prepared by double vacuum arc melting electron beam melted starting stock. Schedules for the processing of the arc cast ingots to the required starting billet sizes are outlined in Figures 1[2] and 2[2].

A slice was taken from the starting billet material of heat DX-606 and was used to determine a heat treatment which would produce an "as fine as possible" recrystallized grain size. The as-received billet material had a duplex microstructure consisting of areas of elongated grains and areas exhibiting complete recrystallization, as shown in Figure 3. Primary breakdown of this ingot was by extrusion (See Figure 1). The billet temperature of 3000°F was above the recrystallization temperature of T-111, thus, recrystallization occurred during extrusion and/or while cooling from the extrusion operation. The extrusion ratio of 2.58:1 was insufficient to cause homogeneous deformation during the extrusion process as evidenced by the preferential location of the recrystallized grains. After conditioning of the extrusion by grinding, it was swage forged to approximately 4-1/2 inches in diameter at 1200°C (2200°F). The billet was heated in an argon purged Inconel retort. The small reduction during forging (approximately 10%) was insufficient to break up the recrystallized grains which were present in the extrusion (See Figure 3).

Annealing the extruded and forged heat DX-606 billet for 1 hour at 1600°C (2900°F) produced a fully recrystallized microstructure. As shown in Figure 4, the grain size resulting from this treatment was mixed, some areas having a fairly fine grain size (ASTM 5-6) and adjacent areas having a grain size of about ASTM 3. Annealing DX-606 for 1 hour at 1500°C (2700°F) resulted in about 95% recrystallization as determined by metallographic examination. The Phase II T-111 starting billets were annealed for 1 hour at 1600°C (2900°F) which was considered a satisfactory thermal treatment for producing an "as fine as possible" recrystallized grain size.

The T-111 Phase II billets were dye penetrant inspected prior to final machining, and no evidence of surface defects was found. However, ultrasonic

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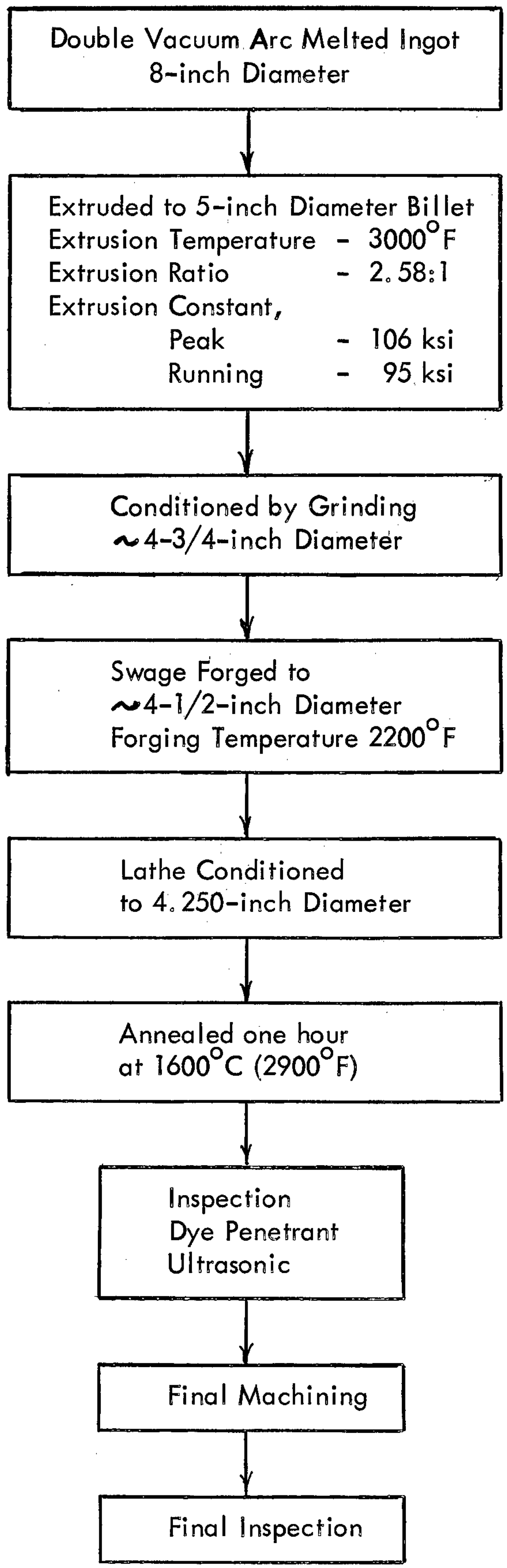

Figure 1 - Processing Schedule for Phase II T-111 Extrusion Billet Material, Heat DX-606

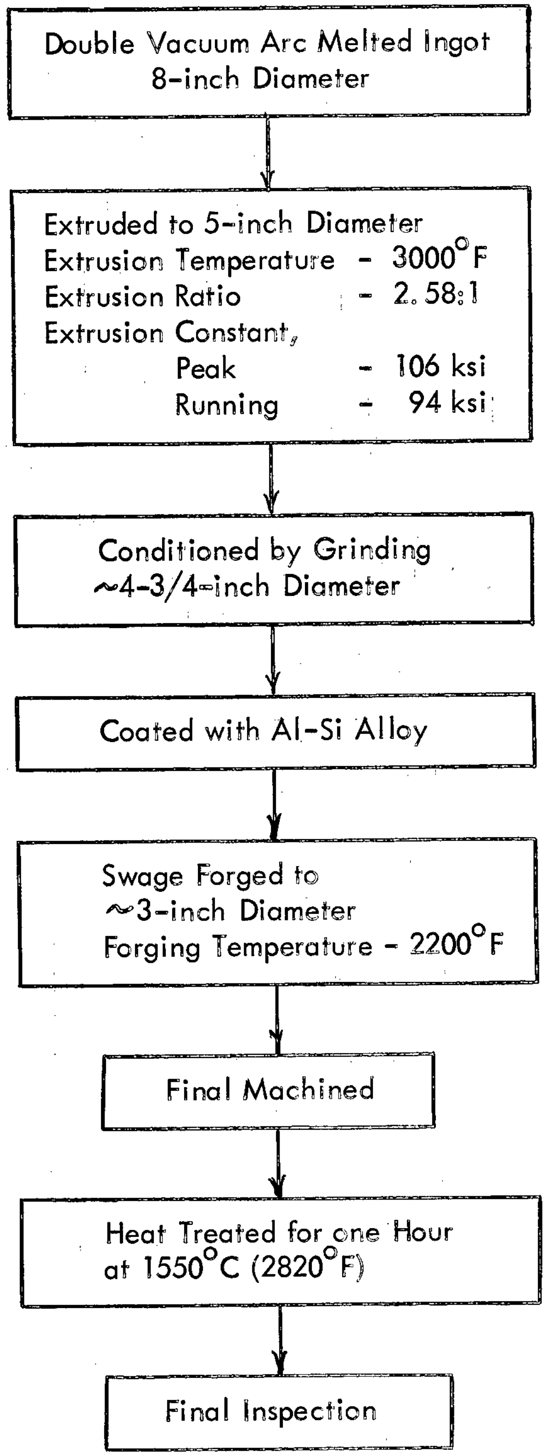

Figure 2 - Processing Schedule for Phase I T-111 Extrusion Billet Material, Heat DX-571

Figure 3 - Microstructure of "As-Received" Phase II T-111 Extrusion Billet Material, Heat DX-606, 100X

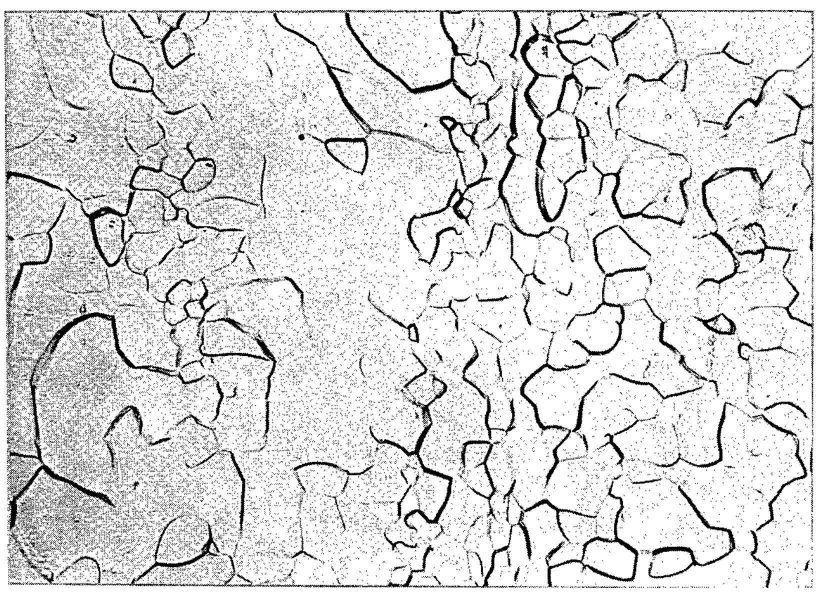

Figure 4 - Microstructure of Phase II T-111, Extrusion Billet Material after Annealing One Hour at 1600°C (2900°F), Heat DX-606, 100X

inspection revealed internal defects in three of the billets. The size and location of the defects are given in Table II.

TABLE II - Location of Ultrasonic Indications, Phase II
T-111 Billet Material, Heat DX-606

Identification	Defect Size	Location
DX-606-II-1	None	--
DX-606-II-2	~.027" dia.	center of billet
DX-606-II-3	~.059" dia.	~1" from OD and 1-3/4" from end
DX-606-II-4	~.077" dia.	center of billet
	~.039" dia.	~1" from OD and 4" from end

The centrally located defects will be removed during the final machining of the tube blank extrusion billet. There will be two starting billets which will each have a defect at approximately mid radius of the tubular starting billet. It is not anticipated that these defects will have a deleterious effect during extrusion.

T-111 Phase I extrusion billets were taken from heat DX-571 (Figure 2). The composition of Heat DX-571 was given in Reference 1, and as discussed earlier, is similar to DX-606. A 1/4-inch thick slice, taken from the as-forged billet stock, was used to determine the effect of annealing temperature on microstructure. The purpose of this heat treatment study was to establish the treatment required to produce a microstructure in the Phase I billets similar to that in the Phase II billet material. After heating for one hour at 1500°C (2700°F) a completely recrystallized microstructure with a grain size of ASTM 4-6 was achieved. After annealing for 1 hour at 1600°C (2900°F) the recrystallized grain size also was approximately ASTM 4-6. The microstructure for the DX-571 after one hour at 1600°C is shown in Figure 5. Based on the results of this heat treatment study, the Phase I T-111 extrusion billets were annealed for 1 hour at 1550°C (2800°F) prior to extrusion.

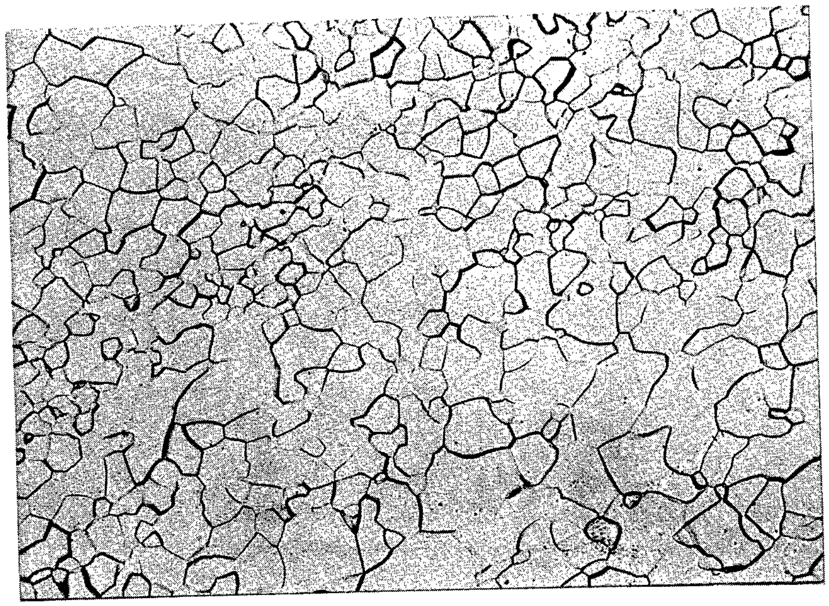

Figure 5 - Microstructure of Phase I T-111, Extrusion Billet Material after Annealing for One Hour at 1600°C (2900°F), Heat DX-571, 100X

After heat treatment, the billets were inspected using dye penetrant and ultrasonic techniques. No evidence of defects was detected. The T-111 Phase I extrusion billets are shown in Figure 6. Data pertinent to the extrusion billets are listed in Table III.

TABLE III - Phase I T-111 Extrusion Billet Data

Billet Identification	Annealed Hardness (DPH)	Weight (lbs)	Diameter (inches)	Length (inches)
DX-571-I-1	218	17.4	2.740	5.176
DX-571-I-2	221	17.1	2.740	5.080

The Phase II B-66 tube blank extrusion billets are shown in Figure 7. Dye penetrant and ultrasonic inspection did not reveal any defects in these billets. Starting B-66 Phase II billet material was processed from an 8-inch diameter double vacuum arc melted ingot by high temperature extrusion. As-extruded the microstructure consisted of approximately 50% recrystallized grains, ASTM 6, as shown in Figure 8. The "as-extruded" hardness of 228 DPH, equivalent to the fully annealed hardness for B-66, remained unchanged after heating sections of the Phase II billet material to 1600°C (2900°F). The effect of annealing temperature on hardness is listed in Table IV.

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TABLE IV - Hardness as a Function of Annealing Temperature for As-Extruded B-66, Heat DX-601T*

Specimen	Annealing Temperature (°C)	(°F)	Diamond Pyramid Hardness, 10 kg Load
DX-601T-II-5	As extruded **		228
DX-601T-II-1	1300	2370	225
DX-601T-II-2	1400	2550	226
DX-601T-II-3	1500	2730	223
DX-601T-II-4	1600	2900	227

*One-hour anneals

**Extruded from 8-inch diameter as-cast ingot to 4.5-inch diameter billet. Extrusion temperature 2900°F.

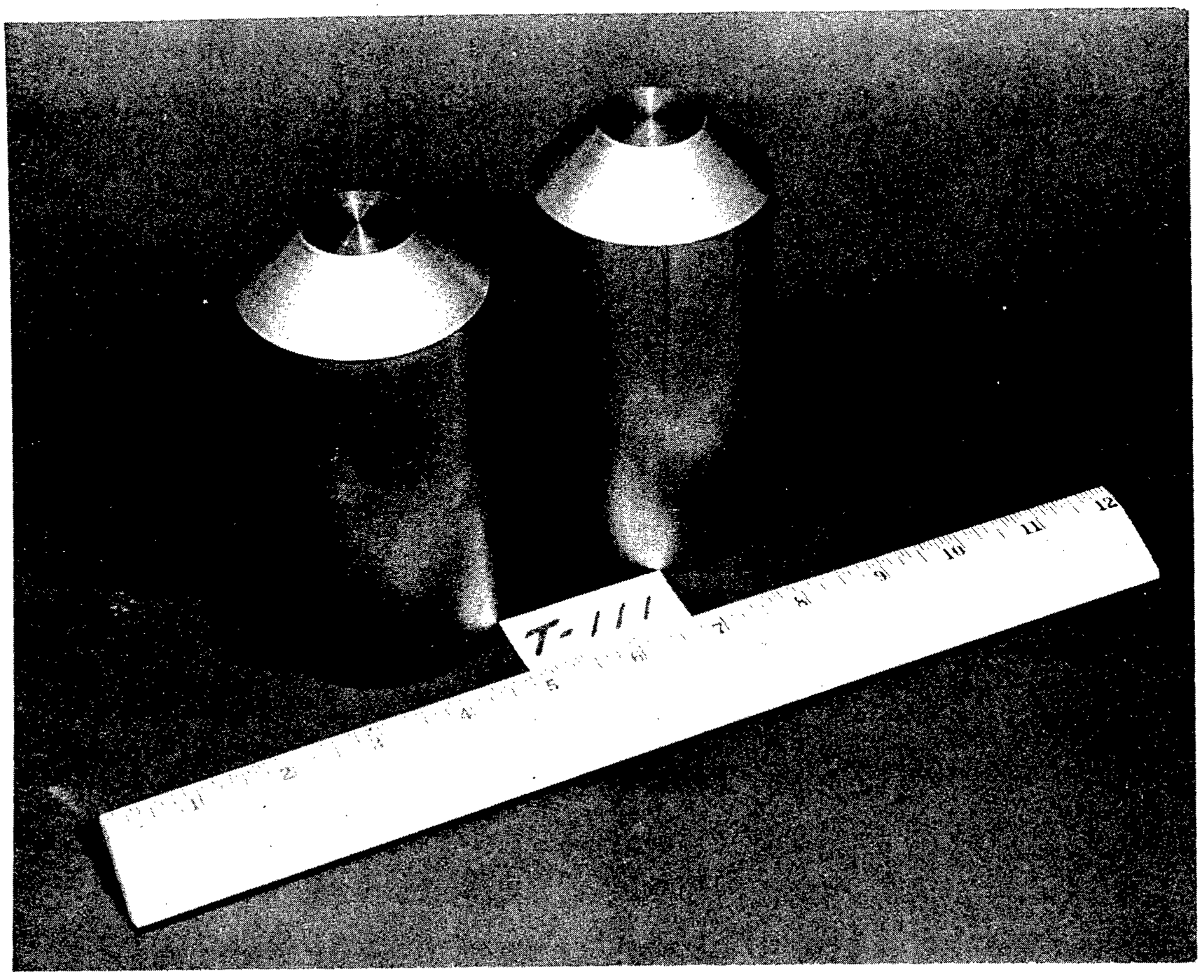

Figure 6 - Phase I T-111 Extrusion Billets, Heat DX-571

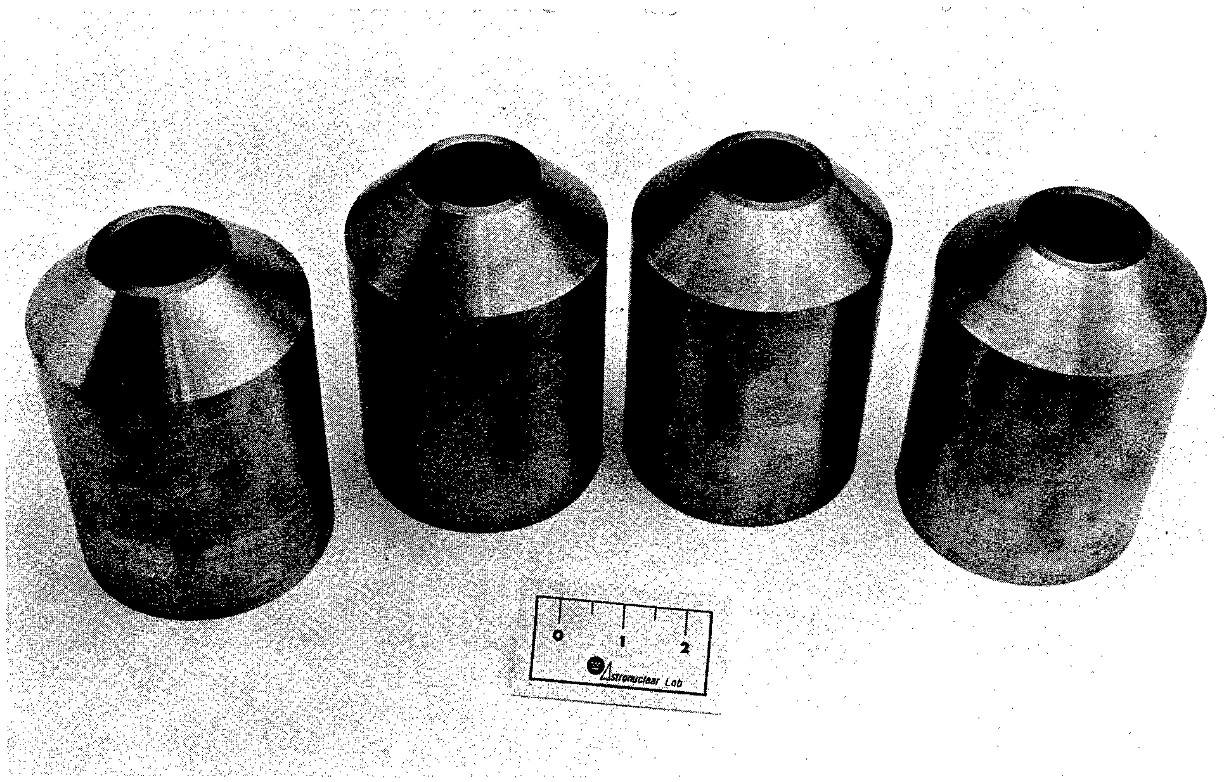

Figure 7 - Phase II B-66 Extrusion Billets, Heat DX-601T

Figure 8 – Microstructure of "As-Received" Phase II B-66 Extrusion Billet Material, Heat DX-601T, 100X

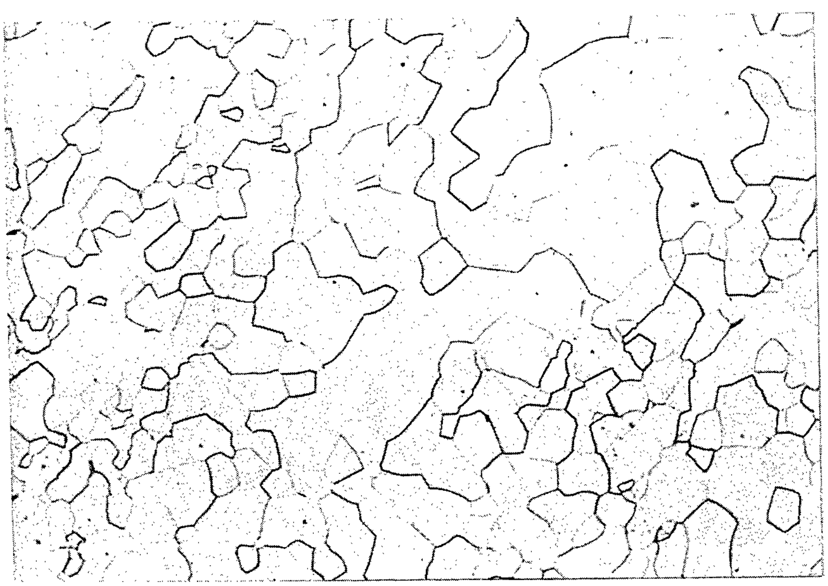

Figure 9 – Microstructure of Phase II B-66 Extrusion Billet Material after Annealing for One Hour at 1400°C (2550°F), Heat DX-601T, 100X

After annealing for one hour at 1400°C (2550°F), the B-66 extrusion had a partially recrystallized microstructure, the recrystallized grains having a size of ASTM 4-6 (Figure 9). Annealing 1 hour at 1500°C (2730°F) provided a completely recrystallized microstructure with a grain size of ASTM 3 or larger. Since the hardness data indicated that annealed mechanical properties had been recovered in the as-extruded billet, and that a completely recrystallized microstructure could not be produced without considerable grain growth, the as-extruded microstructure was selected as the optimum starting condition for the Phase II B-66 extrusion billets.

Starting material for the B-66 Phase I extrusion evaluation billets was processed by swage forging a portion of the extrusion from which the Phase II billets were obtained. The processing schedule for the starting billet material is shown in Figure 10 and the final machined Phase I B-66 extrusion billets are shown in Figure 11. The microstructure of the as-forged B-66 Phase I starting material is shown in Figure 12. After heating for one hour at 1100°C (2000°F), little change was observed in the microstructure although there was a perceptible hardness decrease. The effect of annealing temperature on the hardness of the Phase I B-66 starting billet is given in Table V.

TABLE V - Hardness as a Function of Annealing Temperature for Extruded and Forged B-66, DX-601T

Specimen	Annealing Temperature (°C)	(°F)	Diamond Pyramid Hardness, 10 kg Load
DX-601T-I-0	As worked*		272
DX-601T-I-1	1100	2000	236
DX-601T-I-2	1200	2200	234
DX-601T-I-3	1300	2370	229
DX-601T-I-4	1400	2550	226
DX-601T-I-5	1200**	2200	235

*Extruded from 8" diameter to 4.55" diameter at 2900°F. Forged to 3" diameter at 2200°F.

**Annealed for two hours. All other anneals one hour.

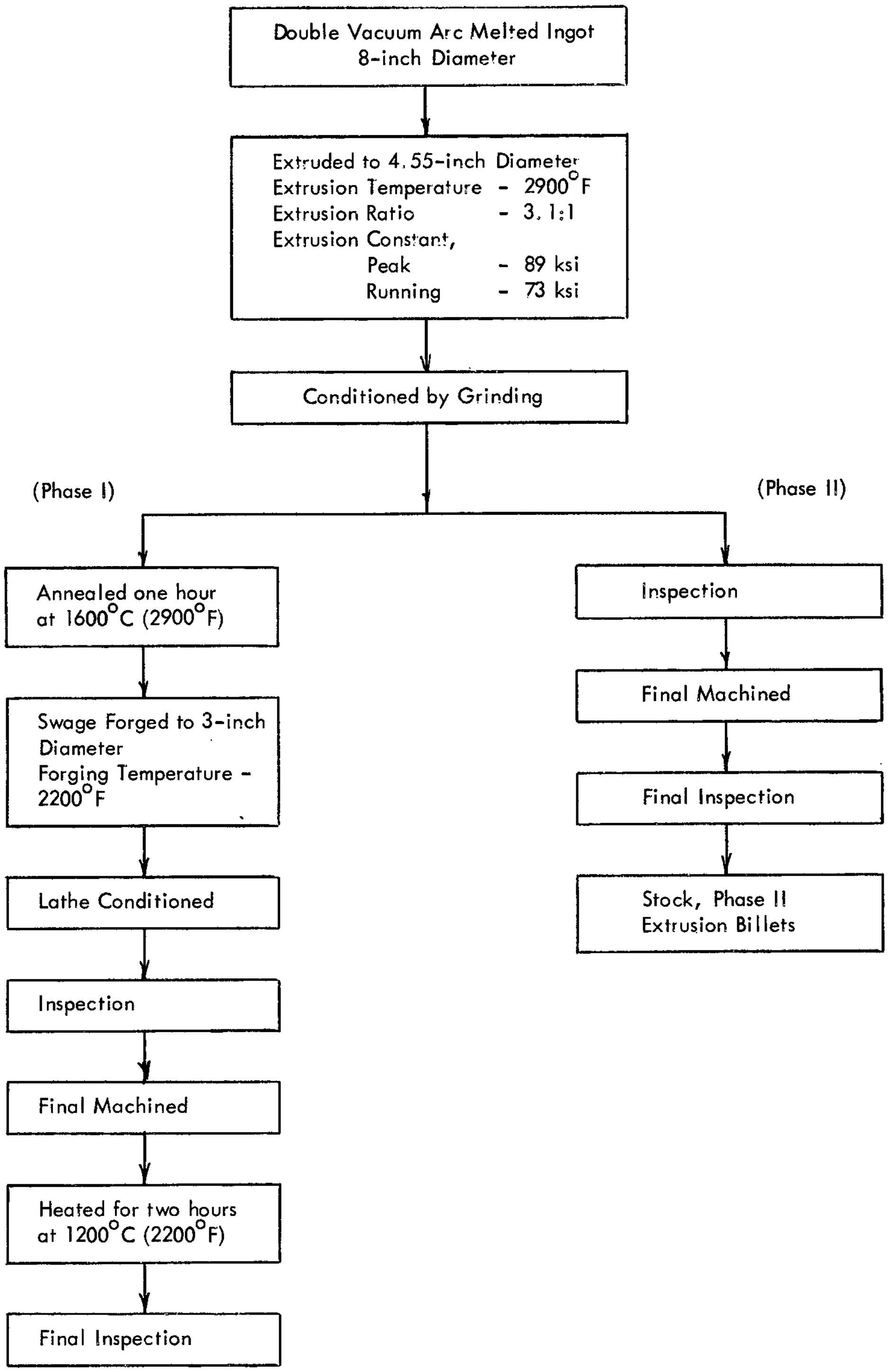

Figure 10 - Processing Schedule for Phase I and Phase II B-66 Extrusion Billet Material, Heat DX-601T

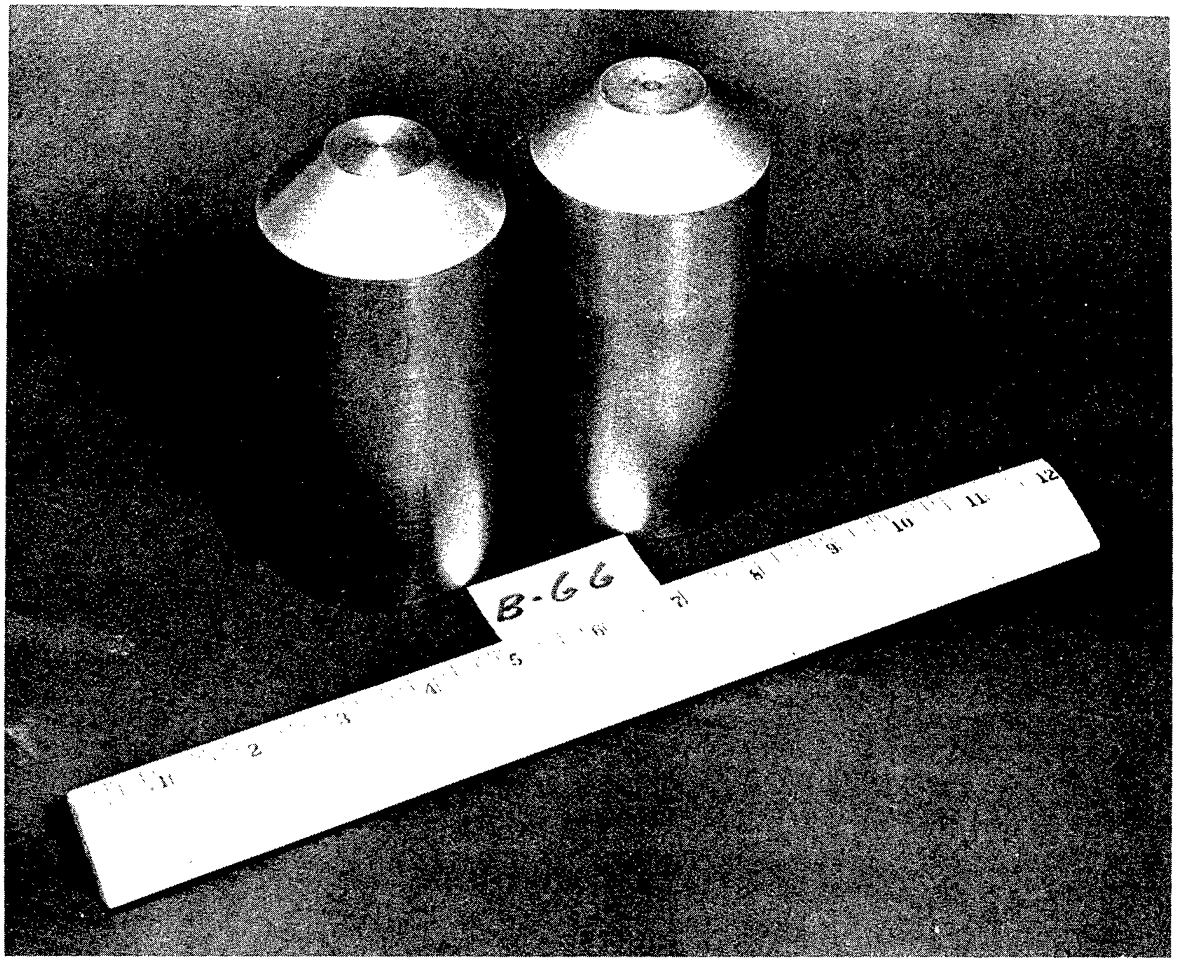

Figure 11 - Phase I B-66 Extrusion Billets, Heat DX-601T

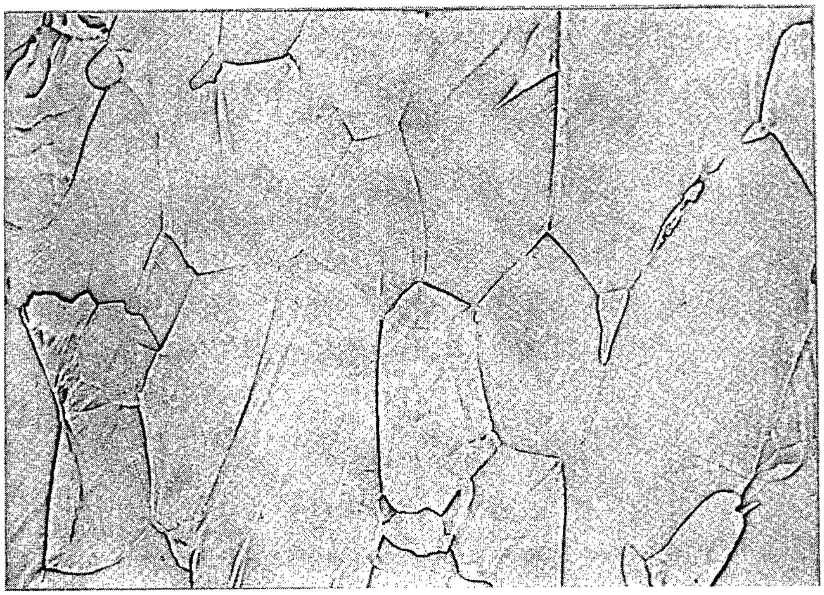

Figure 12 - Microstructure of "As-Received" Phase I B-66 Extrusion Billet Material, Heat DX-601T, 100X

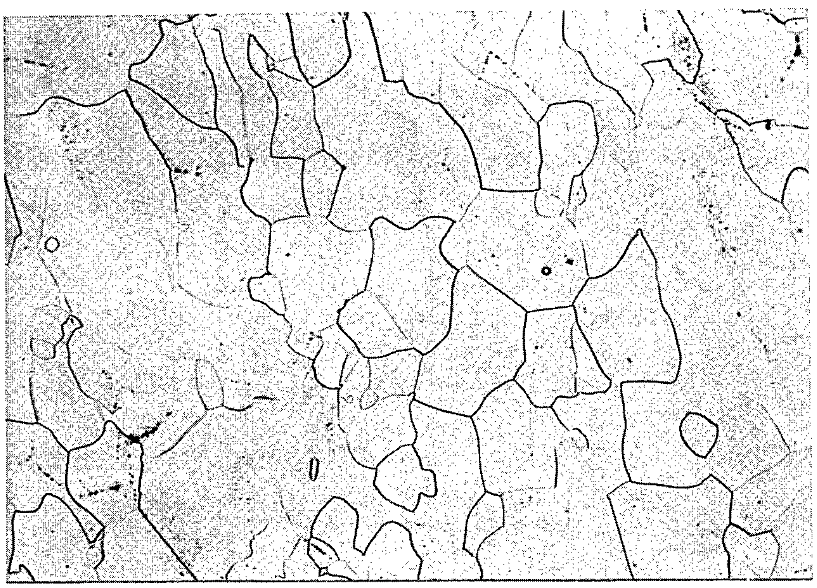

Figure 13 - Microstructure of Phase I B-66 Extrusion Billet Material after Heating One Hour at 1300°C (2370°F) Heat DX-601T, 100X

Recrystallization was essentially complete after heating for 1 hour at 1300°C (2370°F), with the recrystallized grain size approximately ASTM 3 (Figure 13). Although annealing 1 hour at 1200°C (2200°F) had little effect on the as-received microstructure, heating for 2 hours at 1200°C (2200°F) resulted in the formation of some recrystallized grains (Figure 14). A microstructure similar to that in the Phase II starting billets could not be obtained in the Phase I extrusion billets by heat treatment since the observed recrystallized grain size for the Phase II billet material was ASTM 6 (Figure 8) while the recrystallized grain size for the Phase I billet material was ASTM 3. This anomalous recrystallization behavior is not understood at the present time. It would be expected that the Phase I billet material would have a lower recrystallization temperature with a finer recrystallized grain size, since it was worked a greater amount at a lower temperature. The Phase I B-66 extrusion billets were heat treated for 2 hours at 1200°C (2200°F). This treatment provided recovery of annealed mechanical properties and caused the formation of some recrystallized grains. Dye penetrant and ultrasonic inspection did not reveal any defects in the Phase I billets. Table VI lists the data pertinent to the B-66 Phase I extrusion billets.

TABLE VI - Phase I B-66 Extrusion Billet Data

Billet Identification	Annealed* Hardness (DPH)	Weight (lbs)	Diameter (inches)	Length (inches)
DX-601T-I-1	235	8.8	2.730	5.191
DX-601T-I-2	232	8.8	2.735	5.207

*2 hours at 1200°C (2200°F)

B. Extrusion Evaluation

Extrusion was accomplished using a 1400-ton high speed extrusion press at Nuclear Metals, Inc. The mild steel clad billets were heated to temperature in an argon purged retort and were soaked at temperature for approximately one hour prior to extrusion. The clad billets were extruded through a bare cone type die. The extrusion results are summarized in Table VII and the extrusions are shown in Figures 15 and 16. After extrusion, the mild steel cladding was removed by pickling.

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TABLE VII - Extrusion Data for Phase I B-66 and T-111 (b)

Billet (b) Identification	Extrusion Temperature (°F)	Extruded Diameter (inches)	Reduction Ratio	Extrusion Speed (inches per minute)	Extrusion Constants (ksi) Upset	Extrusion Constants (ksi) Running	Comments
B-66-1	2200°F	1.12	7:1	275	94	84	Surface excellent, light rattlesnaking near extrusion nose
T-111-1	2200°F	1.09	7:1	175	92	92	Surface excellent, press stalled after approximately 85% extruded
B-66-2[a]	2200°F	1.18	7:1	1000	102	78	Moderate rattle-snaking and shallow circumferential cracks along entire extrusion
T-111-2	2300°F	1.10	7:1	1000	84	80	Surface excellent, light rattlesnaking near extrusion nose

[a] During heating, retort collapsed permitting mild steel can to oxidize. The press stalled on initial extrusion attempt. Billet was remachined, then re-canned.

[b] Billets listed in order of extrusion.

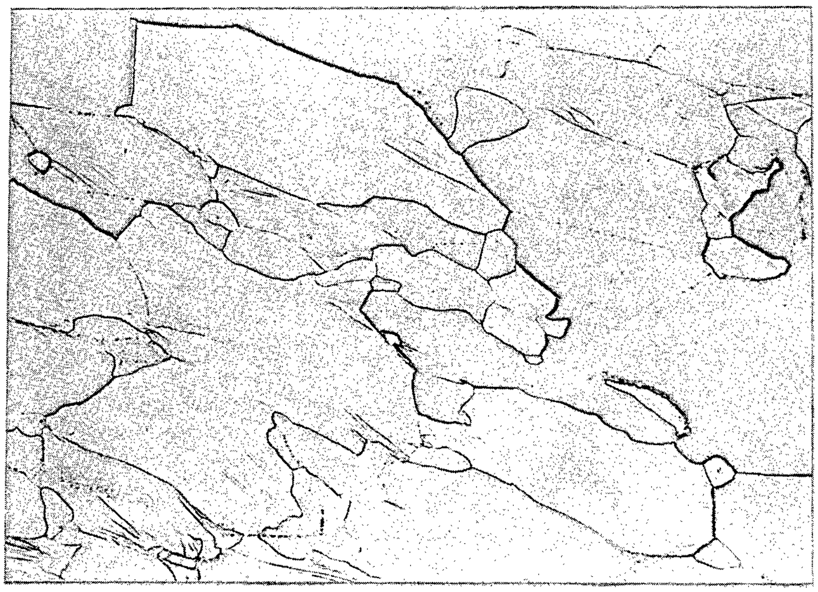

Figure 14 - Microstructure of Phase I B-66 Extrusion Billet Material after Heating for Two Hours at 1200°C (2200°F), Heat DX-601T, 100X

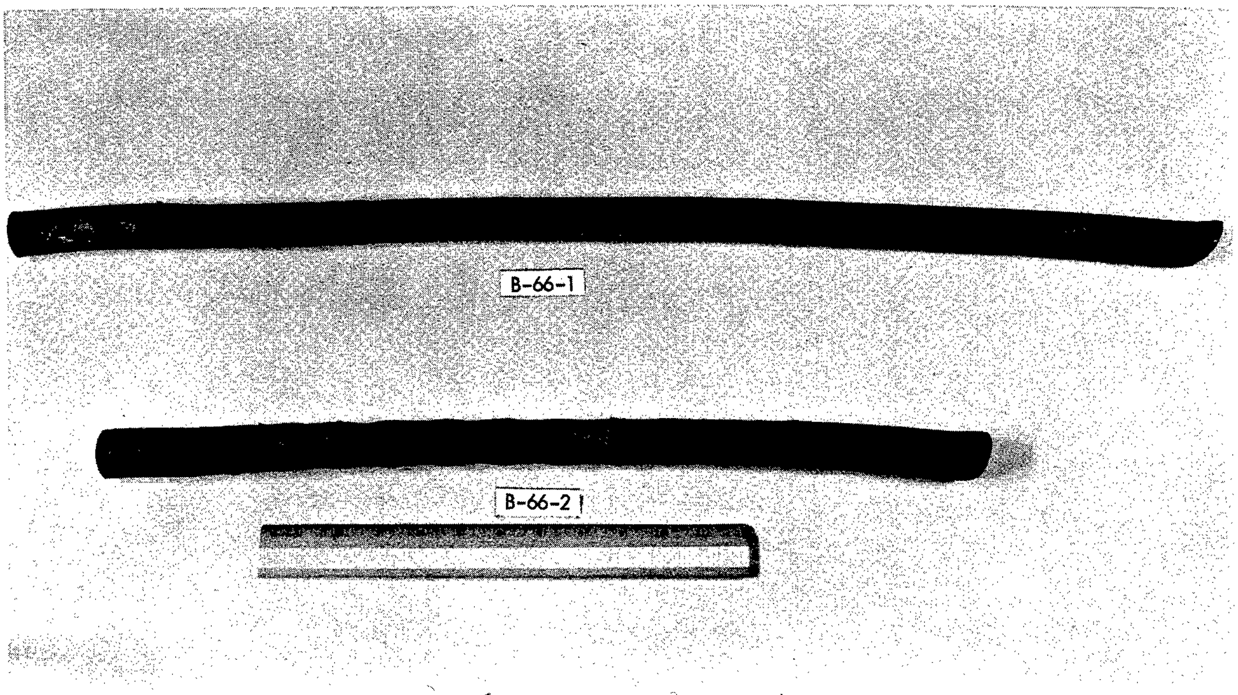

Figure 15 - Phase I B-66 Extrusions, Heat DX-601T

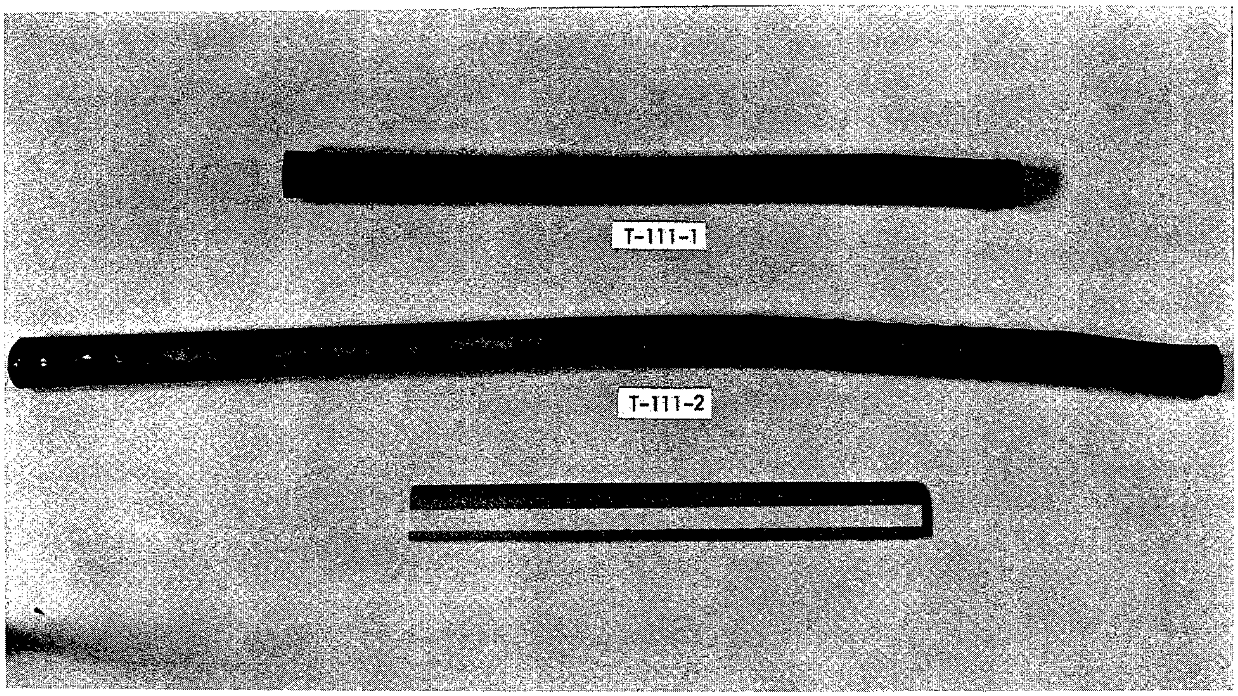

Figure 16 - Phase I T-111 Extrusions, Heat DX-571

During heating, the top front portion of the retort sagged, permitting excessive oxidation of the steel cladding on the B-66-2 billet. The oxide was not completely removed during the billet transfer operation causing the press to stall when extrusion was attempted. Both B-66-2 and T-111-2 billets were allowed to cool to room temperature, the cans were stripped, the billets reclad, and then extruded.

Both T-111 billets had excellent as-extruded surfaces. Increasing both the extrusion temperature and speed had no apparent effect on surface quality, although light rattlesnaking was observed near the nose end of the T-111-2 extrusion billet, (Figure 17). Increasing the extrusion speed from 275 to 1000 inches per minute for B-66 resulted in degradation of the as-extruded surface. As shown in Figure 18, B-66-2 exhibited transverse circumferential cracking the entire length of the billet and moderate rattlesnaking occurred along the length of the extrusion. The as-extruded surface for the B-66 extrusions is shown in Figure 18. It is estimated that the crack depth is 0.02-0.04 inch.

It had been estimated that the tube blank extrusions would be successful if the extrusion constant for the solid rod extrusions did not exceed 100 ksi[3]. From the limited extrusion evaluation conducted, it appears that B-66 and T-111 will be successfully extruded as tube blanks. The extrusion parameters selected for the initial B-66 and T-111 tube blank extrusions, based on the results of the Phase I evaluation, are listed in Table VIII.

Type →

TABLE VIII - Parameters for the Phase II B-66 and T-111 Tube Blank Extrusions

Alloy	Extrusion Temperature (°F)	Extrusion Speed (inches per minute)
B-66	2200	300 - 500
T-111	2300	1000

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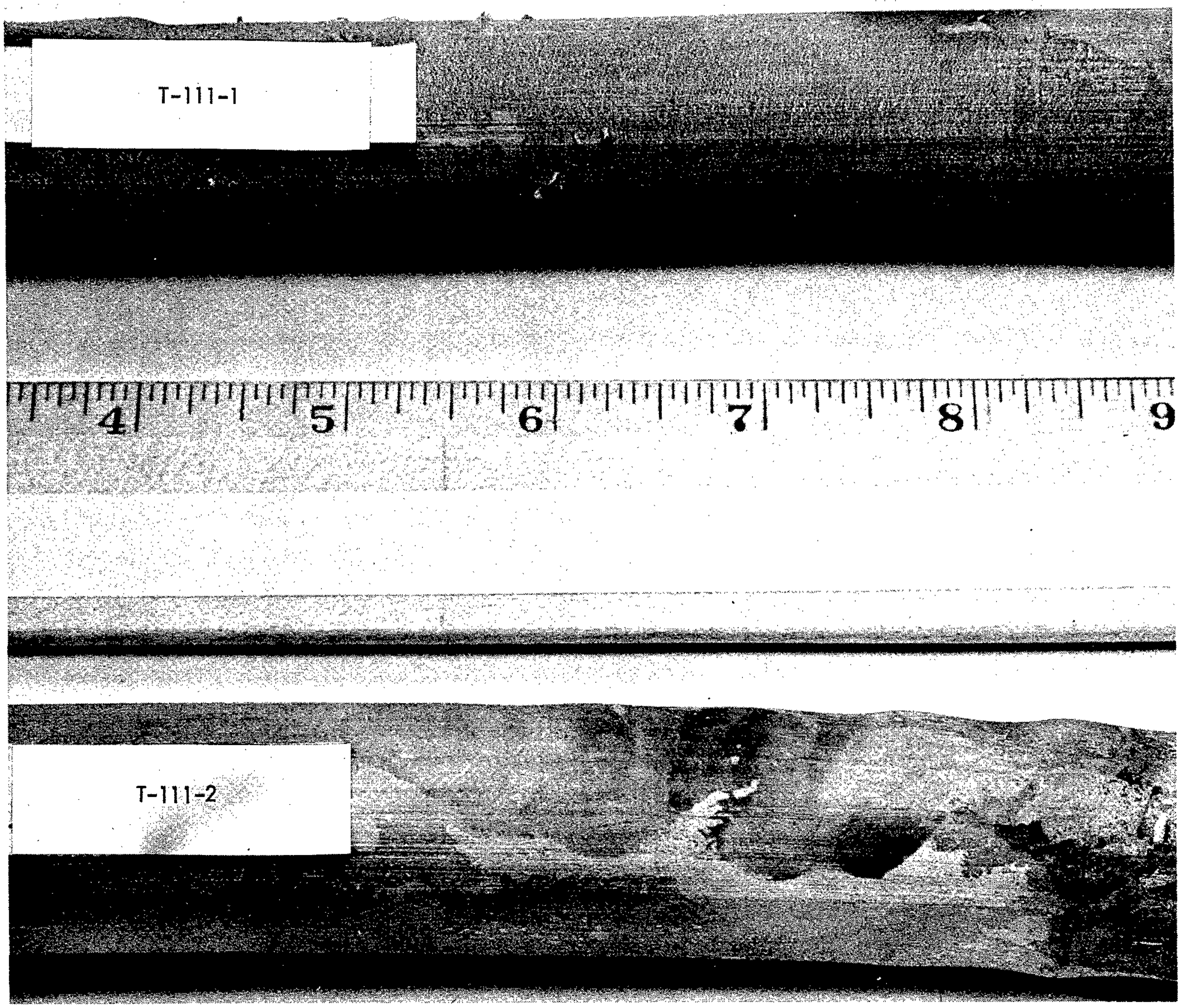

Figure 17 - Phase I T-111 "As-Extruded" Surface, Heat DX-571

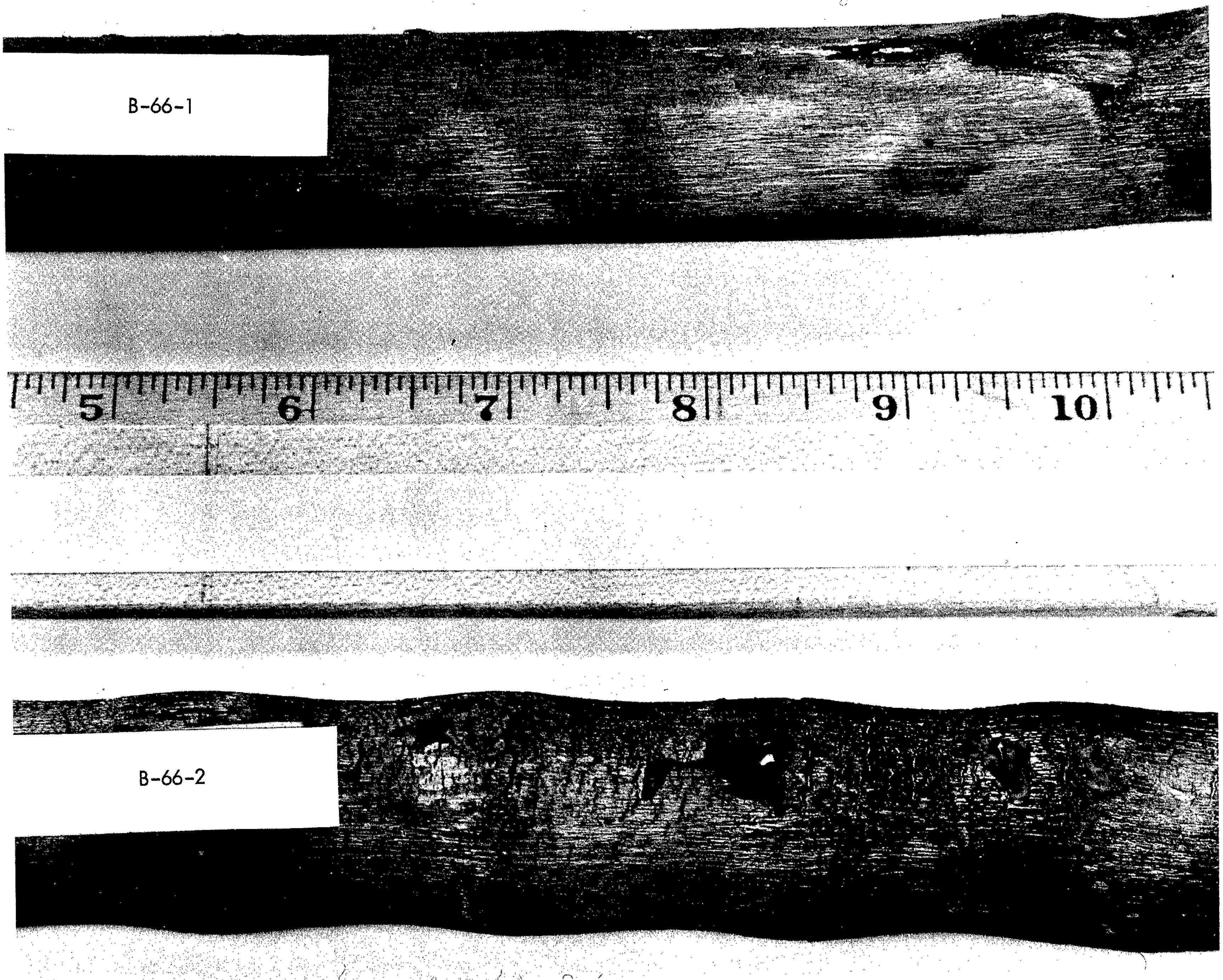

Figure 18 - Phase I B-66 "As-Extruded" Surface, Heat DX-601T

The B-66-1 extrusion was straightened by hammer forging at 950°C (1750°F). After being straightened, the extrusion was lathe conditioned and centerless ground to give the 0.963" diameter x 15-1/8" long rod, shown in Figure 19. Metallurgical evaluation of the as-worked microstructure is in progress. A heat treatment that will produce a grain structure most beneficial for the subsequent hot drawing evaluation will result from this investigation. Evaluation of the hot tube drawing characteristics of the B-66 will be done at the Superior Tube Company.

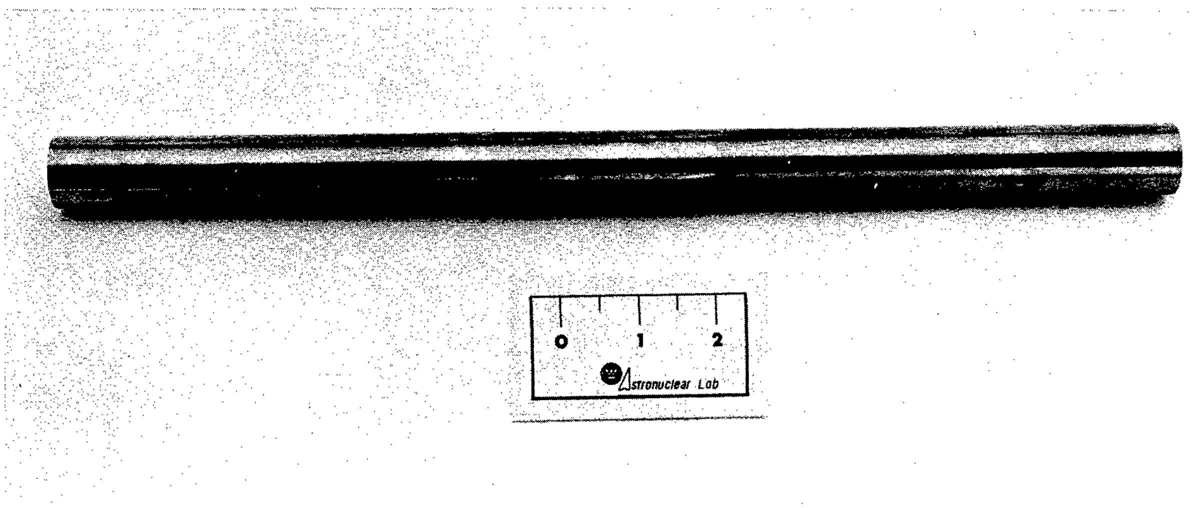

Figure 19 - Phase I B-66 Conditioned Extrusion, Heat DX-601T

III. FUTURE ACTIVITIES

During the next reporting period, the hot drawing evaluation of B-66 will be completed. The Phase II tube blank extrusions will be accomplished and the initial tube blank reductions started. The metallurgical evaluation of the Phase I extrusion billets will also be completed during the next reporting period.

IV. REFERENCES

1. Buckman, R. W. - "Development of High Strength Columbium and Tantalum Alloy Tubing", 1st Quarterly Progress Report, WANL-PR-(N)-001

2. Bishop, E. C. - Private communication.

3. Toeffin, W. - Private communication.