

Lawrence Berkeley National Laboratory

Recent Work

Title

NUCLEAR MAGNETIC RESONANCE STUDY OF PRECIPITATION IN ALUMINUM-4% COPPER

Permalink

<https://escholarship.org/uc/item/2cq0r7qv>

Authors

Plaza-Meyer, Elias
Perra, Mark Walter.

Publication Date

1973-09-01

c.1

NUCLEAR MAGNETIC RESONANCE STUDY
OF PRECIPITATION IN ALUMINUM-4% COPPER

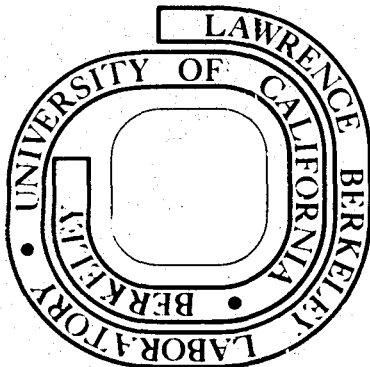
Elias Plaza-Meyer and Mark Walter Perra

September 1973

Prepared for the U. S. Atomic Energy Commission
under Contract W-7405-ENG-48

For Reference

Not to be taken from this room



RECEIVED
LAWRENCE
RADIATION LABORATORY

NOV 16 1973

LIBRARY AND
DOCUMENTS SECTION

c.1

LBL-1883

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

NUCLEAR MAGNETIC RESONANCE STUDY OF PRECIPITATION
IN ALUMINUM -4% COPPER

Elias Plaza-Meyer and Mark Walter Perra

Inorganic Materials Research Division, Lawrence Berkeley Laboratory and
Department of Materials Science and Engineering, College of Engineering;
University of California, Berkeley, California

ABSTRACT

Using the Bloch induction method of observation the kinetics of zone formation in the Al-4% Cu system has been studied by continuous monitoring of the Al-27 nuclear magnetic resonance (NMR) absorption signal during isothermal aging. It was demonstrated that the absorption intensity is a parameter which is strongly influenced by the distribution of solute atoms. The exponent of time in the approximate law for diffusion-controlled growth, $X = 1 - \exp[-(t/\tau)^n]$, was calculated to be $n \sim 2$ for $t \ll \tau$ and $n \sim 1$ for $t \sim \tau$ for the formation of GP I zones at 150°C, in agreement with Ham's theory for the growth of platelets of constant thickness.

INTRODUCTION

A number of different techniques have been used to study the mechanism and kinetics of precipitation in supersaturated metallic solid solutions. Most of these techniques do not follow the process continuously and in very many it is extremely difficult to correlate changes in the physical property being measured with the precipitation process itself.

The present investigation uses the nuclear magnetic resonance technique to study precipitation in the Al-Cu system. This technique was selected because it is known that the Al-27 resonance intensity is strongly influenced by solute concentration through quadrupole interactions and, further, changes in NMR signal parameters should be more clearly attributable to the rearrangement of solute atoms.

QUADRUPOLE EFFECTS

The nuclear resonance of Al-27 undergoes a large reduction in amplitude when it is alloyed, Fig. 1.¹ It is now well established that this reduction in amplitude arises from the oscillatory distribution of screening charge about the solute atoms. The interaction of the charge oscillation with the nuclear electric quadrupole moment of Al-27 nuclei which are in the vicinity of a solute atom perturbs the energy levels of these nuclear dipoles and shifts their transitions appreciably.

At points of cubic symmetry in crystals electric field gradients, and hence quadrupole interactions, vanish.² In the absence of an electric quadrupole interaction the $2I + 1$ magnetic energy levels are equally spaced and the corresponding $2I$ absorptions are superposed. (Here I is the nuclear spin quantum number, which for Al-27 is $5/2$.) The addition of solute will destroy the cubic symmetry in the vicinity of solute atoms and will introduce local electric field gradients. These field gradients interact with the Al-27 quadrupole moment in the Hamiltonian describing the nuclear energy and remove the degeneracy of the various allowed transitions. This interaction shifts the various transitions unequally and splits the resonance line into its $2I$ components. According to first order perturbation theory, there will be a shift in the frequencies of the satellite transitions but not in the central ($m = \frac{1}{2} \rightarrow -\frac{1}{2}$) transition. The resonance frequencies of nuclei which are sufficiently near a solute atom will be shifted strongly, so the signal from these Al nuclei will pass beyond the limits of the line observed causing reduction in intensity

An expression derived by Kohn and Vosko,³ and Blandin and Friedel,⁴ gives the electric field gradient $eq(r)$ in terms of the change in charge density $\Delta\rho(r)$ introduced by an impurity atom;

$$eq(r) = \frac{8\pi e}{3} \alpha \Delta\rho(r)$$

where r is the distance from the impurity atom and α is a constant determined by the electronic structure of the host metal and is called

a core enhancement factor. The charge density $\Delta\rho(r)$ in this equation denotes only the smooth part of the screening charge density, which carries with it a rapidly varying part arising from the interaction with ion cores of host atoms. The screening charge falls off as r^{-3} multiplied by a sinusoidal function of r .

Fukai and Watanabe¹ have used the above expression to calculate the contribution of Al-27 nuclei to the resonance as a function of nuclei distance from the solute atom for Cu, Ag, and Zn solutes. They considered an ideally random, primary solid solution of c atomic fraction of solute. Using the "wipe-out" model, which states that the satellite intensity should follow the function $(1 - c)^n$, n was established to be 236 for Cu and 205 for Ag for c less than 10^{-2} . The exponent n in this model is interpreted to be the number of solvent atoms in a sphere around a solute atom which has an electric field gradient sufficient to remove their satellite transitions from the central transition. The function $(1 - c)^n$ is the probability that a solvent nucleus has no solute atom inside this sphere, provided the solution is ideally random.

Ordering or clustering will have a profound effect on resonance intensity. Because a factor causing quadrupole perturbation is lattice symmetry, a redistribution of solute atoms will directly influence the intensity. If an ordering process produces cubic symmetry around the observed nuclei, the intensity will be greater than for random placement of the surrounding atoms. Similarly, if the process produces a lower symmetry, the intensity will be diminished. Clustering

of solute atoms will in general increase the resonance intensity because part of the lattice would then be depleted in solute and consequently fewer solvent atoms would then be sufficiently near a solute atom to have their satellite transitions perturbed. In clustering or precipitation processes, then, the intensity can be used as a measure of the composition of the solvent-rich phase and can be used as a measure of the extent of the process.

EXPERIMENTAL

A. Preparation of Samples

The alloys were prepared in one inch diameter by 9 inch long ingots from material of 99.999% purity. The alloys were melted in a graphite crucible under an argon atmosphere. After heating in a 10 kW induction furnace, the melts were poured into a spinning chill-mold. The ingots were homogenized at 530°C for 3 days in an evacuated chamber, and water quenched. No large scale segregation was evident in the microstructures of the ingots.

The powder for the resonance experiments was prepared by filing the ingots at room temperature with #4 files in an automatic filing machine. The iron introduced by filing was removed as far as possible with a small magnet. Only those particles which passed through a 250 mesh sieve, but not through a 400 mesh sieve were accepted. This restricted the particle size to between 62 and 35 microns, which is smaller than the RF skin depth in these alloys.⁵ Nearly 50% of the filings were within this range of size.

Samples for hardness measurements, 0.2 inches thick by 1 inch diameter circular discs, were sliced from the ingots with an abrasive saw. These discs were solution treated at 530°C for 48 hours in an argon atmosphere and were then quenched directly into a Dow 710 oil bath which was at the appropriate aging temperature. The Rockwell E scale was used for hardness testing.

B. NMR Experimental

Experiments were conducted using a modified Varian Associates model V-4311 wide line NMR spectrometer, operated at a fixed frequency of 8.134 mc. To stabilize the base line and to increase the signal-to-noise ratio, the steady magnetic field H_0 was modulated with the field H_m . The resulting signal was phase-detected. A PAR (Princeton Applied Research) model HR-8 lock-in amplifier was used to provide the phase-sensitive detection and the modulation field. The reference output of the lock-in amplifier was amplified with an audio amplifier and then applied to the modulation coils in the Varian crosscoil probe, Fig. 2. A modulation frequency of 153 Hz was used, which satisfies the criteria $\omega_m T_1, \omega_m T_2 \ll 1$ (where T_1 is the spin-lattice relaxation time and T_2 is the spin-spin relaxation time of the Al-27 nucleus) so that the derivative of the absorption signal was observed. The corresponding modulation amplitude was 1.38 gauss. The amplitude of the audio modulation was adjusted to be as large as possible without causing modulation broadening. The audio phase was adjusted by selecting the phase which gave maximum intensity for the derivative

signal. The power setting of 0.5 watt was used for the leakage detection of the RF field.

The gross magnetic field, H_0 , was swept at a rate that varied from 10 to 30 gauss per minute depending on the signal obtained and the rate of change of signal parameters with time. This rate is compatible with the line width in that there was negligible saturation. The time constant of the RC filtering circuit in the lock-in amplifier was adjusted so that it was less than one tenth the time spent on the resonance.

A sample which is in the crosscoil induction probe, Fig. 2, experiences both a magnetic field H_0 and a weak radio frequency field H_1 from the transmitter coils. The combination of the absorptive sample, radio frequency field and magnetic field can be represented electrically by a resonant circuit with a complex impedance. The real part of the impedance is called the absorption mode and the imaginary part is called the dispersion mode. As the magnetic field is swept through resonance, the impedance changes radically and the receiver coil detects this change. The type of mode that is received depends on the coupling between the receiver coils and the fields in the probe. The coupling is controlled mechanically by four paddles. As these paddles are moved in the transmitter coil field, the coupling between the receiver and the fields in the probe varies. Before an interpretable signal in any experiment can be received, the paddles must be adjusted so that either pure absorption or pure dispersion is obtained.

In the Varian crosscoil induction probe, extreme temperatures can damage the RF transmitter coil and the modulation coils. Also, because of mechanical balancing of the absorption and dispersion modes, the thermal expansion of balancing paddles upsets modal balance. In order to protect the coils and to attain the maximum coupling stability, a dewar insert, Fig. 3, was constructed so as to allow for a flow of room temperature air to pass between the coils of the probe and the dewar's outer wall. Because the signal-to-noise ratio depends strongly upon the filling factor of the receiver coil, the receiver coil was attached to the inside of the dewar. In this arrangement, the annular region for the flow of temperature regulating gas and the glass wall of the sample tube were the only regions within the coil not containing the sample. The receiver coil was made of 18 turns of 10 mil copper wire and had an inside diameter of 0.488 inches. The coil was cemented to the inner wall of the dewar with a ceramic cement which was able to withstand the temperature extremes in the experiment.

The sample temperature was regulated by passing dry nitrogen gas through the annular space between the sample and the dewar insert. For high temperature experiments (above 100°C) a 1 1/2 inch, 700 watt tube furnace, which was driven by a regulated power supply, was used to heat the gas. A 12 inch long graphite cylinder was placed within the tube of the furnace to facilitate heat transfer to the gas and to insure temperature uniformity of the heated gas. A particular fixed temperature was maintained by adjusting the nitrogen gas flow

rate and the furnace current. The sample temperature was controlled to $\pm 1^\circ\text{C}$ and was monitored continuously throughout the experiment on an Esterline Angus Strip Chart potentiometer.

The sample tube for high temperature experiments, Fig. 4, consisted of two concentric thin-walled pyrex tubes: the outer tube was 11 mm OD and the inner tube was 4 mm OD. The sample was contained within the annular space between the inner and outer tubes and the reference sample, V-51, was contained within the inner tube. Both the inner and outer chambers were evacuated. An extra long seal-off tip was left on the sample tube for re-evacuating after samples were changed.

Before an experiment at high temperatures was begun, a dummy sample was placed in the spectrometer and the desired temperature was reached. The sample tube was then exchanged with the dummy sample tube at the beginning of the experiment so that thermal equilibrium in the probe was reached as quickly as possible. The dispersion mode was eliminated from the signal as quickly as possible by adjusting the paddles. The absorption mode signal was then recorded as precipitation in the sample proceeded.

Data from the experiment was recorded both on a strip chart recorder and on punched paper tape.

RESULTS AND DISCUSSION

A. Hardness Data

To confirm that the alloy which had been prepared for the NMR experiments had aging characteristics similar to those characteristics which had been found in experiments by other workers, in particular Hardy⁶ and Silcock et al.,⁷ hardness vs. time curves were established for the alloy at 150°C, Fig. 5, and at 230°C, Fig. 6. In Hardy's early work on this system, two distinct types of hardness curves were obtained. He found that at temperatures of 110°C and 130°C, hardening occurred in two stages separated by a flat plateau of constant hardness. At higher temperatures, 190°C and 220°C, he found one stage aging curves. The later x-ray work by Silcock indicated that for the two stage aging curves the initial rise in hardness resulted from the formation of GP I zones and that the second rise was due chiefly to the presence of the GP II structure. Further, Silcock established that the one stage aging at the higher temperatures was attributable to the formation of the θ' precipitate.

Two stage and single stage aging behavior are both clearly demonstrated in Figs. 5 and 6, respectively. It is noted for both of these curves that hardening did not begin immediately, and that there was even an initial decrease in hardness. Hardy's results were similar but did not show such a pronounced effect. Hardy pointed out that the initial drop in hardness was probably due to the relief of quenching strains. The time to attain peak hardness was longer in

this work than in Hardy's work. For example, Hardy's 220°C water quenched specimen reached peak hardness after 3 hours whereas the 230°C specimen in this work reached peak hardness after about 7 hours. This probably reflects differences in specimen sizes and quenching rates.

B. NMR Results at 150°C and 230°C

Figure 7 shows a typical NMR absorption derivative curve which was obtained during the isothermal aging of Al-4% Cu at 230°C. A simultaneous digital record of the absorption derivative was made on punched paper tape.

Al-27 absorption signal parameters such as intensity and spectral location were compared with the corresponding signal parameters from a known amount of V-51 powder. This powder was encapsulated within the sample tube, Fig. 4. Since V-51 has a gyromagnetic ratio very close to that of Al-27, the V-51 resonance occurs only a few hundred gauss downfield from the Al-27 resonance when the radio frequency is 8.134 mc. A reference signal was thus established throughout each isothermal aging experiment so that changing parameters within the spectrometer would automatically be taken into account. Also, because the same mass of Al-27 was used in each aging experiment, the integrated intensities from experiments at different temperatures could appropriately be compared. By comparing the resonance frequency of the sample to the resonance frequency of the V-51 reference, it was established that no perceptible Knight shift occurred during aging. Similarly, no line

broadening occurred. The signal intensity, however, was strongly influenced by the rearranging of solute atoms during aging.

Figures 8 and 9 show the intensity of the absorption signal vs. time for aging at 150°C and 230°C, respectively, normalized to the intensity of the V-51 signal. Several features of these curves are worth noting. First, an incubation period was not observed. The initial rate of intensity change was the most rapid and the rate decreased with elapsed time. For these samples there was a ten to fifteen minute period while the sample temperature was increasing and the probe temperature was stabilizing before a signal could be recorded. Given the high rate at which clustering is known to occur at low temperatures, it can reasonably be stated that the zones had been nucleated well before the NMR signal was obtained and that the observed intensity changes correspond to a competitive growth process. Next, the final intensity was greater in the 150°C curve than in the 230°C curve, which is consistent with the slightly lower solubility of the copper at the lower temperature. Finally, the absorption intensity in the 230°C curve had become nearly constant with time at 20 hours whereas the intensity in the 150°C curve was unmistakably increasing at 280 hours.

When the NMR intensity is plotted vs. the log of the aging time, Figs. 10 and 11, similar to the hardness curves, Figs. 5 and 6, several features of the curves are emphasized. Unlike the hardness curves, there was no noticeable incubation period, i.e. the intensity was increasing from the earliest time at which it could be recorded.

There is a considerable amount of scatter in the data, which was caused by the continuing readjustment of the coupling between the transmitter and receiver coils which was necessary throughout the experiment. Within the limit of experimental error, however, the curves consist of a series of straight line segments.

On the 150°C curve there is a distinct point at which the slope of the curve increases. The time at which this change of slope occurs corresponds quite closely to the time on the 150°C hardness curve at which the second hardening stage (GP II formation) has begun. Presumably if the experiment had been continued for several hundred more hours, a third stage would have been reached during which the intensity would have become constant with time. (Demand for the use of the spectrometer by others was too high to allow for such time-consuming work.) The NMR intensity vs. log time curve at 230°C shows a sharp decrease of slope after four hours. The time at which this decrease occurs precedes slightly the time at which the maximum in the single stage hardening due to θ' occurs. Although assigning specific events to the NMR curves on the basis of events which are known to occur on corresponding hardness curves at the same time would be questionable, comparisons should at least give an indication of the sequence of events in the NMR curves.

Consider the first segment of the curve in Fig. 10, the formation of GP I at 150°C. For the purposes of determining a growth law for the GP I zones, we make the following assumptions:

1. That the increase in intensity of the Al-27 signal is due only to solute depletion.¹

2. That the fraction of solute transformed is equal to the fractional increase in absorption intensity, i.e.

$$X = \frac{c_i - c}{c_i - c_f} = \frac{I - I_i}{I_f - I_i}$$

where c_i and c_f are the initial and final concentrations of solute in solution for the process and I_i and I_f are the corresponding intensities. (The wipe-out model relating composition and intensity is appropriate only for very dilute solutions.)

3. That only GP I formation occurs during this period.⁸ The intensity vs. log time, in this period, can be mathematically represented as a straight line. The usefulness of this representation is limited, however, because physical models have not been developed which predict reactions that proceed with the log of the time.

A kinetic equation which is frequently used in the description of diffusion-controlled growth processes is

$$X = 1 - \exp [-(t/\tau)^n]$$

where X is the fraction of solute transformed and n is called the growth exponent. Zener and Wert⁹ suggested that the physical significance of n could be interpreted in terms of the geometrical shape of the particles. Many electron micrography and x-ray diffraction studies¹⁰ have shown that even during their later stages GP I zones

are platelets one to two atoms thick. The thickness of the zones does not increase during growth because of strain energy involved in the coherent precipitation. According to the Zener-Wert theory for diffusion-controlled growth of platelets of constant thickness, the growth exponent is 2. More recently, Ham¹¹ has shown that the growth exponent for this case in the short time approximation ($t \ll \tau$) is 2, but has also shown for longer times ($t \sim \tau$) that the exponent is 1.

We may rewrite the expression for the fraction of transformed solute and take the natural log of both sides,

$$\ln \left(\frac{1}{1-X} \right) = \left(\frac{t}{\tau} \right)^n .$$

Again taking the log of both sides,

$$\ln \ln \left(\frac{1}{1-X} \right) = n \ln t - n \ln \tau$$

so that if $\ln \ln \left(\frac{I_f - I_i}{I_f - I} \right)$ is plotted vs. $\ln t$, the slope gives n and the intercept gives $n \ln \tau$.

This curve has been plotted in Fig. 12 from the GP I stage of the 150°C aging curve. I_i is the resonance intensity extrapolated to $t = 0$ (although this extrapolation may be inaccurate) and I_f is the resonance intensity at $t = 13$ hours. There is an unmistakable decrease in slope with increasing time in Fig. 12. The approximate slope of this curve is plotted vs. time in Fig. 13 where it is seen that the initial slope is near 2 and the final slope is slightly less than one. A plot of

$$\ln \ln \left(\frac{I_f - I_i}{I_f - I} \right) \text{ vs. } \ln t$$

for the 230°C curve in Fig. 14 indicates that the slope is nearly constant throughout the first half of the aging process and has a value of approximately 1.

It is tempting to claim that the result for the growth exponent for GP I is consistent with and corroborates Ham's theoretical results.¹¹ However, there could be considerable inaccuracy involved in extrapolating to $t = 0$. Also, Ham's theory requires that the particles always be small in comparison with their mean separation. Specifically, for discs of constant thickness, it requires that the mean separation be at least 10 times the disc diameter. Nicholson and Nutting¹² used the thin foil technique to study the structure of GP I in water-quenched Al 3.4%Cu which had been aged at 130°C for 16 hours and found a mean zone separation of from 100Å to 150Å and a mean zone diameter of 80Å. It follows then that Ham's analysis should only be applied to the very early stages of zone formation while the zones are only about 10Å to 15Å in diameter.

CONCLUSIONS

The Al-27 NMR signal in an Al-4%Cu alloy increases in intensity during aging at 150° and 230°C. This change can be directly attributed to solute depletion in the matrix. Assuming that the rate of precipitation depends only on the rate of solute depletion, then the fractional increase in NMR intensity can be correlated to the fraction of solute transformed.

Our results analyzed in this way seem to confirm the approximate laws for diffusion controlled growth of second phase particles developed by Ham¹¹ for the case of platelets of constant thickness.

ACKNOWLEDGEMENTS

We would like to express our appreciation to Professor Earl R. Parker, University of California, Berkeley, for his continuous encouragement and guidance.

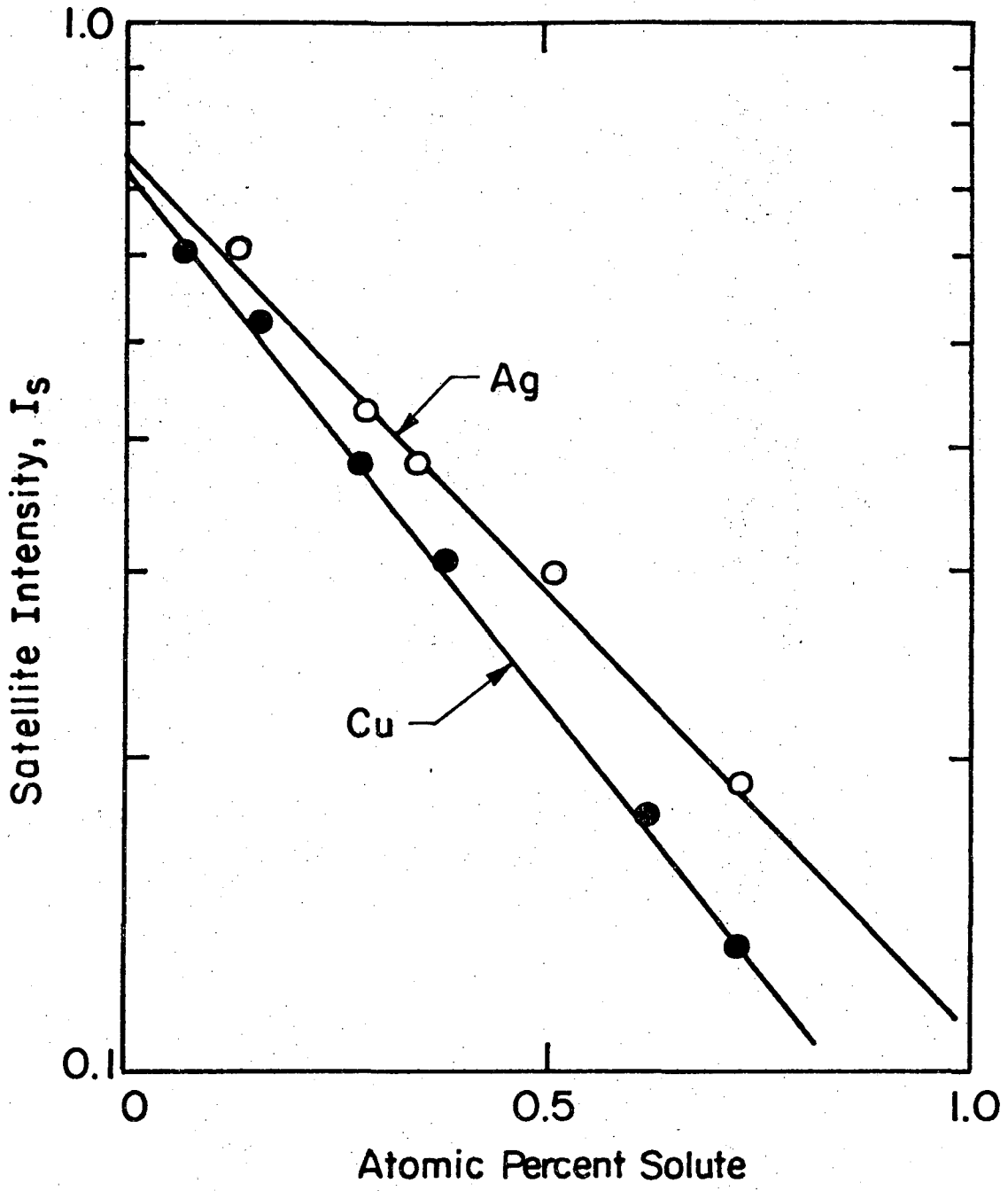
This work was performed under the auspices of the United States Atomic Energy Commission.

REFERENCES

1. Y. Fukai and K. Watanabe, Phys. Rev. B2, (7), 2353 (1970).
2. T. Rowland, Prog. Mat. Sci. 9 (1), 1 (1961).
3. W. Kohn and S. Vasco, Phys. Rev. 119, 912 (1960).
4. A. Blandin and J. Friedel, J. Phys. Radium 21, 689 (1960).
5. A. Chapman, P. Rhodes, Proc. Phys. Soc. (London) B70, 345 (1957).
6. H. K. Hardy, J. Inst. Metals 79, 321 (1951).
7. J. M. Silcock, T. J. Heal and H. K. Hardy, J. Inst. Metals 82, 239 (1954).
8. A. Guinier, Solid State Physics 9, 293 (1959).
9. C. Wert, Thermodynamics in Physical Metallurgy, ASM, 178-200 (1950).
10. A. Kelly and R. B. Nicholson, Progress in Materials Science, (Permagon Press) 10, 149 (1963).
11. F. S. Ham, J. Appl. Phys. 30, 1518 (1959).
12. R. B. Nicholson and J. Nutting, Phil. Mag. 3 (8), 531 (1958).

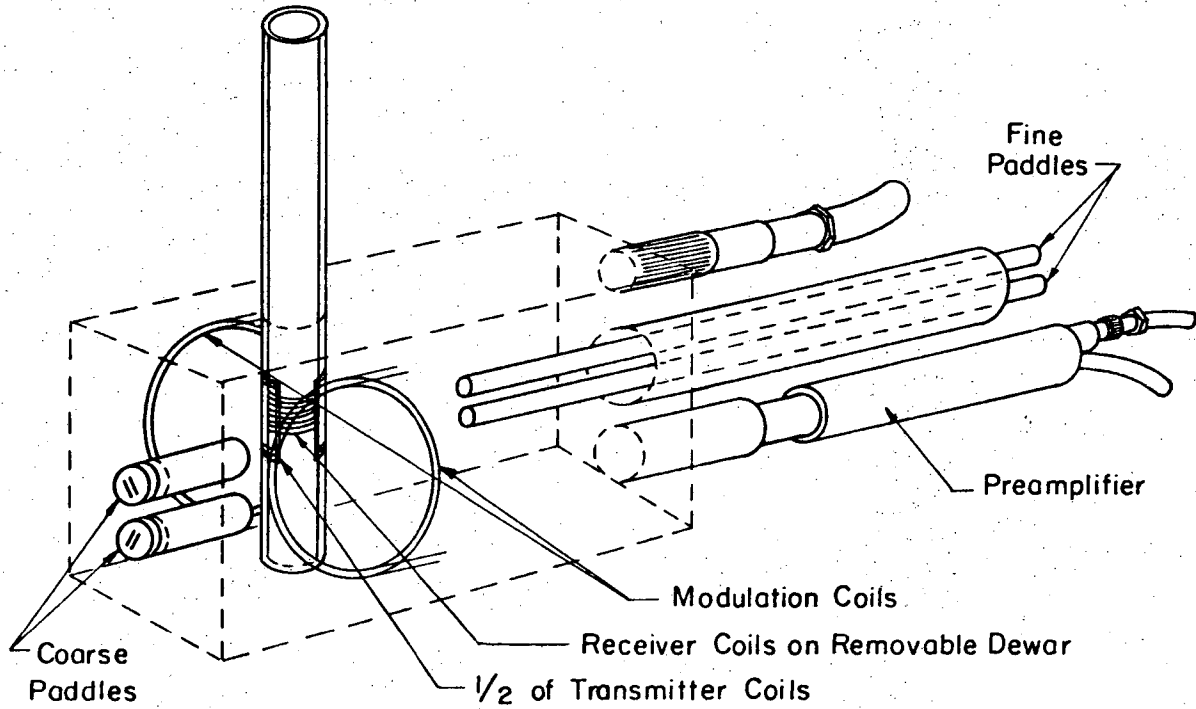
FIGURE CAPTIONS

- Fig. 1. The variation in satellite intensity with atomic percent solute: Cu and Ag in aluminum. (Satellite intensity is equal to the observed intensity minus the intensity from the central transition.) Reference 1.
- Fig. 2. Crosscoil induction NMR probe with removable dewar insert.
- Fig. 3. Dewar insert with receiver coil.
- Fig. 4. High temperature sample tube with V-51 reference sample.
- Fig. 5. Rockwell E hardness vs log time showing two-stage aging behavior. Aging of Al-4% Cu at 150°C.
- Fig. 6. Rockwell E hardness vs log time showing one-stage aging behavior. Aging of Al-4% Cu at 230°C.
- Fig. 7. NMR absorption derivative of Al-27.
- Fig. 8. The variation of absorption intensity with time at 150°C. Absorption intensity normalized to V-51 reference signal.
- Fig. 9. The variation of absorption intensity with time at 230°C. Absorption intensity normalized to V-51 reference signal.
- Fig. 10. The variation of absorption intensity with log time at 150°C.
- Fig. 11. The variation of absorption intensity with log time at 230°C.
- Fig. 12. A plot of $\ln \ln\left(\frac{1}{1-X}\right)$ vs $\ln t$ for the GP I reaction in 150°C aging.
- Fig. 13. A plot of the slope of Fig. 17 vs time. The growth exponent during GP I zone formation.
- Fig. 14. A plot of $\ln \ln\left(\frac{1}{1-X}\right)$ vs $\ln t$ for 230°C aging.



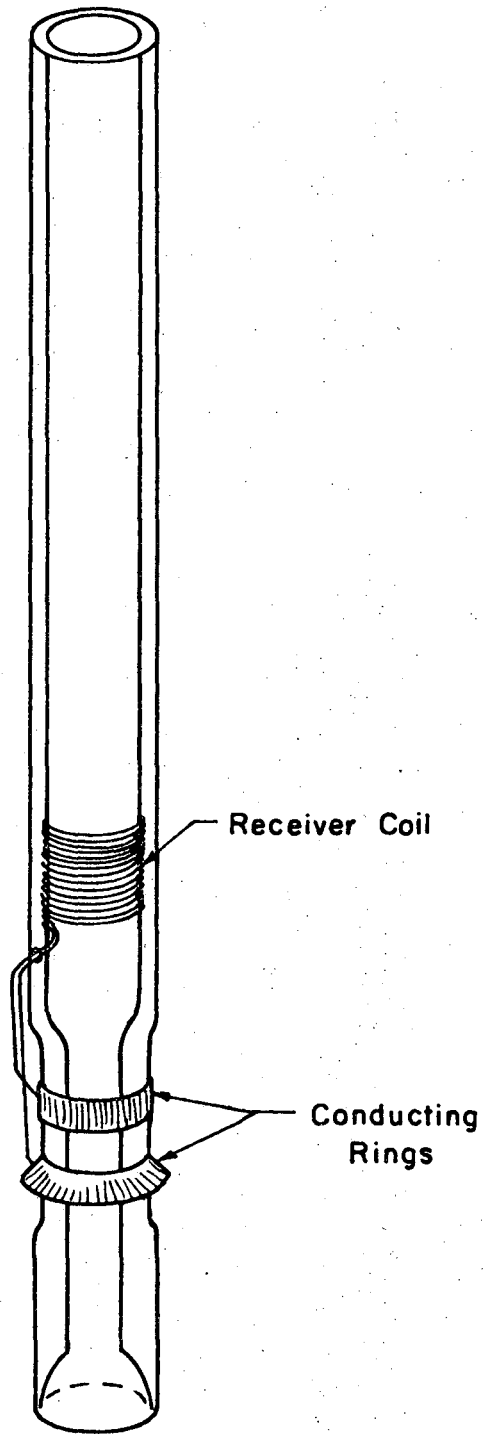
XBL 725-6317

Fig. 1



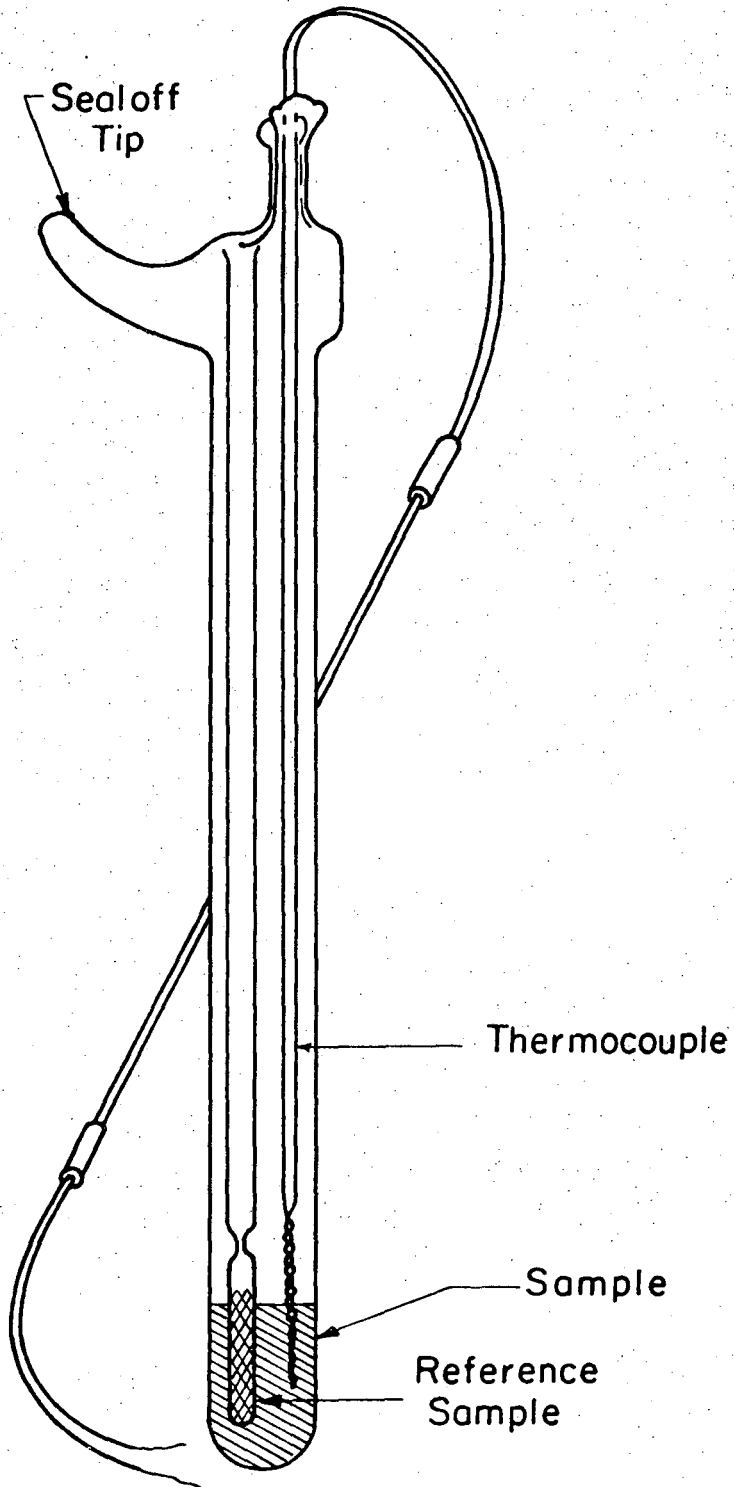
XBL 725-6321

Fig. 2



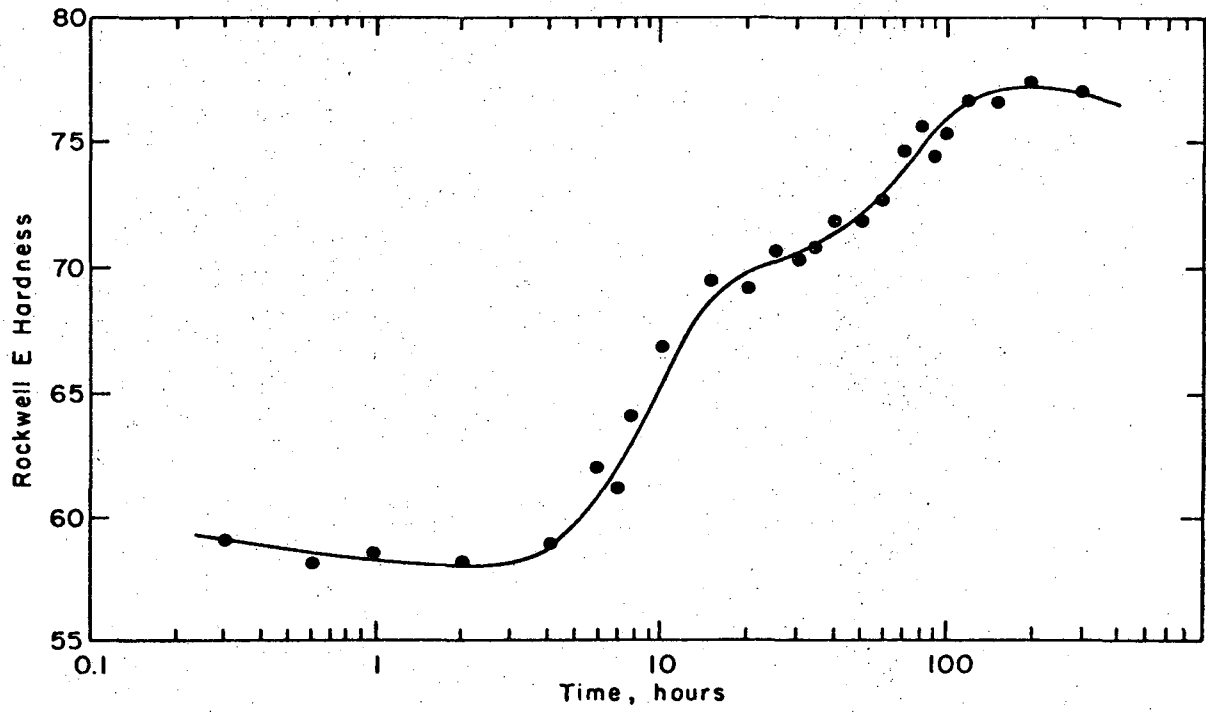
XBL 725-6322

Fig. 3



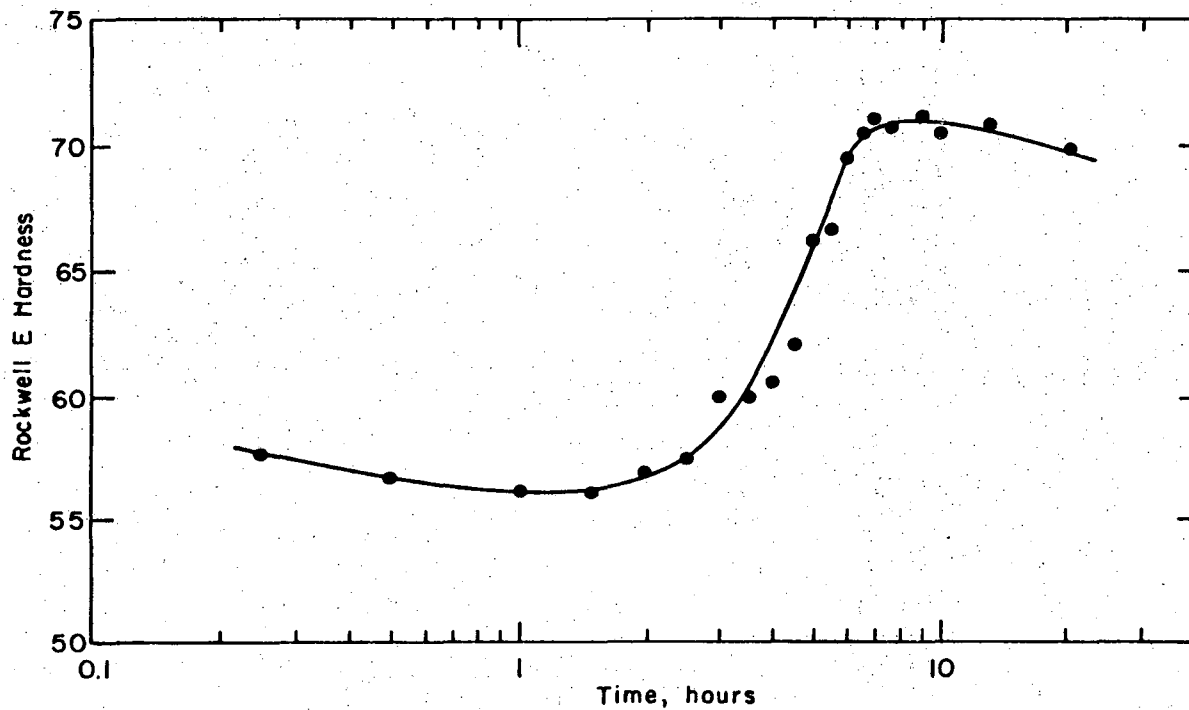
XBL 725-6324

Fig. 4



XBL 725-6325

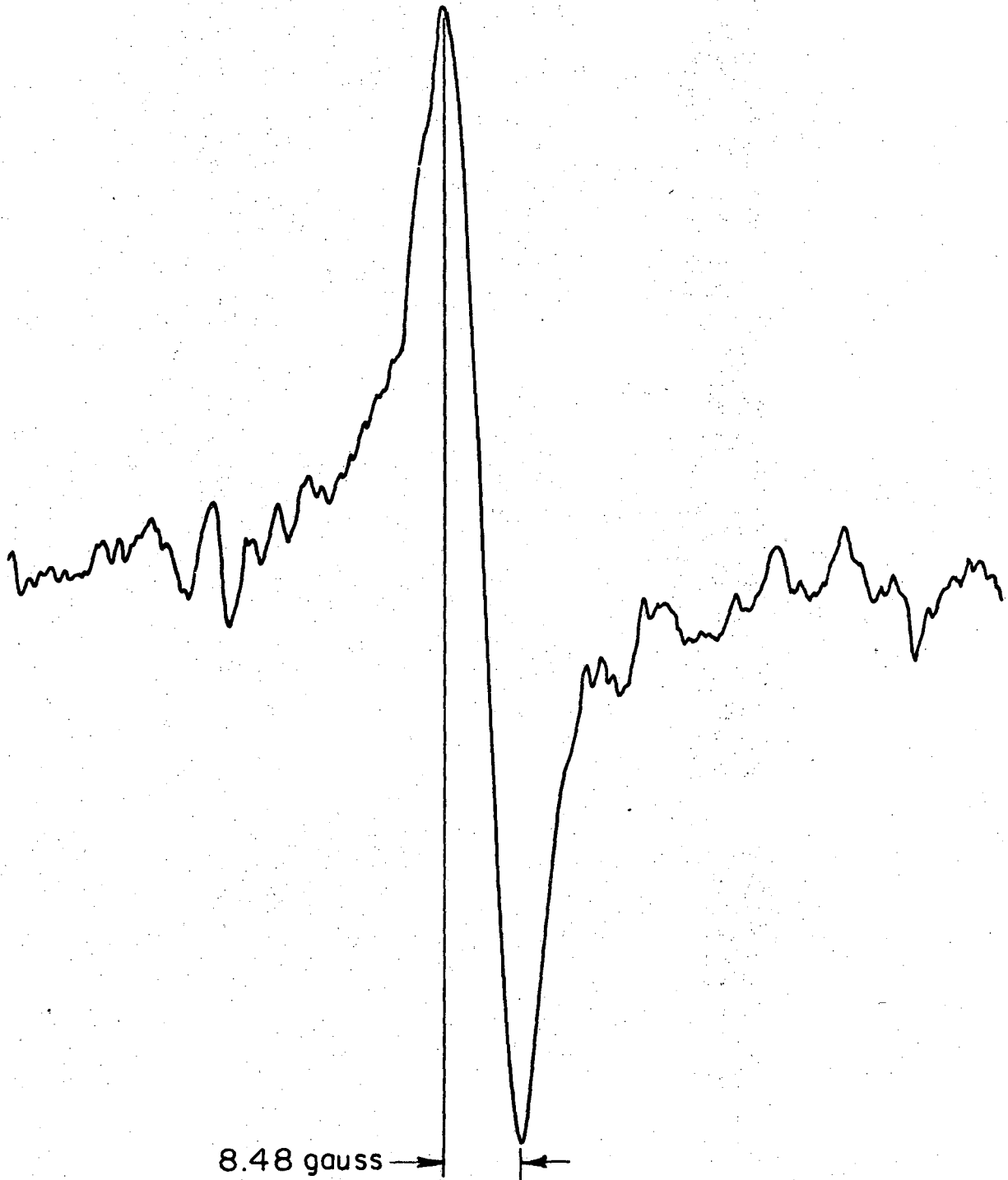
Fig. 5



XBL725-6326

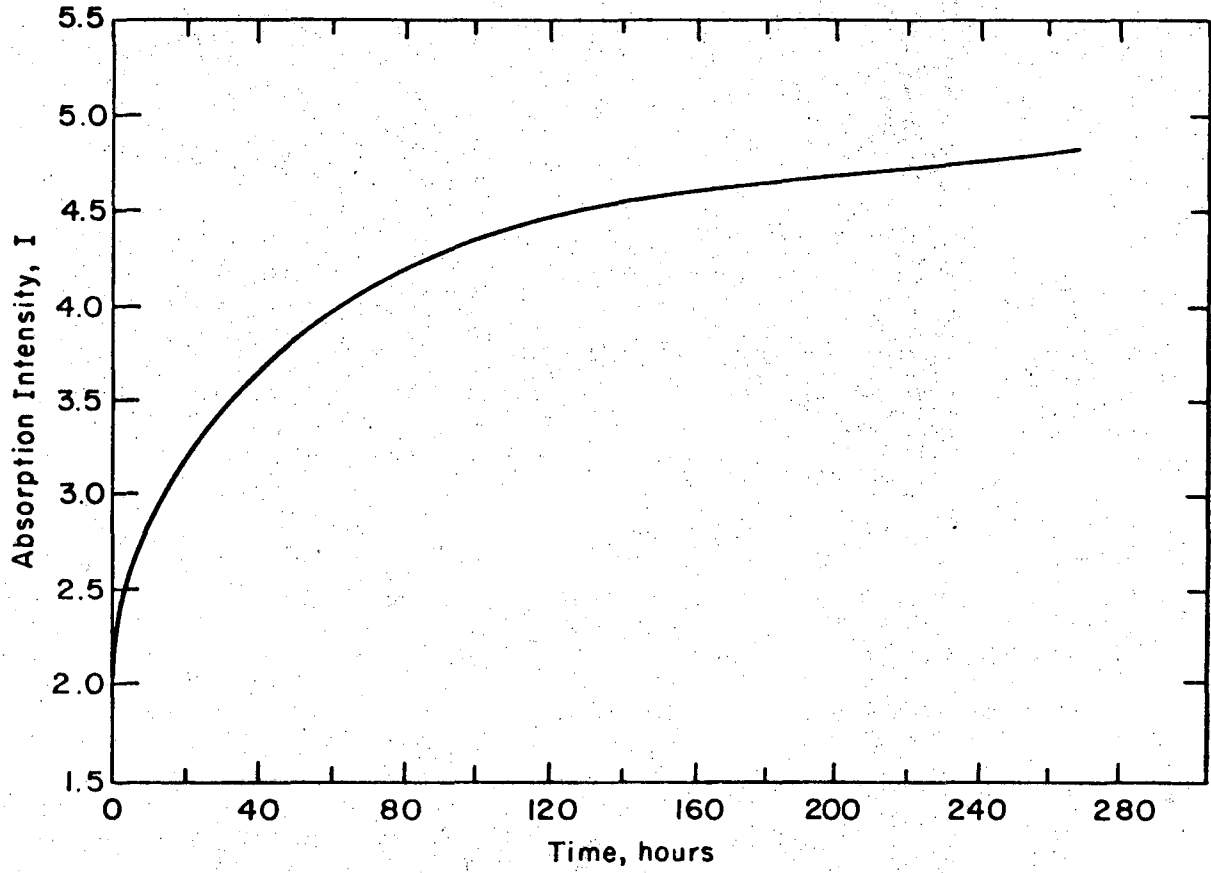
Fig. 6

-25-



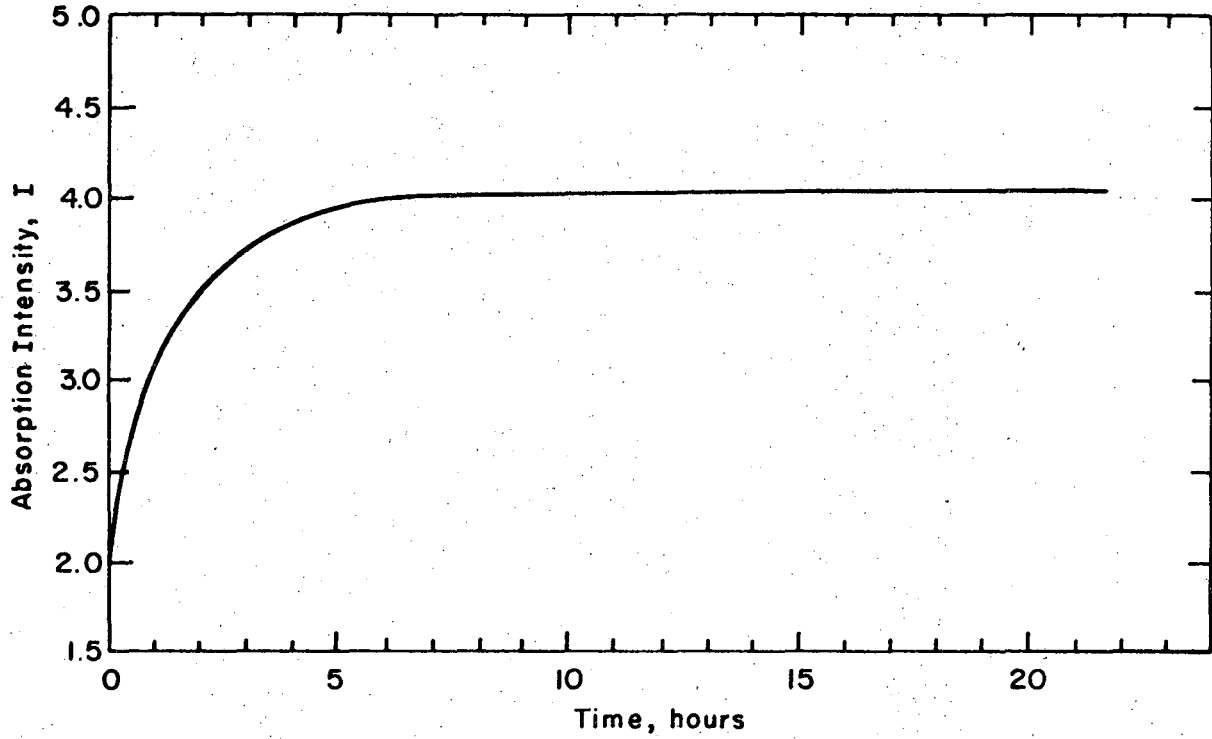
XBL 725-6327

Fig. 7



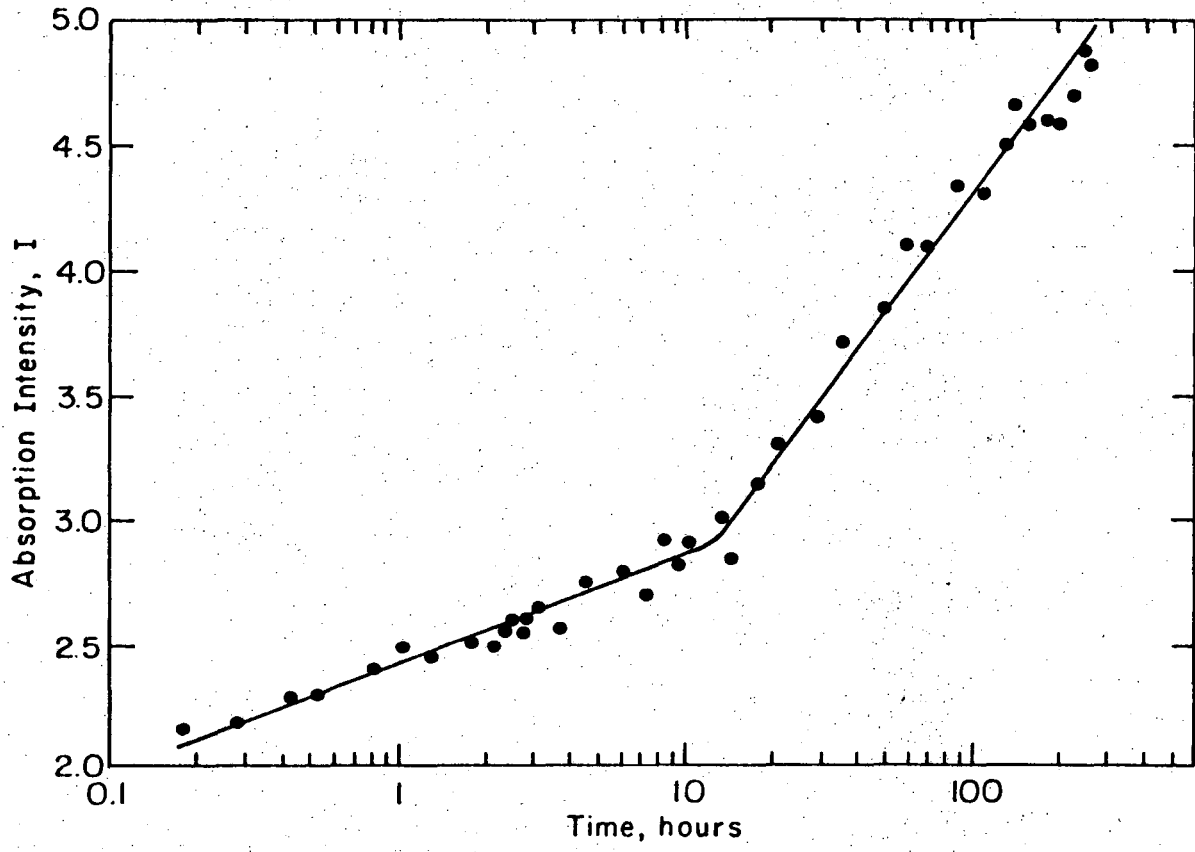
XBL 725-6329

Fig.8



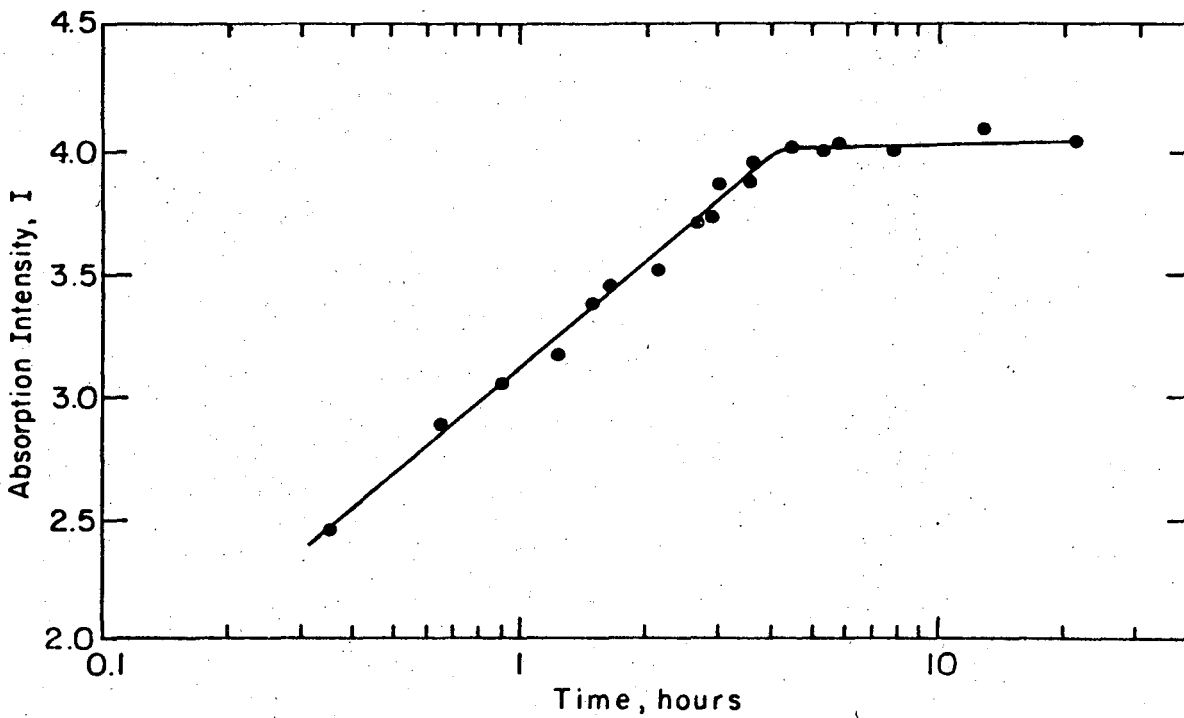
XBL 725-6330

Fig. 9



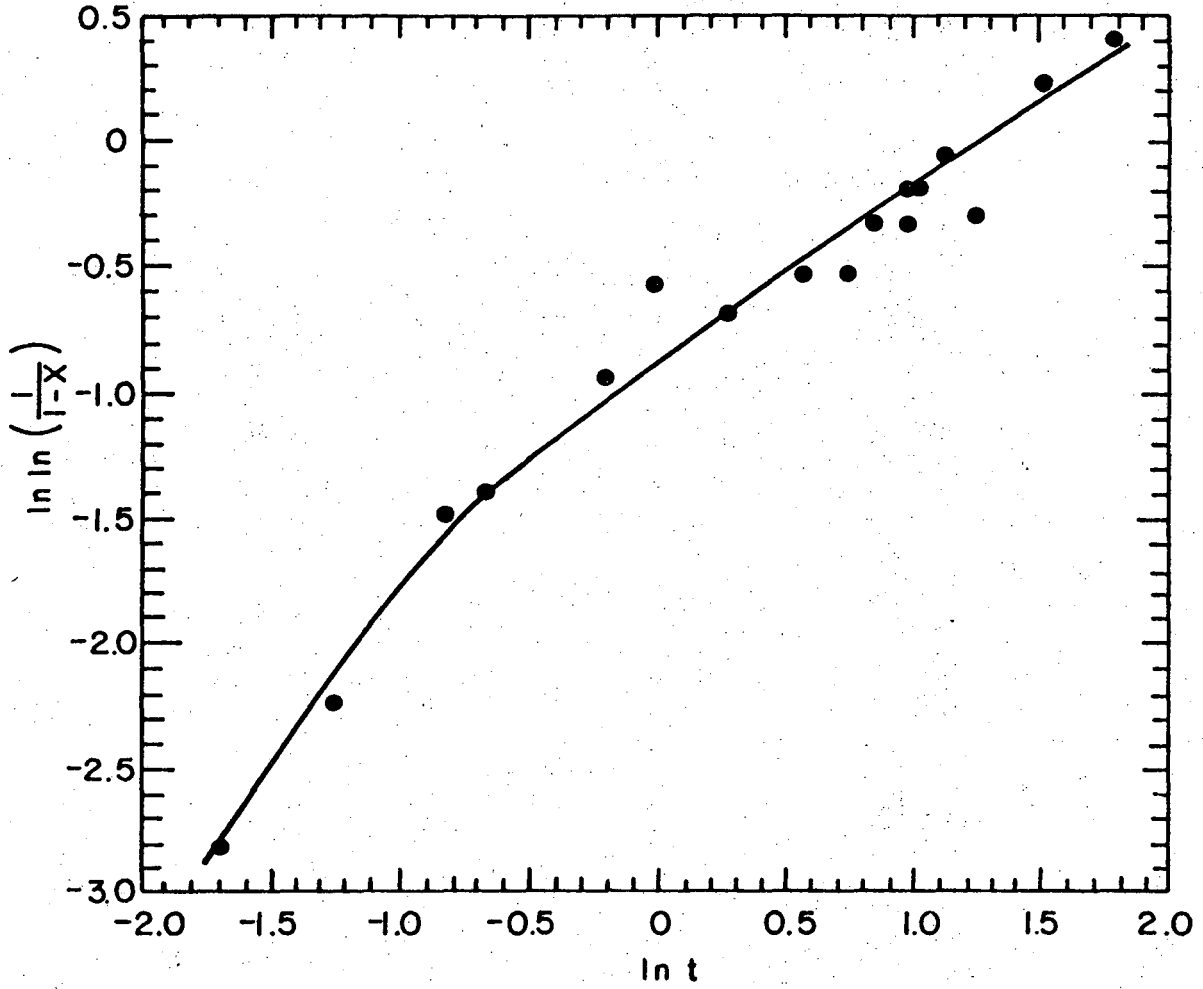
XBL 725-633I

Fig. 10



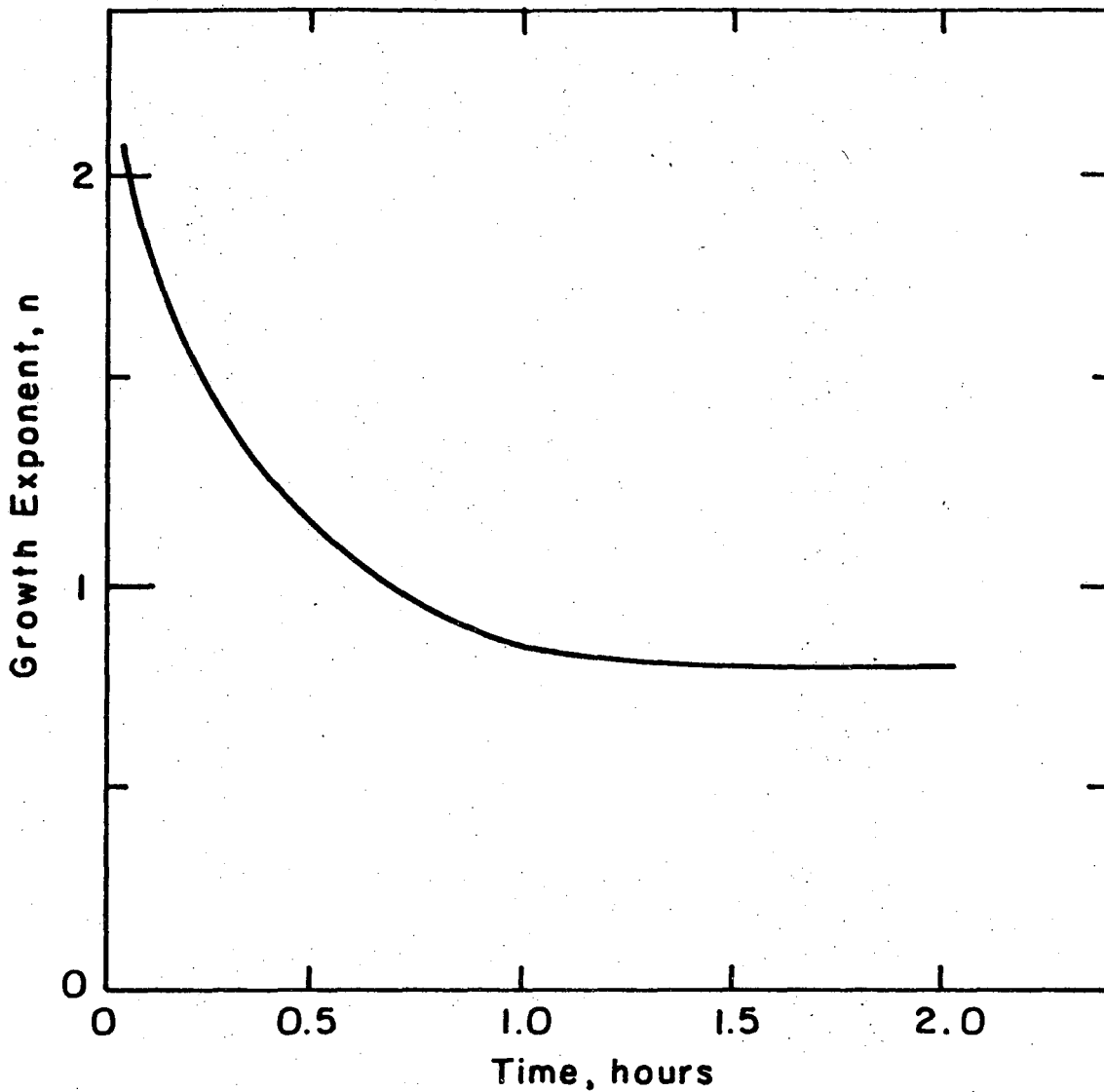
XBL725 6332

Fig.11



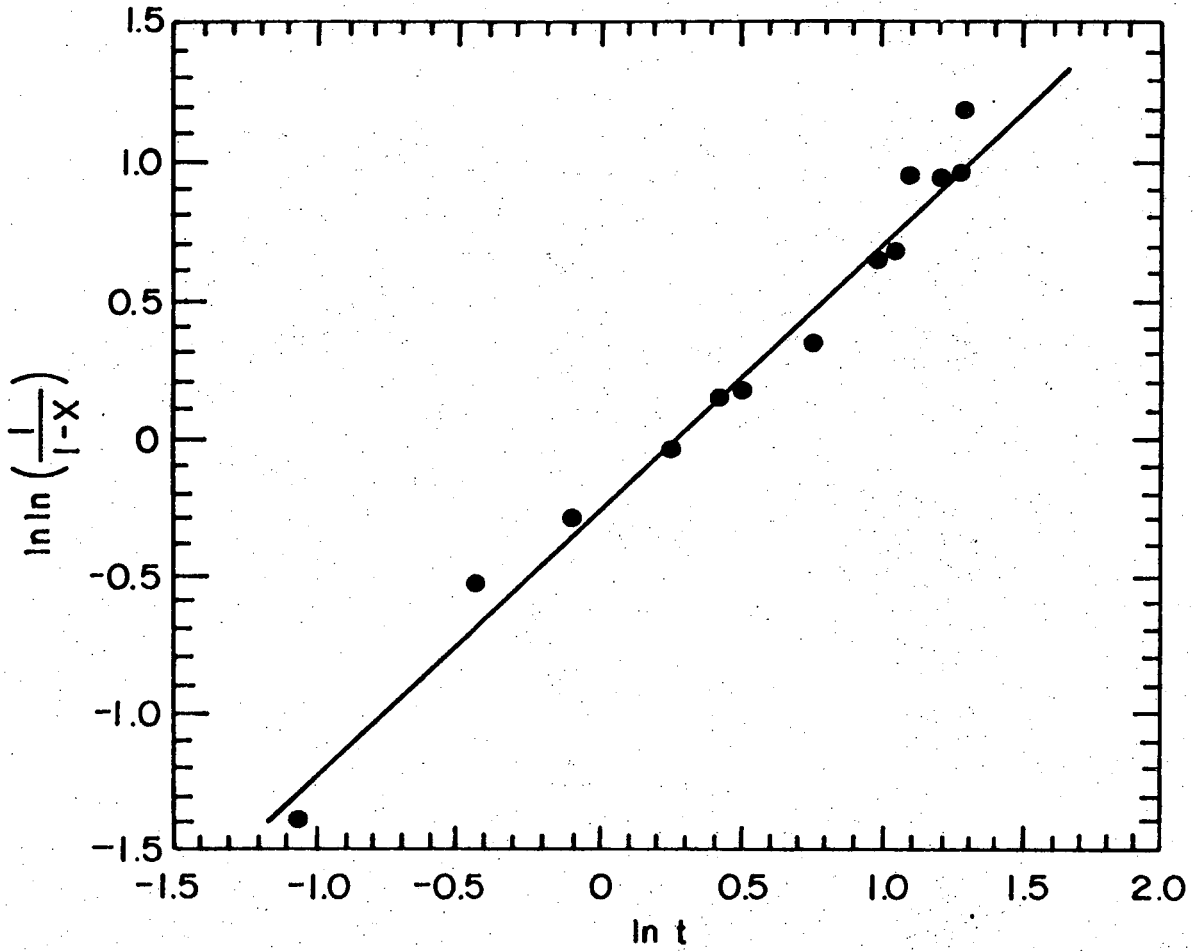
XBL 725-6333

Fig. 12



XBL 725-6334

Fig. 13



XBL 725-6335

Fig. 14

0 0 0 0 3 9 0 . 8 5 4

LEGAL NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

TECHNICAL INFORMATION DIVISION
LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720