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Thermal Simulation of Quenching Uranium-0.75% Titanium* Alloy in Water

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Abstract

A computer model, THE QUENCH SIMULATOR, has been developed to simulate and predict in detail the behavior of U-Ti alloy when quenched at high temperature (about 850°C) in cold water. The code allows one to determine the time- and space-dependent distributions of temperature, residual stress, distortion, and microstructure that evolve during the quenching process. Such analysis is vital for optimizing product characteristics and avoiding undesirable effects such as bowing, cracking, and voids. Of specific importance is the capability to incorporate the complex metallurgical phase transformations, based on a given continuous cooling transformation (CCT) diagram for U-0.75 Ti alloy. The nonlinear temperature- and microstructure-dependent properties, as well as the cooling rate-dependent heats of transformation, are incorporated into the model. The complex

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boiling heat transfer with its various regimes and other thermal boundary conditions are simulated. Experiments to determine the necessary physical, thermophysical, and mechanical properties for the various microstructures and the applicable boiling curve have been performed and incorporated into the model. Both sudden submersion and gradual controlled immersion can be applied. The final output provides detailed information on the predicted characteristics, including microstructure distribution, in tabulated form, contour plots, and color-coded graphics for one-, two-, or three-dimensional geometries. The code is general enough to potentially be used for other quenched materials and with other quenching liquids. A parametric and sensitivity study has been performed demonstrating the importance of the thermal boundary conditions applied for achieving certain product characteristics. The thermal aspects of the model and its application are discussed and demonstrated.

1. Introduction

The purpose of this study is to develop the analytical tools necessary for optimizing the quenching process of uranium-0.75% titanium (U-0.75% Ti) alloy. The goal in quenching the alloy is to achieve maximum hardening both in quantity and quality by changing the microstructure from the γ phase, which is stable at temperatures above 723°C, to the desired α' phase (called martensite) through rapid cooling. In general, the faster the cooling rate the larger is the percentage of the martensite formed and the deeper is the zone of the desired transformation. Although the tools developed in this study are general, it is more convenient to consider a specific process and a specific shape. In this case, a cylindrical form is assumed with 32-mm diameter and 360 mm long. The material being quenched, which is initially at 850°C, may be either submerged at once into a 20°C pool of water or is immersed gradually at a constant rate. To increase the cooling rate, the water in the pool can be agitated from the bottom, or horizontal high velocity jets can be impinged on the quenched material.

A quenching process, however, can result in a number of production difficulties. Too slow a cooling rate can result in too small a percentage of the desired martensite material; too rapid a cooling rate can result in overstresses which may crack and/or deform the part or it may create internal voids inside the material. Nonuniform boundary conditions as well as nonuniform material composition may also cause axial distortion (bow). Factors that may produce nonuniform boundary conditions include:

1. nonuniform surface finish,
2. nonuniform oxide coating on the surface,
3. nonvertical orientation of the immersing cylinder,
4. nonuniform fluid dynamic,
5. cooling jets aimed nonuniformally.

The analytical tools being developed should be useful for improving the production process so that martensite formation will maximize; whereas cracks, deformation, and voids will be minimized or be eliminated altogether. Such analytical tools should simulate the quenching process both on the surface (thermal boundaries) and internally (metallurgical microstructure transformation including the complex process of energy release during the transformation). The simulator should predict the time- and space-dependent distributions of temperature, residual stresses, distortion, and microstructure that evolve during the quenching process. The data required for that simulation, such as the thermal boundary conditions (boiling curves) and the thermophysical and mechanical properties, were not readily available and had to be generated through either theoretical or experimental studies, as part of the objective of this study.

2. THE QUENCH SIMULATOR and its Components

To achieve the objective discussed in the introduction, general computer programs had to be chosen which will allow maximum flexibility in modifying and incorporating the necessary features of the quenching process. The programs chosen were:

HEATING6 - a finite difference three-dimensional transient heat transfer program (Ref.1) and

ADINA - a finite element three-dimensional stress and deformation program (Ref. 2).

These two programs are very general and can be used to simulate very complicated situations involving thermal and structural aspects. However, five unique features of the U-Ti alloy quenching process required development and incorporation of special subroutines:

1. simulation of the exothermic microstructure transformations whose transformation temperatures, and heats of transformation, are complex functions of time-temperature relationships;
2. simulation of nonlinear thermal boundary conditions encountered in the boiling process;
3. simulation of the moving boundary conditions created by the controlled immersion motion of the quenching cylinder relative to the water surface;
4. simulation of temperature and phase dependent thermophysical and mechanical properties of the various microstructues;

5. simulation of an impinging horizontal jet aimed at the vertical immersing cylinder.

Each of the above issues had to be determined and/or developed through both analytical and experimental studies since the basic data were not always available.

The microstructures of interest for the present study are those predicted by the continuous cooling transformation (CCT) for U-0.75% Ti (Fig. 1). The desired α' phase (martensite), forms by rapid cooling to temperatures below 615°C, and is stable below 400°C but decomposes to the undesirable ($\alpha + \delta$) microstructure when held between 615°C and 400°C. As can be seen from Fig. 1, complete conversion of the alloy to α' would require a cooling rate of approximately 200°C/s, whereas cooling rate of less than 20°C/s results in essentially no martensite formation at all and 100% formation of the ($\alpha + \delta$) phase. During any phase transformation heat is liberated, causing corresponding delay in the rate of cooling at the specific location undergoing transformation. A special user-supplied subroutine named HEATGN, which simulates the CCT diagram represented in Fig. 1, was developed and incorporated into HEATING6. For each given node and time step, the amount of each phase present can be read by a subroutine named PHSMAP, from the CCT diagram as a function of its temperature, the time since it passed the critical temperature, and its past history. The amount of heat of transformation released at a given node is determined and entered into the problem as a heat generation rate for the current time step.

The following thermophysical and mechanical properties, which were of prime importance to this study, have been determined experimentally as a function of temperature for the γ, α', and $(\alpha+\delta)$ microstructures and incorporated into the QUENCH program as user-supplied information:

heats of phase transformation,

temperatures of phase transformation,

specific heat,

density,

thermal diffusivity,

thermal conductivity,

thermal expansion coefficient,

Young's modulus,

Poisson's ratio,

strain hardening modulus,

yield strain,

ultimate strain,

yield stress.

This determination was extremely difficult because of the transient nature under which phase decomposition occurs. The methods, constraints, and assumptions used in these experiments are discussed in Ref. 3.

The most important aspect of the quenching process is the heat transfer rate between the quenched material and the quenching fluid. This rate is controlled by the boundary conditions between the solid surface and the surrounding fluid, in this case, water. This boundary condition is dominated by the boiling heat transfer phenomena, which is a very complex process and its behavior is very sensitive to numerous geometrical, physical, and thermodynamical characteristics of both the quenching fluid and the quenched material. The "correct" boiling curve to be used in this study will need special investigation which requires careful experimental and analytical development. For the time being, a general boiling curve, shown in Fig. 2, has been developed based on the most applicable steady state correlations available in the open literature and is used throughout the current work. Because of the high initial temperature of the quenching alloy, the dominating heat transfer mechanism is that of film boiling. The nucleate boiling zone with its highly efficient heat transfer mechanism exists only within a narrow band of temperatures and at the tail end of the quenching process. Techniques that are available for augmenting and reshaping the nature of the boiling curve include: 1. adding resistance between the quenched solid and the boiling liquid by coating material, 2. adding a thin layer of low effusivity ($k\rho Cp$) material such as zirconia (Ref. 4), 3. subcooling of the quenching liquid (Ref. 5), 4. polishing or increasing the roughness of the solid surface (Ref. 6), 5. agitation and direct water jetting which tends to destabilize the film boiling regime and augment heat transfer (Ref. 7), and 6. use various combinations of additives and types of quenching fluids (Refs. 5 and 8).

All of the above techniques are based on destabilizing the film boiling regime (that means creating solid liquid contact) sooner in the quenching process and maintaining a nucleate boiling state for as long as possible. The analytical tools developed in this study can be extremely useful in studying the potential effects of these techniques through parametric analysis. Boiling correlations normally call for an experiment to be performed with the specific combination of solid and liquid being used. Such an experiment is indeed currently in progress for the quenching of U-0.75% Ti alloy in water. However, there are two main problems associated with performing an applicable experiment. First, the quenching process on hand involves a very fast transient (cooling from about 850°C to 20°C in about 15 s), whereas the traditional approach of correlating boiling curves is directed toward steady state relationship (no time parameter involved). Many investigators, including Peyayopaskul and Westwater (Ref. 9), Bergles and Thomson (Ref. 10), and Bui and Dhir (Ref. 11), have shown that the boiling curve generated under fast transient is quite different from the one generated for steady state under the same conditions. Other investigators including Lin and Westwater (Ref. 12), Moreaux and Beck (Ref. 5), and Henry (Ref. 13) have indicated the dependence of the boiling curve (primarily the destabilization of the film boiling regime) on the thermal properties of the material itself, k, ρ, and C_p in the form of a product ($k\rho C_p$) called superficial effusivity. Second, the material of interest is U-0.75% Ti. The thermophysical properties of its various microstructures and its matallurgical characteristics during transformation are all somewhat uncertain. However, the data reduction methods used to determine the heat transfer coefficient from experimental data, such as the inverse heat transfer solution method by Beck (Ref. 14), are strongly dependent on good knowledge of the rest of the parameters and a good direct solution of the system at hand.

Recognizing the above uncertainties, a careful experimental plan is being implemented for developing the desired thermal boundary conditions. Fortunately, the film boiling regime itself, which prevails during the majority of the quenching process, is not very sensitive to either the surface characteristics or the rate of transient cooling. It is proposed that the experimental data be reduced and correlated to include and reflect the transient effects, if those are determined to be important, through a newly introduced time parameter: the rate of cooling on the wall surface, i.e., ($\Delta Tw/\Delta t$) as shown in Fig .3. Such a representation of the transient relationship will allow its use in a general transient code such as the QUENCH simulator.

Another unique boundary condition is the immersion process of the cylinder into the water through the water interface. This requires changing the appropriate heat transfer coefficients as each specific node moves from the environment of natural convection and radiation into the environment of boiling. To do that, a scheme was devised and implemented in a user subroutine called CONVTN that keeps track of the cylinder movement. If only part of the nodal area is submerged, then only that part is subjected to boiling heat transfer conditions, while the remainder is subjected to natural convection and radiation to the ambient air.

Finally, the simulation of the horizontal stationary impinging jet also poses a unique boundary condition in relation to the vertically moving cylinder. As the cylinder moves, the zone of water impingement changes positions along a vertical line on the surface. The simulation of that process has been developed and incorporated into the model, but not in time to be used in this study. Instead, the point jet is assumed to act as a slot jet which is applied vertically all along the length of the cylinder.

3. The General Quench Simulator

The unique features described in the previous section were integrated into the general heat transfer code HEATING6 to provide the thermal component of the QUENCH simulator and into the general structural code

ADINA to provide the stress and deformation component. To this package a number of service routines was added to facilitate interfaces and to provide the desired plots and graphics. Figure 4 represents an overall perspective of the relationship between the various analytical and experimental tasks in the program.

The general code needs to be tailored to the specific case to be analyzed by providing its geometry, dimensionality and the proper boundary conditions. It is also possible to expand the applicability of the code to other alloys and maybe processes by providing a new set of thermophysical and mechanical properties and an appropriate HEATGN and PHASE subroutines, the two subroutines which reflect the metallurgical behavior of the alloy during phase transformation.

The final output of the code provides detailed, time- and space-dependent information on each of the predicted characteristics which include:

temperatures,

cooling rates,

microstructures,

all stress components,

deformation and distortion.

This information is provided in tabulated form in time-dependent plots for any selected single node or in iso-contour plots for any desired cross section and time. Color graphics of the same contours are also be provided.

4. Thermal Parametric Studies

The objective of the parametric studies was to demonstrate the capability of the techniques employed in the analysis and to study the effects that various parameters in the quenching process have on martensite formation. A second objective was to observe trends and then formulate plans for future studies to improve production, including the various ways of augmenting boiling characteristics (agitation, jets, additives, coating, etc.). The operating parameters as well as material and boundary condition characteristics covered in this study include:

effect of temperature-dependent properties,

effect of heat of transformation,

effect of water temperature,

effect of jet impingement,

effect of oxide coating,

effect of immersion rate,

effect of boiling curve variations,

effect of insulating the bottom.

Four designated heat transfer models have been prepared for performing the thermal parametric studies. This allowed a more economical use of human and computer resources depending on the objective of the parameter being studied. The four models simulated were:

1. one-dimensional radial (R) model which was used to study the effects of temperature-dependent properties, heat of transformation, water temperature, uniform oxide coating, and boiling curve variations on sudden submersion cases;
2. two-dimensional model in R and Z directions, which was used to study

the effects of immersion rates;

3. two-dimensional model in R and ϕ directions, which was used to study the effects of nonsymmetrical boundary conditions on sudden submersion cases;

4. three-dimensional model in R, ϕ, and Z directions which was used to study any of the effects listed above including immersion and nonsymmetry, but was obviously much more costly to run. This was also the only model that could be used by the stress component of the code to simulate a bow deformation.

These four models were used to perform the parametric study by first running a base case on the one-dimensional model with "standard" parametric values and then successively altering the investigated parameter and comparing results to the base case. A summary of the cases run is given in the following table.

The base case was run using the "standard" parameters which were as follows: sudden submersion, no oxide coating, constant thermophysical properties, correct heats of transformation, standard "chopped" boiling curve (standard "chopped" boiling curve uses heat transfer coefficient of 0.02 W/mm-K below T of 178°C for nucleate boiling, and 0.0025 W/mm-K above T of 178°C for film boiling), symmetrical boundary conditions, and 20°C quenching water. Figure 5A shows the temperature as a function of time for three nodes of interest: the surface node, the center node, and a node in between (8 mm radius). One can clearly see the effect that heat of transformation has on the central node. The effect on the surface node is unnoticeable because of the extremely high heat transfer rate which dominates the process. Also very noticeable is the effect of the

Case	R	Th	Z	V	O	P	H	BCN	Tb	DIS
base	33			0	N	C	+	S	20	
1	33			0	N	T	+	S	20	
2	33			0	N	C	0	S	20	
3	33			0	N	C	+	S	100	
4	34			0	Y	C	+	S	20	
5	33			0	N	C	+	0.0025/ 0.04	20	
6	33			0	N	C	+	0.0025/ 0.01	20	
7	33			0	N	C	+	0.0025	20	
8	33			0	N	C	+	0.005	20	
9	33			0	N	C	+	0.0075	20	
10	33			0	N	C	+	0.02	20	
11	33			0	N	C	+	5 S	20	
12	33			0	N	C	+	10 S	20	
13	17	13		0	N	C	+	S	20	
14	17	13		0	N	C	+	S	20	90
15	5		25	6.35	N	C	+	S	20	
16	5		25	7.62	N	C	+	S	20	
17	5		25	9.14	N	C	+	S	20	
18	11	7	25	7.62	N	C	+	S	20	

Nomenclature used in the above table include.

R,TH,Z = number of nodes in radial, angular and axial directions
V = immersion velocity (0 = submersion), mm/s
OC = oxide coating (Y = yes, N = no)
P = properties
H = heat of transformation (+ = yes, o = no)
BCN = boundary conditions (S = standard boiling curve) (see text for discussion,fluxes in W/mm.K)
Tb = boundary temperature (bulk water), C.
DIS = angular distribution of BCN (nonsymmetry)

transition from film boiling to nucleate boiling on the surface node. The same effects can also be seen in Fig. 5B where the cooling rates ($\partial T/\partial t$) are shown.

Case 1 applies the physical properties as a function of temperature to study the effect when compared to using constant properties. There was very little difference between the two cases. For that reason, it was decided to use constant properties throughout the study to reduce computer cost.

Case 2 was run with no heats of transformation included. Comparing the results shows that the effect is very considerable. Figure 6 demonstrate that comparison side by side. It can be seen that there is a major delay in the cooling rate as a result of the heats of transformation involved. This demonstrates very convincingly the importance of the QUENCH simulator in studying the quenching process.

Case 3 applies a quenching water temperature of 100°C instead of 20°C and the impact is as expected: surface as well internal nodes temperatures are raised by about 80°C with the cooling rates being slower but not by much.

Case 4 applies a 200-micron coating of zirconia on the outer surface to study the effect it has on the boiling characteristics. The original idea was to test the known fact that some resistance coating on a boiling surface can, under certain conditions, cause the film boiling to be replaced by nucleate boiling and, therefore, improve cooling rates. This case apparently did not perform as it was hoped, and the cooling rates were substantially lower. This, however, requires further studies in order to achieve, if possible, the right conditions.

Cases 5 to 12 are all designed to study the effects of certain alterations in the boiling curve for either the whole boiling curve, only the nucleate boiling regime part, or only the film boiling regime part. The interest in running such cases is twofold; first, to find out the sensitivity of martensite formation to the correctness of the values used in the boiling curve and, second, to find out if efforts to augment and "reshape" the boiling curve are worthwhile. Cases 5 and 6 showed that the effect of changing the heat transfer coefficient for the nucleate regime on temperatures or cooling rates on both the surface nodes or internal nodes is negligible until a shift to nucleate boiling occurs. However, by the time that shift occurs, all the temperatures including the central node, are much below the transformation temperatures of the alloy. The net result is no effect at all on martensite formation.

Cases 7, 8, 9, and 10 take the other approach in which the film heat transfer coefficient is multiplied by a factor of 1, 2, 3, and 8, correspondingly, and assumed constant. As opposed to Cases 5 and 6, these cases demonstrated a very strong influence on both the temperature profiles as well as on the final martensite formation. This can be seen very well in Fig. 7 for percent of martensite formation (100% martensite was achieved in Case 10). It became very clear then that changes in the film boiling regime have a very marked influence on martensite production, whereas similar changes in the nucleate boiling regime do not.

In order to test the combined effect of changing both the nucleate and film boiling regimes, the heat transfer values of both have been multiplied by a factor of 5 and 10 in Cases 11 and 12, correspondingly. Again, a very marked influence can be seen on both the temperature profiles and the martensite formation. This, however, is primarily the result of increased film boiling heat transfer coefficient.

The overall conclusion that can be derived from Cases 5 to 12 is that the heat transfer coefficient has a dominant effect on the quenching process with most of the influence coming from changes in the film boiling regime. However, that relationship will be dependent on the relative rate between heat transfer by conduction radially inward versus the transient rate of cooling of any given node. For any increase in cooling rate at any specific point to cause an improvement in martensite formation, it needs to reach that point before transformation to $(\alpha + \delta)$ has occurred. Any cooling rate increase after that time is useless as much as martensite formation is concerned. Therefore, the above derived conclusions might be different for different alloys or even for the same alloy but of different dimensions or shape.

Case 13 is a two-dimensional (R-ϕ) case in which the influence of nonsymmetrical boundary conditions were studied, and the resulting nonsymmetrical martensite formation was very apparent.

Cases 15, 16, and 17 are two-dimensional (R,Z) models designed to study the influence of immersion rates.

Case 18 is a three-dimensional (R,Z,ϕ) model in which the effects of both nonsymmetrical boundary conditions and immersion can be seen at the same time. A three-dimensional model is also necessary for performing stress analysis to study the resulting deformation and bow. An immersion rate of 7.62 mm/s and nonsymmetrical standard boiling curve were used in this case. Nonsymmetrical boundary conditions were simulated by applying a standard boiling heat transfer at the angle of 0.0 degrees which was gradually increased up to twice that value at the angle of 180 degrees. Figure 8 shows the temperature and phase distributions in 10-s intervals for the (R-Z) cross section at 180 degrees angle, whereas Fig. 9 does it for temperatures on a (R-ϕ) cross section at 180-mm elevation. The sequential process of martensite formation with time as the cylinder penetrates the quenching pool can be clearly seen in Fig.88 and the nonsymmetrical distributions are very noticeable in Fig. 9.

5. Conclusions and Recommendations

The study demonstrated the feasibility of analytically simulating in great detail the complex quenching process of U-0.75% Ti alloy. The models developed take into account most of the factors influencing the process, including the complex phase transformation diagram, and are capable to analytically predict temperature distributions, cooling rates, stresses, deformation, phase composition, and possibly even the prediction of void formation, all as functions of both position and time. Such an analytical tool can be used to understand, study, and optimize the quenching process and improve the desired martensite formation both in quality and quantity.

The thermophysical and mechanical properties of U-0.75% Ti, as well as the phase transformation temperatures and the related heats of transformation, have been determined through experiments which were guided by the data requirements of the models developed. Other experiments are under way to determine the proper boiling heat transfer coefficient to be used.

It was concluded that heat of transformation, which constitutes about 19% of the total heat transfer involved in the process, is a significant factor affecting the metallurgical behavior of the alloy during the quenching operation.

On the other hand, it was concluded that the process is not very sensitive to the dependence of the property data on temperature. For this reason, there are no immediate plans for obtaining more accurate physical and mechanical data. It was demonstrated that nonsymmetrical boundary conditions can have a major impact on the nonsymmetry of the product including the creation of bowing deformation.

The thermal boundary conditions constitute a very important factor in the metallurgical behavior during quenching. However, most of the influence is in the early part of the process before phase transformation occurs at any specific point. The film boiling regime is the regime of most importance, whereas the nucleate boiling regime plays a minor role in influencing the final product. This, however, may not always be the case because of the time lag involved in the penetration of the thermal effect from the surface, where it initially starts, toward the central portion of the body being quenched. The size, shape, and thermophysical properties of the quenched alloy will determine the importance of this relationship. Effort in augmenting the heat transfer should consider these facts.

It is recommended that effort in the future should be put in further determining the relative importance of various parameters and conducting experiments to either acquire the necessary data or augment factors, such as the boiling curve characteristics, which were determined to have the most potential for improving the quench process. It is also recommended that the models developed in this study be applied to other alloys than U-0.75% Ti as well as to other material science processes where detailed information on internal behavior is desired or necessary.

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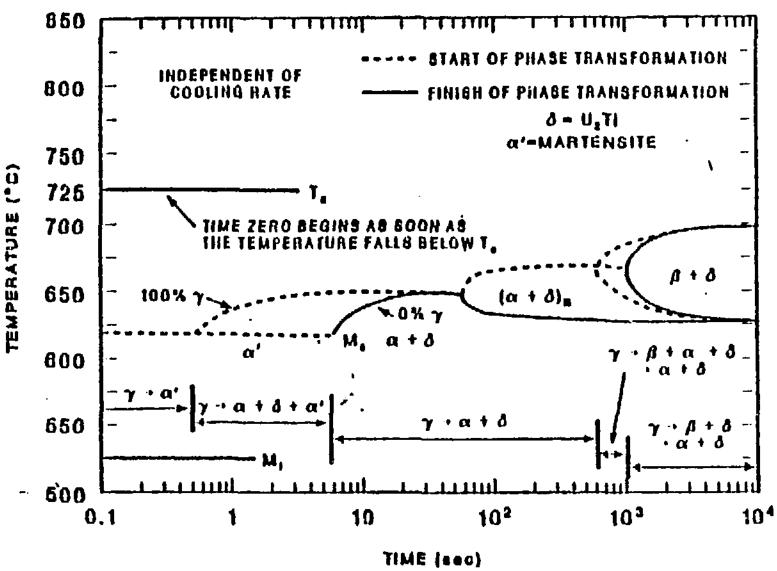


Fig. 1. Phases of U-0.75Ti alloy as functions of time and temperature for continuous cooling.

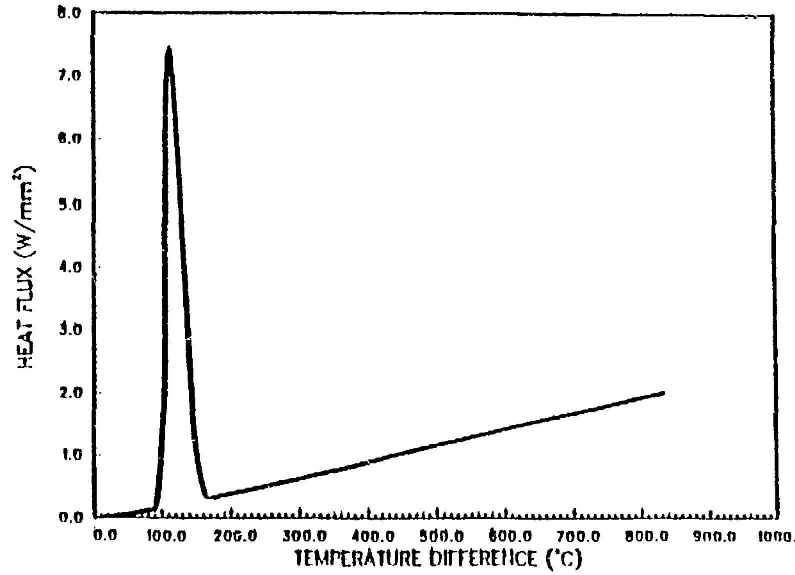
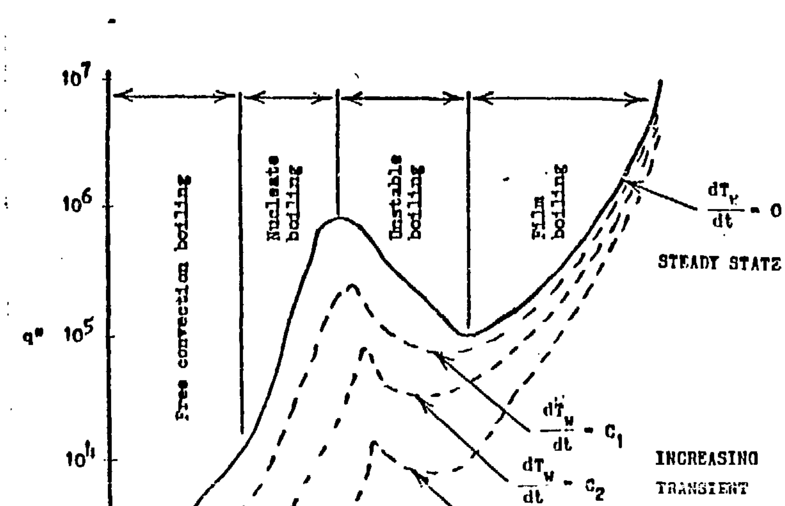
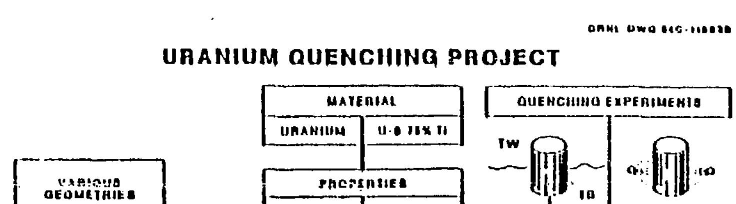


Fig. 2. Linear plot of heat flux as a function of wall temperature difference ($T_w - T_b$).





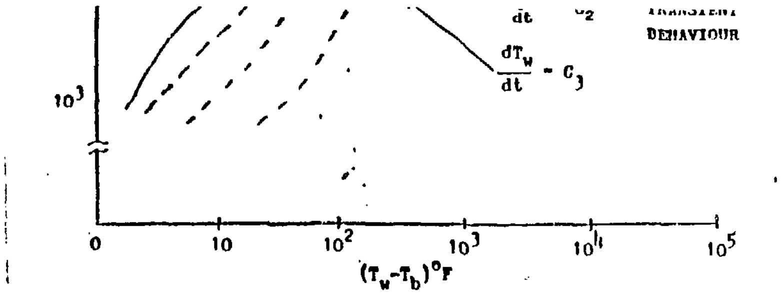


Fig. 3. Proposed way of correlating transient effects of boiling heat transfer. Wall cooling rate is a parameter.

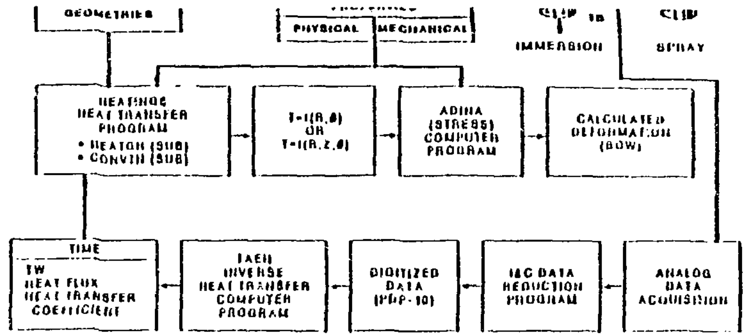


Fig. 4. Relationship of analytical and experimental efforts on quenching project.

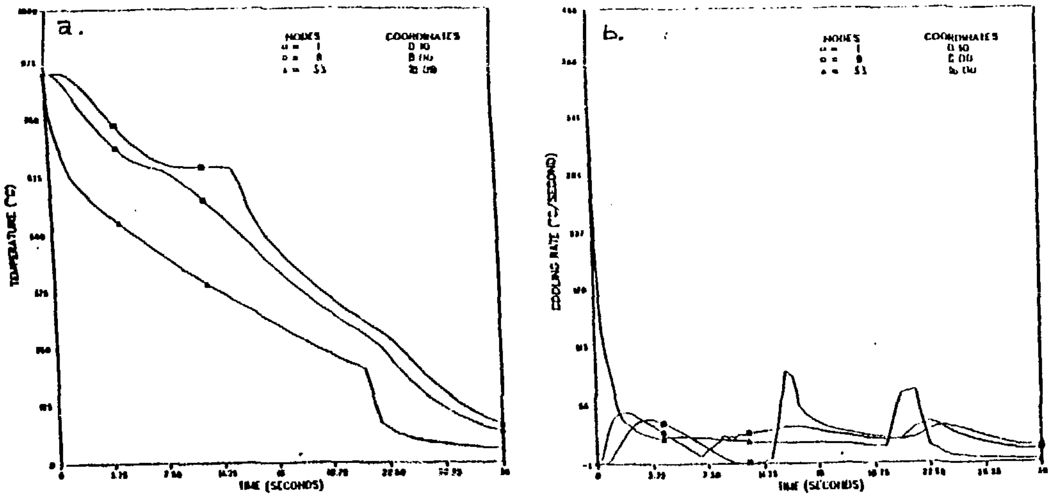


Fig. 5. Node temperatures (a) and cooling rates (b) as functions of time for 1-d model with phase transformations considered (Base Case).

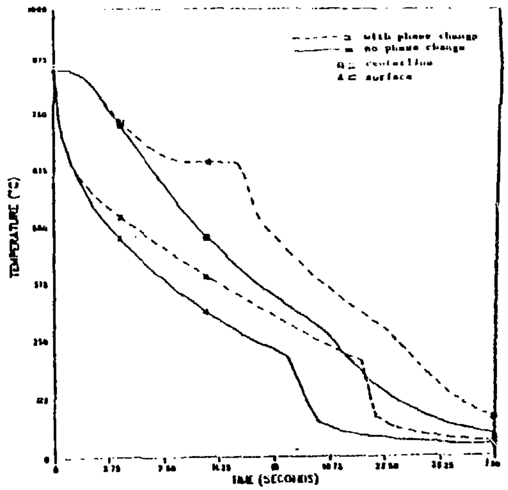


Fig. 6. Comparisons of surface and centerline temperatures as functions of time for 1-d model with and without heats of phase transformation (Case 2).

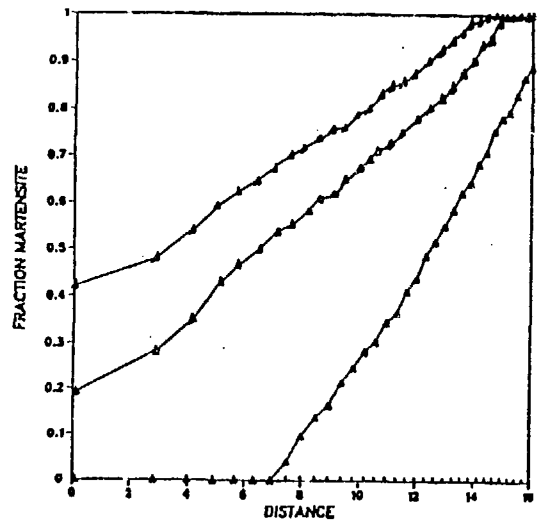
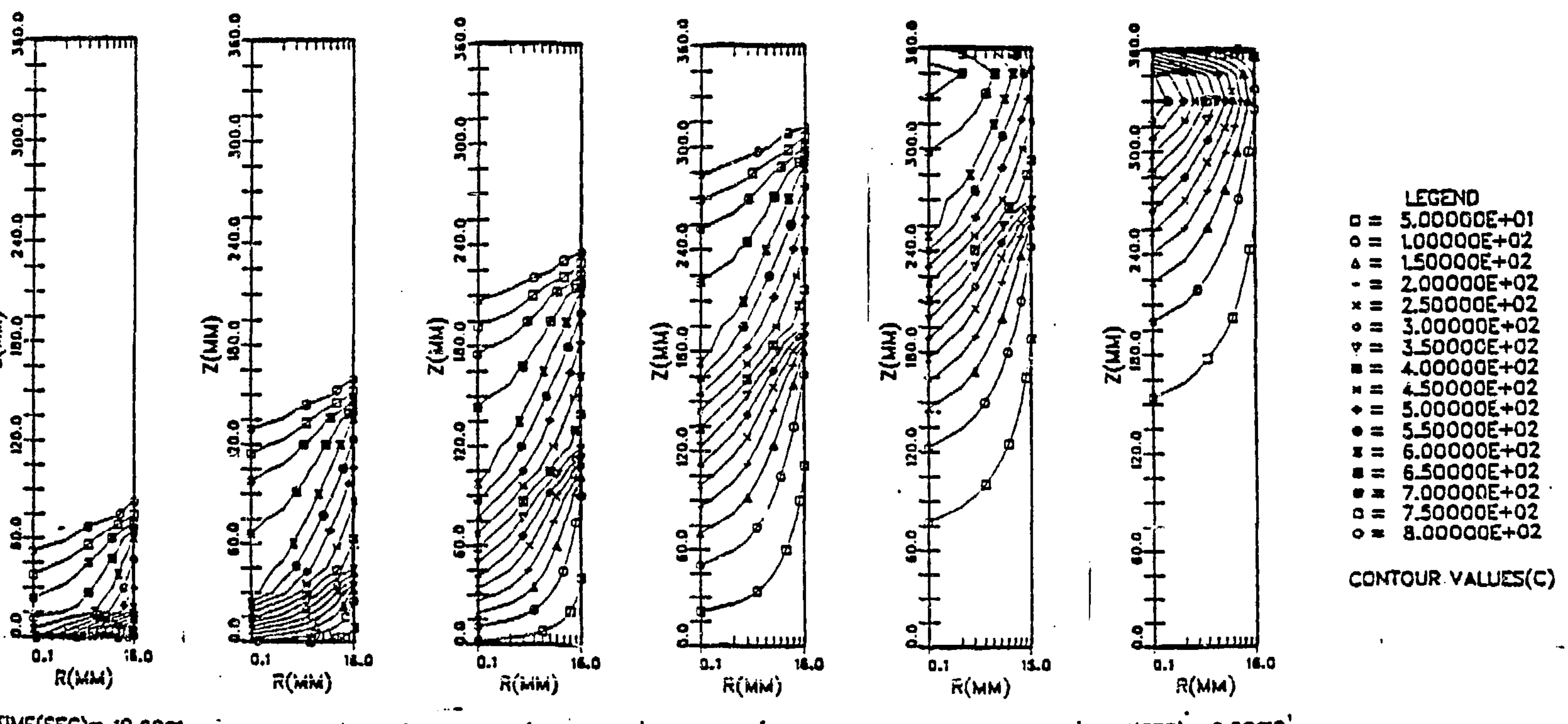


Fig. 7. Fraction of martinsite at the final time as a function of film heat transfer coefficient (Cases 7, 8, and 9).

a.



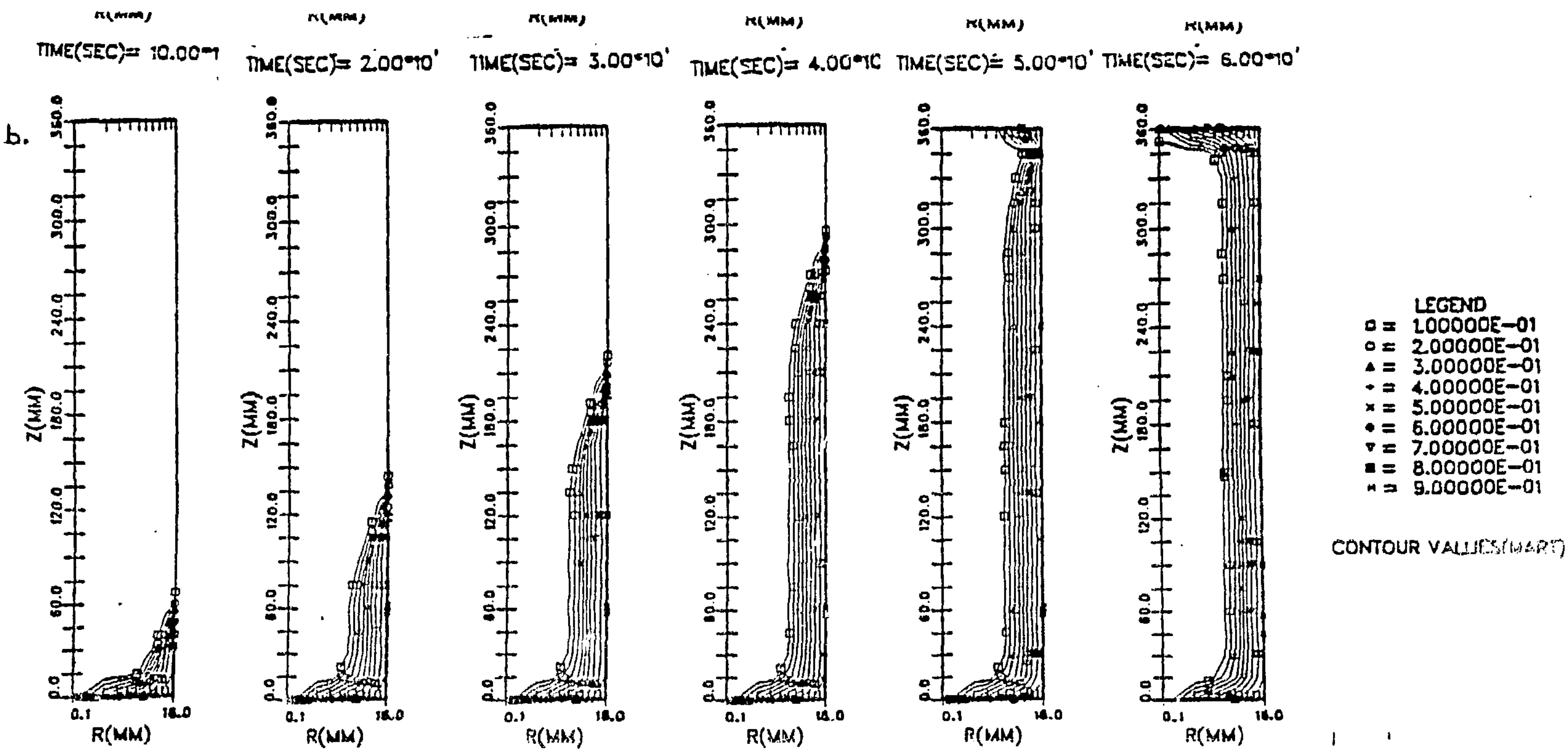


Fig. 8. Isotherms (a) and phase distributions (b) for three-dimensional (R, O, Z) immersion model at an immersion rate of 7.62 mm/sec at the plane O = n during a 1-min transient at 10-sec intervals (Case 18).

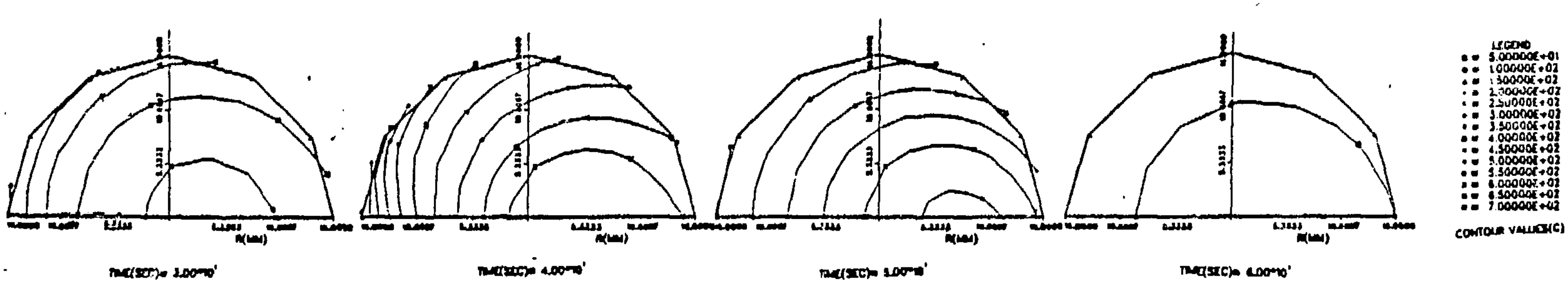


Fig. 9. Isotherms for three-dimensional (R, O, Z) model at plane Z = 180 mm during a 1-min transient at 10-sec intervals (Case 18).