

NASA Technical Memorandum 82898

A Search for Equilibrium States

(NASA-TM-82898) A SEARCH FOR EQUILIBRIUM STATES (NASA) 13 p HC A02/MF A01 CSCL 20M N83-20808

Unclas
G3/77 03121

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November 1982



A SEARCH FOR EQUILIBRIUM STATES

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ABSTRACT

An efficient search algorithm is described for the location of equilibrium states in a search set of states which differ from one another only by the choice of pure phases. The algorithm has three important characteristics: (1) it ignores states which have little prospect for being an improved approximation to the true equilibrium state; (2) it avoids states which lead to singular iteration equations; (3) it furnishes a search history which can provide clues to alternative search paths.

INTRODUCTION

The numerical computation of the composition and other properties of equilibrium states has been a continuing challenge for many years. As computers have become more powerful, people have attempted larger and more complex equilibrium calculations. These more complex problems naturally require greater amounts of computer time but, more importantly, they increase the probability that numerical obstacles to successful computations will arise. The potential impediments to success include inadequate initial estimates, convergence problems, difficulties with numerical significance and singular iteration equations. Difficulties such as these are often interrelated and more than one of them may arise to plague the unwary thermodynamicist in a particular problem. It is desirable, insofar as possible, to anticipate and avoid numerical problems to enhance the probability of success.

Let me briefly review the nature of equilibrium computations. An equilibrium state corresponds to an extremum in a function over some selected class of candidate states; for example, it might be regarded as a maximum in the entropy or a minimum in a free energy. Thus, in principle, the location of an equilibrium state can be regarded as a two step procedure: the calculation of the thermodynamic properties of all candidate states followed by a comparison of their entropies (or free energies). The greater the number of candidate states, the more difficult and time consuming the computational process and in short order it becomes quite impractical. The number of candidate states usually grows with the number of chemical species being considered. Hence an obvious way to limit the number of states is to restrict the number of species. But sometimes this may be undesirable because in so doing we might unwittingly eliminate the true equilibrium state from consideration.

*Presented, in part, at Calphad XI held at the Argonne National Laboratory, Argonne, Illinois, May 16-20, 1982.

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There is an alternative and generally more practical approach to the location of equilibrium states when there are many states to consider. One can begin at some initial state, explore for a state of higher entropy in the vicinity of this state and then step to an adjacent state of greater entropy. This permits one to ignore states of lower entropy and insures that each successful step will yield a state of higher entropy. The chief disadvantage with this method is that one cannot be certain that a particular starting point will produce a search path that will ultimately reach the true equilibrium state; it could just as easily terminate at a local entropy maximum or terminate prematurely because of numerical difficulties. Further, even if the path were to terminate at the true equilibrium state one might not recognize it as the global maximum. A partial resolution of this dilemma is provided by trying different starting points and comparing the entropies at the end points of the various paths. This is illustrated schematically in fig. 1 for two search paths. In this illustration the first path gives a better approximation to the equilibrium state than the second but neither has located the true equilibrium state.

Clearly the prospect of a successful search is enhanced by increasing the number of successful steps in the search path beginning at a given point. The number of steps can be increased by assiduously avoiding both states which are likely to create numerical problems and states which have little prospect for being the equilibrium state. I shall describe a search algorithm of this type for an important class of thermodynamic states; those obtained from a particular state by the addition of one or more pure phases in equilibrium. The algorithm has three beneficial characteristics: (1) it ignores states which have little prospect for yielding an improved approximation to the true equilibrium state; (2) it avoids states which lead to singular iteration equations; (3) it furnishes a search history which might provide clues to alternative search paths.

EQUILIBRIUM EQUATIONS

Before I can describe the search algorithm it is necessary to look at the equation which must be satisfied by an equilibrium state. The governing equations are a set of nonlinear algebraic equations (Zeleznik and Gordon, 1968; Zeleznik, 1981) obtained from a constrained extremum calculation.

$$\mu_j^\alpha + \sum_{i=1}^{\ell} \lambda_i v_{ij}^\alpha = 0 \quad j = 1, 2, \dots, m_\alpha; \alpha = 1, 2, \dots, p \quad (1)$$

$$\sum_{\alpha=1}^p \sum_{j=1}^{m_\alpha} v_{ij}^\alpha n_j^\alpha - q_i = 0 \quad i = 1, 2, \dots, \ell$$

In (1) the Greek index α is a phase index used to label each of the p phases, the Latin index j labels the m_α species within a phase and the Latin index i labels the ℓ composition constraint relations. The quantity μ_j^α is the chemical potential of species j in phase α and is a function of the phase composition. The λ_i are Lagrangian multipliers associated with

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each of the ℓ constraint relations. The doubly subscripted quantity v_{ij}^α is the set of stoichiometric coefficients representing the moles of component i (i^{th} independent species) in one mole of the j^{th} species in phase α . Finally n_j^α is the moles per unit mass of species j in phase α and q_i gives the moles of component i per unit mass. Suppose that the phases $p_0 \leq \alpha \leq p$ are pure phases. Then for $p_0 \leq \alpha \leq p$, $m_\alpha = 1$ and μ_1^α is just the chemical potential of the pure species μ_α^* which is independent of composition. Thus, in this case, (1) contains a subset of $(1 + p - p_0)$ linear equations which are independent of the composition variables.

$$\sum_{i=1}^{\ell} \lambda_i v_{i1}^\alpha = -\mu_\alpha^* \quad \alpha = p_0, p_0 + 1, \dots, p \quad (2)$$

Naturally any solution of (1) must provide a solution for (2). Conversely, if there is no solution of (2) there can be no solution for (1). Consequently, it is important to focus on those states for which we can be relatively certain that (2) will possess solutions as a means for extending the search path. Fortunately, the properties of linear systems of equations are well understood and this understanding can be applied advantageously to the particular set of equations shown in (2). The relevant ideas are most easily illustrated with a simple numerical example.

A system of linear equations such as (2) can be written most compactly in matrix notation as

$$Ax = c \quad (3)$$

where A is a rectangular matrix, c is a known column vector and x an unknown column vector. Linear equations are classified as either possessing solutions (consistent equations) or not possessing solutions (inconsistent equations). Consistent equations may possess one or many solutions. The matrix A and the vector c together determine whether the equations are consistent or inconsistent. For example let A be a 3×4 rectangular matrix.

$$A = \begin{pmatrix} 1 & 2 & 3 & 4 \\ 2 & 3 & 4 & 6 \\ 3 & 5 & 7 & 10 \end{pmatrix}$$

Then x , if it exists, is a four component vector $x' = (x_1, x_2, x_3, x_4)$ and c is a three component vector $c' = (c_1, c_2, c_3)$ where a prime denotes the transpose of a matrix or a vector. Suppose that $c' = \hat{c}' \equiv (0, 1, 1)$. Then it may be readily verified that $x' = \hat{x}' \equiv (1, 1, -1, 0)$ is a solution of (3) and hence, for this choice of A and c , the equations are consistent. As a matter of fact there are infinitely many solutions. To demonstrate this we only need to verify that the two vectors

$$x_1^0 = \begin{pmatrix} 1 \\ -2 \\ 1 \\ 0 \end{pmatrix} \quad x_2^0 = \begin{pmatrix} 0 \\ -2 \\ 0 \\ 1 \end{pmatrix}$$

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satisfy $Ax_1^0 = 0 = Ax_2^0$. Such vectors are said to be in the null space of A . Thus the vector $x = \hat{x} + \beta_1 x_1^0 + \beta_2 x_2^0$ is a solution for arbitrary numbers β_1 and β_2 . Actually this is the general solution to the problem. On the other hand suppose $c' = \tilde{c}' \equiv (1, 1, 1)$. Then it turns that (3) becomes an inconsistent set of equations because if a solution existed it would lead to an inconsistency. To see that this is true we observe that if $w' \equiv (1, 1, -1)$ then $w'A = 0$. But if a solution exists then

$$1 = (1 + 1 - 1) = w'c = w'Ax = 0'x = 0$$

which is clearly inconsistent. A vector such as w is said to be in the left null space of A or, equivalently, in the null space of A' . Thus we see that (3) is solvable for x if and only if c satisfies the condition

$$w'c = c_1 + c_2 - c_3 = 0$$

In general, $Ax = c$ is solvable if and only if $w'c = 0$ for each independent vector w in the left null space of A .

The ideas which were illustrated by example in the preceding paragraph can be established quite generally and are done so in texts on linear algebra. For our purposes the important feature is that each independent vector in the left null space of A imposes an independent constraint which must be satisfied by the vector c if $Ax = c$ is to be consistent. Each additional constraint reduces the likelihood that a given vector c will permit a solution. However, if there are no vectors in the left null space of A then a solution will always exist. If the relation $w'A = 0$ is written out in terms of the components then it can be recognized as implying that the rows of A are linearly dependent. Thus if the rows of A are linearly independent then there will be no vector in the left null space of A and no constraints imposed on c .

Suppose we now choose A to be the coefficient matrix of (2). Then its rows are

$$(v_{11}^\alpha, v_{21}^\alpha, \dots, v_{\alpha 1}^\alpha) \quad \alpha = p_0, p_0 + 1, \dots, p$$

and each row is composed of the stoichiometric coefficients for the species which forms one of the $1 + p - p_0$ pure phases. So long as the pure phase species are independent of each other there will be no restrictions imposed on the permissible values for the right side of (2). Thus, from the previous discussion, it seems desirable to confine a search path to states whose pure phases are independent species. This does not mean that dependent pure phases cannot coexist. For example, on any typical winter day one can find liquid water and ice coexisting and these are certainly dependent species since they have identical chemical formulas. It only means that a state containing both water phases is less likely to arise as an equilibrium state than one containing only one of them. The initial phases of a search for an equilibrium state should be confined to the more likely states. Those states containing dependent pure phases can be searched after all of the independent pure phase states have been explored. As a practical matter candidate states with dependent pure phases may have to be treated as special cases rather than as part of a general search procedure.

THE SEARCH ALGORITHM

We can now incorporate the ideas from the foregoing discussion in a general search algorithm over a set of thermodynamic states which differ from one another only in the choice of independent pure phases.

- Step 1. Specify an initial state with p independent pure phases.
- Step 2. Specify a search set of pure species.
- Step 3. Calculate properties of the state.
- Step 4. If any pure phases are negative remove all of them and go to Step 3.
- Step 5. For each species in the search set calculate

$$\phi \equiv \mu_j^* + \sum_{i=1}^k \lambda_i v_{ij} \quad \text{where } \mu_j^* \text{ is the chemical potential of the}$$

- Step 6. j^{th} species and v_{ij} is its set of stoichiometric coefficients. Select as the $(p+1)^{\text{th}}$ pure phase a species from the search set for which $\phi < 0$, ϕ is a minimum over the search set and such that the species is independent of the p phases already present.
- Step 7. If a $(p+1)^{\text{th}}$ pure phase is located go to Step 3, otherwise terminate the search.

The criterion $\phi < 0$ for pure phase inclusion, Step 6, is used in the current NASA equilibrium program and was also used by its ancestral programs (Gordon and McBride, 1976; Gordon and Zeleznik, 1963; Zeleznik and Gordon, 1962). The simultaneous removal, in Step 4, of all pure phases with negative moles differs from the procedure used in the NASA equilibrium programs where only one such phase is removed. The reason for this change is that it introduces an asymmetry in the search path which should help to minimize the occurrence of closed loop paths. Occasionally it may also accelerate progress toward the final state. The requirement for independent species in Step 6 has a twofold purpose. First, it restricts the search to independent pure phases which have the greatest prospect for yielding a solution. Second, it avoids the class of singular iteration equations associated with dependent pure phases.

AN APPLICATION OF THE SEARCH ALGORITHM

I shall illustrate the use of the search algorithm by applying it to a large, difficult problem that arose in a study of the corrosion of turbine blade materials by deposits from high temperature combustion gases (Santoro et al., 1981, 1980). The problem involves 19 chemical elements present in widely varying amounts as is shown in Table 1. Further, because many of the species of interest are present in trace amounts it was desirable to calculate accurately mole fractions as small as 10^{-20} . The large disparity in elemental amounts and the requirement for high accuracy in the mole fractions presented a severe challenge to the numerics. Even with double precision arithmetic (approximately 14 digits) the results are just above the numerical noise. Calculations were carried out at one atmosphere pressure and a temperature of 1000 K. The initial states were always composed of the gaseous phase plus var-

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ious pure phases. Nearly 500 different chemical species were considered in the equilibrium computations.

Table 2 shows the initial and final states for six different search paths, the number of steps in each path and the Gibbs free energy of the final state. Paths 3 and 4 terminated at the same state while paths 5 and 6 also terminated at identical states. The common terminus of paths 3 and 4 represents the best estimate of the equilibrium state as judged by the value of the Gibbs free energy. The paths themselves are schematically presented in Table 3 where "+" denotes an addition of a pure phase, "-" denotes a deletion of a pure phase and "±" represents a null step. A null step means that the algorithm would have added a pure phase but could not do so because it would have been linearly dependent on the pure phases already present. Paths 3 and 5 are not shown. Path 3 has no steps and path 5 has two steps, the removal of Al_2SiO_5 followed by the addition of CaAl_4O_7 .

Path 1 begins at an initial state containing only the gaseous solution and terminates in a tight three step loop without encountering any null steps. Table 4 shows the properties of states encountered by path 1. The initial state for path 2 is a modification of the final state for path 1. Neither Al_2TiO_5 nor $\text{Ca}_4\text{Ti}_3\text{O}_{10}$ were chosen for the initial state of path 2 because they were involved in the tight loop terminating path 1. The species Ti_4O_7 was replaced by TiO_2 and $\text{Al}_6\text{Si}_2\text{O}_{13}$ was replaced by Al_2O_3 and SiO_2 with which it is linearly dependent. Finally, Fe_3O_4 was replaced by Fe_2O_3 and the species CaSO_4 , Na_2SO_4 , PbSO_4 and ZnO were added. The ensuing search path contains two null steps; one involving Al_2SiO_5 to terminate the search and another involving Al_2TiO_5 at step 2. Although Al_2TiO_5 was involved in a null step, it was subsequently added to the system at step 8.

The initial states for paths 3 and 4 are derivatives of the final state for path 2. Since path 2 was completed with an Al_2SiO_5 null step it suggests that new paths be tried which include it as a pure phase. Thus, for path 3, $\text{Al}_6\text{Si}_2\text{O}_{13}$ was omitted from consideration in the search and was replaced by Al_2SiO_5 in the initial state. This search terminated with no additional steps being taken. For path 4, $\text{Al}_6\text{Si}_2\text{O}_{13}$ was again omitted from consideration and was replaced in the initial state by the combination of Al_2O_3 and SiO_2 .

Like paths 3 and 4, the initial states for 5 and 6 are closely related to the final state for 2. Rather than omitting $\text{Al}_6\text{Si}_2\text{O}_{13}$, it was Al_2O_3 which was excluded from the four species dependent set of Al_2O_3 , SiO_2 , Al_2SiO_5 and $\text{Al}_6\text{Si}_2\text{O}_{13}$. For path 5 the species $\text{Al}_6\text{Si}_2\text{O}_{13}$ was supplanted by Al_2SiO_5 in the initial state. For path 6 it was replaced by SiO_2 .

CONCLUSIONS

An algorithm has been described which offers considerable assistance in the determination of equilibrium states containing pure phases. Its utility increases with the complexity of the system being calculated.

ACKNOWLEDGEMENTS

I wish to acknowledge the assistance of Bonnie J. McBride in carrying out the calculations for this paper. I also wish to thank Fred Kohl for calling my attention to the difficult problem used in the example.

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TABLE 1. - DISTRIBUTION OF ELEMENTS IN THE SAMPLE PROBLEM.

	KG-ATOMS/KG
C	0.27960611E-02
H	0.40735049E-02
N	0.51352555E-01
O	0.14417322E-01
S	0.35989835E-05
Ar	0.30643626E-03
V	0.37750290E-09
Ca	0.95960942E-08
Mg	0.15824376E-07
Al	0.71273032E-07
Cl	0.54242432E-07
Na	0.83648392E-08
K	0.49185178E-08
Si	0.68471511E-08
Fe	0.34434377E-08
Zn	0.29413516E-08
Pb	0.92811611E-09
Ba	0.14003181E-08
Ti	0.40147322E-08

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TABLE 2. - PURE PHASE SPECIES IN INITIAL AND FINAL STATES FOR SEARCH PATHS

SEARCH PATH PURE PHASES	1		2		3		4		5		6	
	INITIAL	FINAL	INITIAL	FINAL	INITIAL	FINAL	INITIAL	FINAL	INITIAL	FINAL	INITIAL	FINAL
Al ₂ O ₃			X	X	X	X	X	X	0		0	
SiO ₂			X				X				X	
Al ₂ SiO ₅				X	X		X	X				
Al ₆ Si ₂ O ₁₃		X		X	0		0		X	X		X
CaAl ₄ O ₇		X								X		X
MgAl ₂ O ₄		X	X	X	X	X	X	X	X	X	X	X
TiO ₂			X				X				X	
Ti ₄ O ₇		X										
Al ₂ TiO ₅				X	X	X		X	X	X		X
Ca ₄ Ti ₃ O ₁₀		X										
Na ₂ V ₂ O ₆		X	X	X	X	X	X	X	X	X	X	X
BaSO ₄		X	X	X	X	X	X	X	X	X	X	X
CaSO ₄			X	X	X	X	X	X	X	X	X	X
K ₂ SO ₄				X	X	X		X	X	X		X
Na ₂ SO ₄			X	X	X	X	X	X	X	X	X	X
PbSO ₄			X	X	X	X	X	X	X	X	X	X
ZnO			X	X	X	X	X	X	X	X	X	X
Fe ₂ O ₃			X	X	X	X	X	X	X	X	X	X
Fe ₃ O ₄		X										
STEPS	15		17		0		20		2		8	
GIBBS ENERGY PER UNIT MASS	-2156.7272 6236		-2156.7275 3378		-2156.7275 3734				-2156.7274 7210			

NOTE: X PURE PHASE PRESENT
0 PURE PHASE OMITTED FROM CONSIDERATION IN THE

SEARCH

TABLE 3. - SEARCH PATH DETAILS

STEP	PATH 1	2	4	6
1	+Mg ₂ Al ₄ Si ₅ O ₁₈	+K ₂ SO ₄	+K ₂ SO ₄	+Al ₆ Si ₂ O ₁₃
2	+Ca ₄ Ti ₃ O ₁₀	±Al ₂ TiO ₅	±Al ₂ TiO ₅	-SiO ₂
3	+CaAl ₄ O ₇	+Na ₂ Al ₂ Si ₆ O ₁₆	+Na ₂ Al ₂ Si ₆ O ₁₆	+CaAl ₄ O ₇
4	-Ca ₄ Ti ₃ O ₁₀	-Al ₂ O ₃ -Na ₂ SO ₄ -SiO ₂	-Al ₂ O ₃ -Na ₂ SO ₄ -SiO ₂	+Al ₂ TiO ₅
5	+Al ₆ Si ₂ O ₁₃	+Al ₆ Si ₂ O ₁₃	+CaAl ₄ O ₇	-CaAl ₄ O ₇ -PbSO ₄ -TiO ₂
6	-Mg ₂ Al ₄ Si ₅ O ₁₈	-Na ₂ Al ₂ Si ₆ O ₁₆	+Al ₂ TiO ₅	+CaAl ₄ O ₇
7	+Ti ₄ O ₇	+CaAl ₄ O ₇	-CaAl ₄ O ₇ -PbSO ₄ -K ₂ SO ₄ -TiO ₂	+K ₂ SO ₄
8	+MgAl ₂ O ₄	+Al ₂ TiO ₅	+CaAl ₄ O ₇	+PbSO ₄
9	+Ca ₄ Ti ₃ O ₁₀	-CaAl ₄ O ₇ -PbSO ₄ -K ₂ SO ₄ -TiO ₂	+Al ₂ O ₃	
10	+Na ₂ V ₂ O ₆	+CaAl ₄ O ₇	-CaAl ₄ O ₇	
11	+Fe ₃ O ₄	+Na ₂ SO ₄	+Na ₂ SO ₄	
12	+BaSO ₄	+Al ₂ O ₃	+K ₂ SO ₄	
13	+Ca ₄ Ti ₃ O ₁₀	+K ₂ SO ₄	-Na ₂ Al ₂ Si ₆ O ₁₆ -Al ₂ O ₃	
14	+Al ₂ TiO ₅	-CaAl ₄ O ₇ -Na ₂ SO ₄	+PbSO ₄	
15	-Al ₂ TiO ₅ -Ca ₄ Ti ₃ O ₁₀	+Na ₂ SO ₄	+Al ₂ SiO ₅	
16		+PbSO ₄	+CaAl ₄ O ₇	
17		±Al ₂ SiO ₅	+Al ₂ O ₃	
18			-CaAl ₄ O ₇ -PbSO ₄ -K ₂ SO ₄	
19			+K ₂ SO ₄	
20			+PbSO ₄	

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1 Report No NASA TM-82898	2. Government Accession No	3 Recipient's Catalog No	
4. Title and Subtitle A SEARCH FOR EQUILIBRIUM STATES		5 Report Date November 1982	
		6 Performing Organization Code 505-32-32	
7. Author(s) Frank J. Zeleznik		8. Performing Organization Report No E-1281	
		10 Work Unit No	
9. Performing Organization Name and Address National Aeronautics and Space Administration Lewis Research Center Cleveland, Ohio 44135		11. Contract or Grant No	
		13. Type of Report and Period Covered Technical Memorandum	
12 Sponsoring Agency Name and Address National Aeronautics and Space Administration Washington, D.C. 20546		14. Sponsoring Agency Code	
		15. Supplementary Notes Presented, in part, at Calphad XI held at the Argonne National Laboratory, Argonne, Illinois, May 16-20, 1982.	
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17 Key Words (Suggested by Author(s)) Thermodynamics Equilibria Computation		18 Distribution Statement Unclassified - unlimited STAR Category 77	
19. Security Classif. (of this report) Unclassified	20. Security Classif. (of this page) Unclassified	21. No. of Pages	22. Price*

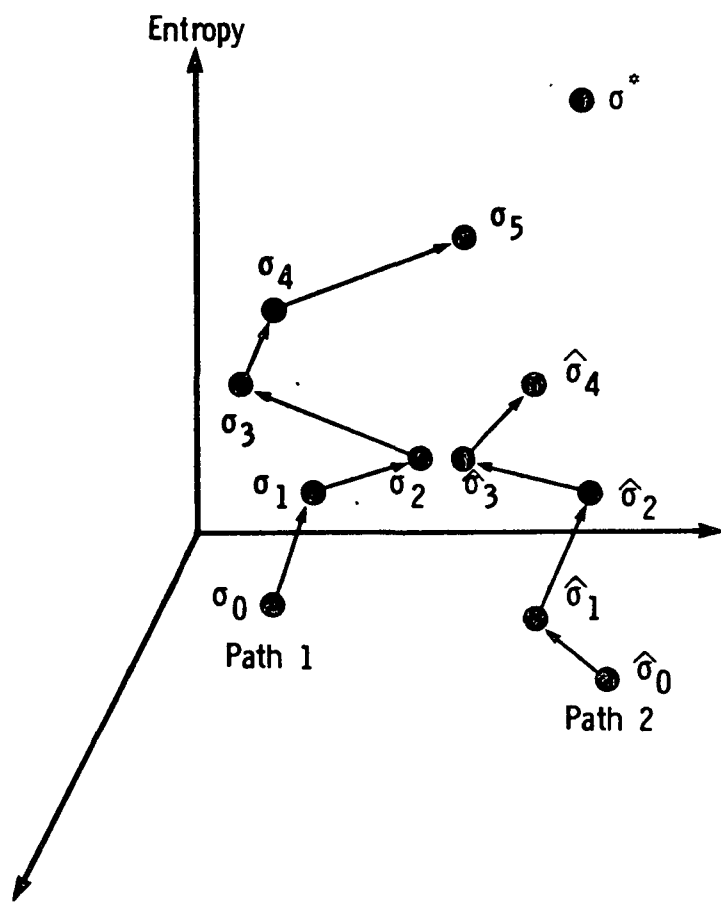


Figure 1. - Two search paths for the equilibrium state σ^* beginning with states σ_0 and $\hat{\sigma}_0$.

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