A NOTE ON THE IMPLICATIONS OF THERMODYNAMIC STABILITY IN THE INTERNAL VARIABLE THEORY OF INELASTIC SOLIDSt

J. B. MARTIN‡

Department of Civil Engineering, University of Cape Town, Rondebosch, Cape Town, South Africa

(Received 26 *December 1973)*

Abstract-Recently developed internal theories for inelastic solids are considered, and the consequences of thermodynamic stability requirements are explored. The results are expressed in a form which permits comparison with the stability postulates of Drucker and Ilyushin.

1. INTRODUCTION

In recent years considerable attention has been given to the study of the thermodynamic foundations of the mechanical behaviour of non-linear solids. These studies (see, for example Coleman and Gurtin [2], Valanis[1], Kestin and Rice [3], Rice [4], Lubliner[5], Nemat-Nasser [6] for recent examples) are based on the concepts of internal variables and, while the degree of generality and the details in various approaches may differ, the fundamental concepts are essentially identical in each case. It appears, however, that the consequences of the requirements of thermodynamic stability have not been fully explored. In this note we shall draw attention to some of these consequences.

In particular, we shall discuss the relation between the consequences of stability in the internal variable formulation and the quasi-thermodynamic postulates introduced in the early stages of the development of classical plasticity by Drucker[7,8] and Ilyushin [9]. For this purpose it will be sufficient that we formulate the thermodynamic description for solids which undergo small deformations, so that the distinction between density and stress in the deformed and undeformed configuration can be ignored. A generalisation of the major part of the work can be accomplished without difficulty.

2. THERMODYNAMIC FORMULATION

In setting down the thermodynamic description of the mechanical behaviour of an inelastic solid we shall follow the formulation of Kestin and Rice [3].

Equilibrium states of an infinitesimal element of an elastic solid satisfy the fundamental equation

$$
u = u(s, \epsilon_{ij}, \xi_{\alpha}) \tag{1}
$$

where u is the specific internal energy (i.e. the internal energy per unit mass), s is the specific

tThe research reported in this paper was supported by the Ollice of Naval Research under Contract NOOO 14-67-A-OI91-0003 with Brown University.

Wrofessor of Civil Engineering, University of Cape Town. (Formerly Professor of Engineering, Brown University.)

248 J. B. MARTIN

entropy, ϵ_{ij} is the strain tensor and ξ_{α} ($\alpha = 1, \ldots, n$) are independent internal variables whose precise nature will depend on the solid under discussion. We shall be concerned with processes in which the element passes through a continuous sequence of states which satisfy the fundamental equation (1). Because such processes in inelastic solids are normally approximated only under slow rates of loading, we shall for convenience refer to them as quasi-static processes.

At any instant during a quasi-static process, therefore,

$$
\begin{split} \dot{u} &= \frac{\partial u}{\partial s} \dot{s} + \frac{\partial u}{\partial \epsilon_{ij}} \dot{\epsilon}_{ij} + \frac{\partial u}{\partial \xi_{\alpha}} \dot{\xi}_{\alpha} \\ &= T\dot{s} + \rho^{-1} \sigma_{ij} \dot{\epsilon}_{ij} - P_{\alpha} \dot{\xi}_{\alpha}. \end{split} \tag{2}
$$

In this equation we identify the temperature T ,

$$
T = \frac{\partial u}{\partial s},\tag{3}
$$

and the stress tensor σ_{ij} ,

$$
\sigma_{ij} = \rho \frac{\partial u}{\partial \epsilon_{ij}},\tag{4}
$$

where ρ is the density. Further, we define P_{α} as being equal and opposite to the internal forces $\partial f/\partial \xi_{\alpha}$ which are conjugate to the internal variables ξ_{α} ,

$$
P_{\alpha} = -\frac{\partial u}{\partial \xi_{\alpha}}.\tag{5}
$$

The reversible work along a quasi-static path is

$$
\dot{W}_0 = \rho^{-1} \sigma_{ij} \epsilon_{ij} - P_\alpha \dot{\xi}_\alpha, \tag{6}
$$

while the rate at which external forces do work on an element of unit mass is

$$
\dot{W} = \rho^{-1} \sigma_{ij} \dot{\epsilon}_{ij}.
$$
 (7)

It follows then that the rate of entropy production per unit mass due to energy dissipation ζ is

$$
\dot{\zeta} = \frac{\dot{W} - \dot{W}_0}{T} = \frac{P_{\alpha} \dot{\xi}_{\alpha}}{T}.
$$
\n(8)

The Second Law requires that $\dot{\zeta} > 0$, and hence, since $T > 0$, at any instant we must have

$$
P_{\alpha}\dot{\xi}_{\alpha}>0.\tag{9}
$$

In order to complete our description of the inelastic solid we must introduce a phenomenological relation between the internal forces P_{α} and the rate of change of the internal variables $\dot{\xi}_{\alpha}$. In the simplest case we may put

$$
\dot{\xi}_{\alpha} = \dot{\xi}_{\alpha}(P_{\beta}), \qquad P_{\alpha} = P_{\alpha}(s, \epsilon_{i}, \xi_{\alpha}). \tag{10}
$$

This equation will be referred to as the rate equation, and is subject only to the restriction that inequality (9) must be satisfied.

Elimination of the internal variables ξ_{α} from equations (3) and (4) by means of equations (5) and (10) will provide what are usually referred to as the constitutive equationsfor the inelastic solid. We shall not concern ourselves with the particular forms that these equations take except to note that the constitutive equations for viscoelasticity, creep and plasticity can each be recovered by a suitable choice of the fundamental equation (1) and the rate equations (10). It is important to note, however, that a history of deformation which satisfies the constitutive relations is a quasi-static process according to our definition.

3. STABILITY CONSEQUENCES

In order to investigate the stability consequences of interest in this note we construct a conceptual isolated system which consists of a homogeneous element of unit mass of the non-linear solid, a reversible heat source and a reversible work source. The reversible heat source is in diathermal contact with the solid, and is at temperature T^o . Its internal energy u_H is given by

$$
u_H = T^{\circ} s_H, \tag{11}
$$

where s_H is the entropy of the heat source. The reversible work source is mechanically coupled to the non-linear solid so that it applies constant conservative forces $\rho^{-1} \sigma_{ij}^{\circ}$ to the element. Its internal energy u_w is given by

$$
u_w = -\rho^{-1} \sigma_{ij}^{\circ} \epsilon_{ij} \tag{12}
$$

to within an arbitrary constant which we may neglect. The total energy of the isolated system is then given by

$$
U = u + uH + uW
$$

= $u(s, \epsilon_{ij}, \xi_{\alpha}) + T^{\circ} s_H - \rho^{-1} \sigma^{\circ}_{ij} \epsilon_{ij}.$ (13)

The energy minimum principle states that the unconstrained equilibrium state of the isolated system is given by the values of the thermodynamic parameters which minimise U subject to the constraint that the total entropy of the system *S,*

$$
S = s + s_{\rm H},\tag{14}
$$

is constant. Taking variations about the unconstrained equilibrium state of the system, and noting that in view of equation (14) $\delta s_H = - \delta s$, we find that

$$
\delta U = \left(\frac{\partial u}{\partial s} - T^{\circ}\right) \delta s + \left(\frac{\partial u}{\partial \epsilon_{ij}} - \rho^{-1} \sigma^{\circ}_{ij}\right) \delta \epsilon_{ij} + \frac{\partial u}{\partial \xi_{\alpha}} \delta \xi_{\alpha}
$$

= $(T - T^{\circ}) \delta s + \rho^{-1} (\sigma_{ij} - \sigma^{\circ}_{ij}) \delta \epsilon_{ij} - P_{\alpha} \delta \xi_{\alpha}.$ (15)

Since arbitrary variations in s, ϵ_{ii} , ξ_{α} are possible, necessary and sufficient conditions for $\delta U = 0$ are that

$$
T = T^{\circ}, \qquad \sigma_{ij} = \sigma^{\circ}_{ij}, \qquad P_{\alpha} = 0. \tag{16}
$$

This equation defines the *unconstrained equilibrium states* of the system. From equations (3), (4), (5) and (16) we may determine the values of the thermodynamic parameters s° , ϵ_{ij}° , ξ_{α}° which characterise the unconstrained equilibrium state (or states) of the conceptual system. Note that this solution implies that ξ_{α} is constant and hence $\dot{\xi}_{\alpha} = 0$ in the equilibrium state. This in turn implies that the rate equation (10) must be such that $\dot{\xi}_\alpha = 0$ when $P_\alpha = 0$.

Using equation (14), U may be written as

$$
U = u(s, \epsilon_{ij}, \xi_{\alpha}) - T^{\circ} s - \rho^{-1} \sigma_{ij}^{\circ} \epsilon_{ij} + T^{\circ} S. \tag{17}
$$

Noting that the Helmholtz free energy of the unit mass of non-linear solid is

$$
f(T,\epsilon_{ij},\xi_{\alpha})=u-Ts,
$$
\n(18)

it is convenient to define

$$
U_p = f(T^{\circ}, \epsilon_{ij}, \xi_{\alpha}) - \rho^{-1} \sigma^{\circ}_{ij} \epsilon_{ij}.
$$
 (19)

The energies *U* and *Up* differ only by a constant. The energy minimum principle now states that *Up* should be a minimum in the unconstrained equilibrium state subject to the constraint that $T = T^o$. Since for a quasi-static process

$$
\dot{f} = \frac{\partial f}{\partial T} \dot{T} + \frac{\partial f}{\partial \epsilon_{ij}} \dot{\xi}_{ij} + \frac{\partial f}{\partial \xi_{\alpha}} \dot{\xi}_{\alpha} \n= -s \dot{T} + \rho^{-1} \sigma_{ij} \dot{\epsilon}_{ij} - P_{\alpha} \dot{\xi}_{\alpha},
$$
\n(20)

this alternative form of the minimum principle provides the same unconstrained equilibrium state that is given equation (16).

Now consider a state adjacent to the equilibrium state and characterised by T° , $\epsilon_{ij}^{(1)}$, $\xi_{\alpha}^{(1)}$. The energy minimum principle requires that

$$
U^p(T^{\circ}, \epsilon_{ij}^{(1)}, \xi_{\alpha}^{(1)}) > U^p(T^{\circ}, \epsilon_{ij}^{\circ}, \xi_{\alpha}^{\circ}). \tag{21}
$$

Substituting from equation (19) and rearranging, this becomes

$$
f(T^{\circ}, \epsilon_{ij}^{(1)} \xi_{\alpha}^{(1)}) - f(T^{\circ}, \epsilon_{ij}^{\circ}, \xi_{\alpha}^{\circ}) > \rho^{-1} \sigma_{ij}^{\circ} (\epsilon_{ij}^{(1)} - \epsilon_{ij}^{\circ}). \tag{22}
$$

Since our isolated system is a conceptual convenience, and since T° , σ_{ij}° can be arbitrarily chosen, this inequality is in fact a restriction on the form of the Helmholtz free energy of the non-linear solid. The restriction can be interpreted in two ways.

First, remembering that $\partial f/\partial \xi_{\alpha} = 0$ for $\xi_{\alpha} = \xi_{\alpha}^{\circ}$, inequality (22) can be regarded as a statement which requires some degree of convexity of the function f . Equation (22) can be written as

$$
f(T^{\circ}, \epsilon_{ij}^{(1)}, \xi_{\alpha}^{(1)}) - f(T^{\circ}, \epsilon_{ij}^{\circ}, \xi_{\alpha}^{\circ}) > (\epsilon_{ij}^{(1)} - \epsilon_{ij}^{\circ}) \frac{\partial f}{\partial \epsilon_{ij}}_{\epsilon_{ij} = \epsilon_{ij}^{\circ}} + (\xi_{\alpha}^{(1)} - \xi_{\alpha}^{\circ}) \frac{\partial f}{\partial \xi_{\alpha}}_{\epsilon_{\alpha} = \epsilon_{\alpha}^{\circ}}.\tag{23}
$$

The precise nature of the requirementsimposed by equation (23) is somewhat complex, and there is probably little to be gained in enumerating them at this degree of generality. It can be noted, however, that a sufficient condition that equation (23) should hold is that $f(T^{\circ}, \epsilon_{ij}, \xi_{\alpha})$ should be strictly convex for T^o arbitrary but constant. This would also ensure that there exists a unique euqilibrium state for the system.

Alternatively, on noting that

$$
f(T^{\circ}, \epsilon_{ij}^{(1)}, \xi_{\alpha}^{(1)}) - f(T^{\circ}, \epsilon_{ij}^{e}, \xi_{\alpha}^{e})
$$

=
$$
\int_{\epsilon_{ij}^{-\circ}}^{\epsilon_{ij}^{(1)}} \frac{\partial f}{\partial \epsilon_{ij}} d\epsilon_{ij} + \int_{\epsilon_{\alpha}^{-\circ}}^{\epsilon_{\alpha}^{(1)}} \frac{\partial f}{\partial \xi_{\alpha}} d\xi_{\alpha}
$$

=
$$
\int_{\epsilon_{ij}^{-\circ}}^{\epsilon_{ij}^{(1)}} \rho^{-1} \sigma_{ij} d\epsilon_{ij} - \int_{\epsilon_{\alpha}^{-\circ}}^{\epsilon_{\alpha}^{(1)}} P_{\alpha} d\xi_{\alpha}, \qquad (24)
$$

equation (22) can be written as

$$
\int_{\epsilon_{ij}^{\circ}}^{\epsilon_{ij}^{(1)}} \rho^{-1}(\sigma_{ij} - \sigma_{ij}^{\circ}) d\epsilon_{ij} > \int_{\xi_{\alpha}^{\circ}}^{\xi_{\alpha}^{(1)}} P_{\alpha} d\xi_{\alpha}, T = T^{\circ} = \text{constant.}
$$
 (25)

This inequality holds for any quasi-static process between the state T° , ϵ_{ii}° , ξ_{α}° and the adjacent state T° , $\epsilon_{ii}^{(1)}$, $\xi_{\alpha}^{(1)}$.

Again, since the system we have created is merely a conceptual convenience, with T° , σ_{ij}° arbitrarily chosen, this result has relevance for an arbitrary element of non-linear material. We may define *relaxed states* of the material characterised by thermodynamic variables T° , ϵ_{ii}° , $\epsilon_{\alpha}^{\circ}$. These states are identified by the condition that $P_{\alpha} = -\frac{\partial f}{\partial \xi_{\alpha}} = 0$. The entropy s^o and the stress tensor σ_{ij}° are then obtained by the relations $s^{\circ} = -\frac{\partial f}{\partial T}$, $\sigma_{ij}^{\circ} = \rho^{-1} \frac{\partial f}{\partial \epsilon_{ij}}$. These relaxed states coincide exactly with the unconstrained equilibrium states of the conceptual system insofar as the material element is concerned.

Now suppose the element is deformed isothermally in a process in which the constitutive equations are satisfied from a relaxed state T° , ϵ_{ij}° , ξ_{α}° to a neighbouring state T° , $\epsilon_{ij}^{(1)}$, $\xi_{\alpha}^{(1)}$. During this actual process the rate equations will be satisfied; nevertheless the process is a quasi-static process (according to our definition) since the internal energy of the element is at each instant during the process given by the fundamental equation (1). Hence inequality (25) applies. Furthermore, because the rate equations are satisfied, from inequality (9),

$$
\int_{\xi_{\alpha}}^{\xi_{\alpha}^{(1)}} P_{\alpha} d\xi_{\alpha} = \int_{\xi_{\alpha}}^{\xi_{\alpha}^{(1)}} P_{\alpha} \dot{\xi}_{\alpha} dt > 0.
$$

It follows then that for any element deformed isothermally from a relaxed state to a neighbouring state, in a process which satisfies the constitutive relations,

$$
\int_{\epsilon_{ij}^{\circ}}^{\epsilon_{ij}^{(1)}} (\sigma_{ij} - \sigma_{ij}^{\circ}) d\epsilon_{ij} > 0.
$$
 (26)

4. PLASTIC MATERIALS

Models for plastic and visco-plastic materials may be characterised by a discontinuity in the rate equation in that P_{α} need not be identically zero when $\dot{\xi}_{\alpha} = 0$. It is convenient to define a function $\psi(P_\alpha)$, and to incorporate into the rate equation (10) the requirement that $\dot{\xi}_\alpha = 0$ when $\psi(P_{\alpha})$ < 0. This will lead to the phenomenon of yielding in the constitutive equations. Behaviour

of this kind generally arises as a deliberate idealisation of the actual rate equations; nevertheless it is a useful idealisation under certain conditions and it is important to consider its implications within the thermodynamic framework.

Returning to the conceptual system described in previous sections, the basic results remain unchanged. However, we must now admit the possibility that the system cannot achieve an unconstrained equilibrium state in a process which begins at a non-equilibrium state and proceeds by the relaxation of constraints. This occurs simply because internal relaxation ceases when $\psi(P_{\alpha})$ < 0 and $\dot{\xi}_{\alpha}$ = 0. The system will reach a *constrained* equilibrium state in which, say,

$$
T = T^{\circ}, \qquad \sigma_{ij} = \sigma_{ij}^{\circ}, \qquad \epsilon_{ij} = \epsilon_{ij}^*, \qquad \xi_{\alpha} = \xi_{\alpha}^*, \qquad P_{\alpha} = P_{\alpha}^* \neq 0, \qquad \psi(P_{\alpha}^*) < 0. \tag{27}
$$

This constrained equilibrium state will depend on the process by which it is achieved even when the Helmholtz free energy f is strictly convex.

We ask whether it is still possible to make statements similar to that of inequality (25) for variations about a constrained equilibrium state. Let us compare the constrained equilibrium state T° , ϵ_{i}^{*} , ξ_{α}^{*} with an adjacent state T° , $\epsilon_{ij}^{(1)}$, $\xi_{\alpha}^{(1)}$. The energy minimum principle is still applicable in the constrained equilibrium state in the sense that the *unconstrained* parameters take on values which minimise U_p for $T = T^{\circ}$. Thus

$$
U_p(T^{\circ}, \epsilon_{ij}^{(1)}, \xi_{\alpha}^*) > U_p(T^{\circ}, \epsilon_{ij}^*, \xi_{\alpha}^*)
$$
\n
$$
(28)
$$

Let us now suppose that $f(T^{\circ}, \epsilon_{ii}, \xi_{\alpha})$ is strictly convex. It follows then that

$$
f(T^{\circ}, \epsilon_{ij}^{(1)}, \xi_{\alpha}^{(1)}) - f(T^{\circ}, \epsilon_{ij}^{(1)}, \xi_{\alpha}^{*}) > (\xi_{\alpha}^{(1)} - \xi_{\alpha}^{*}) \frac{\partial f}{\partial \xi_{\alpha}}_{T^{\circ}, \epsilon_{ij}^{(1)}, \epsilon_{\alpha}^{*}} = -(\xi_{\alpha}^{(1)} - \xi_{\alpha}^{*}) P_{\alpha}^{*}.
$$
 (29)

If we now add inequalities (28) and (29), we find that

$$
f(T^{\circ}, \epsilon_{ij}^{(1)}, \xi_{\alpha}^{(1)}) - f(T^{\circ}, \epsilon_{ij}^{*}, \xi_{\alpha}^{*}) - \rho^{-1} \sigma_{ij}^{\circ} (\epsilon_{ij}^{(1)} - \epsilon_{ij}^{*}) > -(\xi_{\alpha}^{(1)} - \xi_{\alpha}^{*}) P_{\alpha}^{*}.
$$
 (30)

Alternatively, this may be written as

$$
\int_{\epsilon_{ij}^*}^{\epsilon_{ij}^{(1)}} \rho^{-1}(\sigma_{ij} - \sigma_{ij}^{\circ}) d\epsilon_{ij} > \int_{\xi_{\alpha}^*}^{\xi_{\alpha}^{(1)}} (P_{\alpha} - P_{\alpha}^*) d\xi_{\alpha}, T = T^{\circ} = \text{constant.}
$$
 (31)

This inequality will hold for any quasi-static process between the state T° , ϵ_{i}^{*} , ξ_{α}^{*} and the state $T^{\circ}, \epsilon_{ij}^{(1)}, \epsilon_{\alpha}^{(1)}$.

As before, we may now consider an arbitrary element of the material. Let us define a *relaxed plastic state* as any state T^* , ϵ^* , ξ^* for which $\psi(P^*_{\alpha}) < 0$, $P^*_{\alpha} = -\frac{\partial f}{\partial \xi_{\alpha}}$, $s^* = -\frac{\partial f}{\partial T}$, σ^* , ρ^{-1} *af/aEij.*

Suppose that the element is deformed isothermally in a process in which the constitutive equations are satisfied from a relaxed plastic state $T^*, \epsilon_{ij}^*, \xi_{\alpha}^*$ to a neighbouring state $T^*, \epsilon_{ij}^{(1)}, \xi_{\alpha}^{(1)}$. Inequality (31) applies to this actual process.

However, inequality (9) is not sufficient to ensure that

$$
\int_{\xi_{\alpha}^{(1)}}^{\xi_{\alpha}^{(1)}} (P_{\alpha} - P_{\alpha}^{*}) d\xi_{\alpha} = \int_{\xi_{\alpha}^{*}}^{\xi_{\alpha}^{(1)}} (P_{\alpha} - P_{\alpha}^{*}) \dot{\xi}_{\alpha} dt > 0, \qquad (32)
$$

and hence that

$$
\int_{\epsilon_{ij}^*}^{\epsilon_{ij}^{(1)}} (\sigma_{ij} - \sigma_{ij}^*) d\epsilon_{ij} > 0.
$$
 (33)

It is evident that a *sufficient condition* for inequality (32), and hence inequality (33), is

$$
(P_{\alpha} - P_{\alpha}^*)\dot{\xi}_{\alpha} > 0,\tag{34}
$$

for P_α , $\dot{\xi}_\alpha$ associated through the rate equation and $\psi(P^*_\alpha)$ < 0. Note further that if f is a strictly convex function, as assumed, inequality (33) will hold for all choices of the final state and not only for states neighbouring the initial state.

5. CONCLUSIONS

The quasi-thermodynamic postulates for time-independent plastic materials introduced by Drucker[7] and Ilyushin [9] apply to processes in which an arbitrary material element passes through a continuous sequence of relaxed plastic states, and hence may be compared with inequality (33). It is evident that the net work,

$$
\int_{\epsilon_{ij}^*}^{\epsilon_{ij}^{(1)}} (\sigma_{ij} - \sigma_{ij}^*) d\epsilon_{ij},
$$

around a cycle in stress (Drucker) or a cycle in strain (Ilyushin) for either small or unlimited changes in state is not required to be positive by thermodynamic stability considerations. This can be ensured by the addition of an additional assumption, such as inequality (34); it is equally evident that inequality (34) is conceptually identical to Hill's principle of maximum plastic work[lO].

The more general result, given in inequality (26), applies to arbitrary time-dependent materials. This result does not support Drucker's attempted extension[8] of his postulate for time- independent materials to time-dependent materials.

*Acknowledgements-The*writer is indebted to ProfessorsJ. Kestin and J. R. Rice for valuable discussion and advice during the preparation of this paper.

REFERENCES

- 1. K. C. Valanis, Thermodynamics of large viscoelastic deformations, J. *Math. Phys.45,* 197-212 (1966); and, Unified theory of thermomechanical behaviour of viscoelastic materials, *Mechanical Behaviour of Materials under Dynamic Loads,* edited by U. S. Lindholm, p 343. Springer (1968).
- 2. B. D. Coleman and M. E. Gurtin, Thermodynamics with internal state variables, J. *Chem. Phys.* 47, 597 (1967).
- 3. J. Kestin and J. R. Rice, Paradoxes in the application of thermodynamics to strained solids, *A Critical Review of Thermodynamics,* edited by E. B. Stuart, B. Gal-Or and A. 1. Brainard, p. 275. Mono Book Corp. (1970).
- 4. J. R. Rice, On structure of stress-strain relations for time-dependent plastic deformation in metals, 1. *appl. Mech.* 37, 728 (1970).
- 5. 1. Lubliner, On the thermodynamic foundations of non-linearsolids, *Int.* 1. *Non-Linear Mechanics* 7,237-254 (1972).
- 6. S. Nemat-Nasser, On nonequilibrium thermodynamics of continua, Lecture delivered at the ASME Winter Annual Meeting, Nov. 26-30 (1972).
- 7. D. C. Drucker, A more fundamental approach to plastic stress-strain relations, *Proc. 1st U.S. Nat. Congr. Appl. Mech.,* p 487, ASME (1951).
- 8. D. C. Drucker, On the postulate of stability of material in the mechanics of continua, J. Mécanique 3, 235 (1964).
- 9. A. A. Ilyushin, On the postulate of plasticity, Prik. Mat. Mekh. 25, 503 (1961).
- 10. R. Hill, Avariational principle of maximum plastic work in classical plasticity, *Quart.* 1. *Mech. appl. Math.* t, 18 (1948).