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# A comprehensive review of rational irreversible thermodynamics

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ABSTRACT

This research paper presents a critical study of the ‘Rational irreversible thermodynamics’ framework on evaluating its theoretical foundations and practical applications. A comprehensive review of existing framework and theoretical analysis has been carried out. We identify the principles, assumptions, and limitations inherent in rational irreversible thermodynamics. Our findings highlight discrepancies between theoretical predictions and experimental observations, particularly in highly non-equilibrium systems. The critical evaluation presented in this study elucidates the challenges and opportunities in rational irreversible thermodynamics, leading the way for future research.

## 1. Introduction

Rational irreversible thermodynamics, also known as non-equilibrium thermodynamics, deals with systems that are not in thermodynamic equilibrium and involve processes with irreversible effects. This formalism was extended by Coleman (1964) [1], Truesdell (1984) [2], and Noll (1974) [3]. This theory is different from classical irreversible thermodynamics. This is a mathematical theory which is macroscopic and phenological. The principal aim of rational irreversible thermodynamics is to establish a method for deriving constitutive equations. These constitutive equations are analysed using the second law of thermodynamics, based on representation formulas and entropy production for a comprehensive description of physical processes [4, 5]. Irreversible thermodynamics is based on phenomenological laws but RIT follows a deductive approach, ensuring consistency with fundamental thermodynamic principles such as entropy production and material frame-indifference

Rational Irreversible Thermodynamics provides a comprehensive framework for analyzing and understanding non-equilibrium processes. The method of rational thermodynamics resembles to that of mechanics.

## 2. A Comprehensive study of Rational Irreversible Thermodynamics

RIT incorporates several key concepts and assumptions to model non-equilibrium systems effectively [6 , 7].

### 2.1. Conservation Laws

Rational irreversible thermodynamics starts with the conservation laws, which includes conservation of mass, energy, and momentum. These laws provide the foundation for understanding the behavior of systems undergoing non-equilibrium processes. However, there are two slight differences which are necessary. The first is the introduction of a specific rate of energy supply r in the balance of internal energy, which in local form is written as

$$\rho u = -\nabla \cdot q - P:\nabla v + \rho r \tag{1}$$

r is generally referred to as the power supplied or lost by radiation ρ is the mass density, e is the specific internal energy, q is the heat flux vector and ρr represents the energy

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supply per unit volume.

$$\frac{d}{dt}\int_{v} \rho e dv + \int_{s} q . ds = \int_{v} (\rho r + \text{other energy sources}) dv \tag{2}$$

### 2.2. Distinction Between Internal Energy and Other Forms of Energy

The total energy equation usually includes kinetic and potential energy terms. However, in RIT, the internal energy balance is precisely isolated from these contributions to highlight irreversible effects such as dissipation, heat flux, and chemical reactions.

### 2.3. Local Equilibrium Assumption

In classical thermodynamics, systems are typically assumed to be in global equilibrium, meaning the system is uniform, and thermodynamic variables like temperature and pressure are constant throughout.

In RIT, the assumption of local equilibrium is introduced. This means that locally (within small regions of the system), the system is assumed to be in thermodynamic equilibrium, even though the system as a whole may be far from equilibrium. This allows RIT to apply thermodynamic laws at a local level, making it possible to deal with gradients in temperature, pressure, chemical potential and other variables that drive irreversible processes like heat conduction, diffusion and chemical reactions [6, 7].

### 2.4. Linear response assumption

Rational irreversible thermodynamics often assumes that systems respond linearly to small perturbations from equilibrium conditions. This assumption reveals that the response of a system to a small change in its external conditions (e.g., temperature, pressure, or chemical potential) is linearly proportional to the magnitude of that change. The linear response assumption is a powerful tool for analyzing systems near equilibrium, its applicability should be carefully assessed based on the specific conditions of the system under consideration [8-10].

### 2.5. Homogeneous system assumption

The homogeneous system assumption is particularly useful because it allows thermodynamic properties such as temperature, pressure, density, and composition to be described by single-valued functions, simplifying the mathematical formulation of thermodynamic equations and relations. It forms the basis for many thermodynamic models and theories, especially in the context of continuous media. Rational irreversible thermodynamics typically assumes that systems are homogeneous and well-mixed, which means that properties do not vary spatially within the system [10, 11].

### 2.6. Neglect of fluctuations

In Rational Irreversible Thermodynamics (RIT), the neglect of fluctuations refers to the assumption that macroscopic quantities such as temperature, pressure, and concentration are smooth and continuous, without considering the random, microscopic fluctuations that occur naturally in any system. Rational irreversible thermodynamics often neglects fluctuations in thermodynamic quantities, assuming that they are small compared to average values. RIT provides a phenomenological framework that applies to macroscopic systems where microscopic fluctuations average out and are not considered essential to understanding the overall process [13, 14].

### 2.7. Axiom of Memory

This is a fundamental concept in rational thermodynamics that acknowledges the influence of the system's history on its current state and behavior. It recognizes that the thermodynamic state of a system at any given time is not solely determined by its current conditions but also depends on its prior interactions and experiences. This means that the fluxes (like heat, mass, or momentum transfer) at any given moment depend not only on the current thermodynamic forces (like temperature or concentration gradients) but also on the system's previous states [14, 15].

### 2.8. Axiom of Equipresence

Mathematically, the Axiom of Equipresence is expressed by stating that two systems are in the same thermodynamic state if and only if all of their extensive variables are equal. This equivalence relation provides the basis for defining thermodynamic equilibrium and for characterizing the evolution of systems undergoing non-equilibrium processes.

This axiom states that if a variable is present in one constitutive relation, then there is no reason why it should not be present in all the other constitutive equations, until it is proved otherwise. The condition for the presence or absence of an independent variable is essentially determined by the Clausius–Duhem’s inequality. It should be realized that there is no physical justification to such an axiom, which is merely a mathematical convenience in the determination of constitutive relations [14, 15].

### 2.9. Extended Second Law

In classical thermodynamics, the second law is often framed in terms of entropy change for systems in or moving toward equilibrium. However, many real-world processes are irreversible (e.g., heat flow, friction, chemical reactions). Truesdell extended the classical formulations of the second law to non-equilibrium situations, providing a more comprehensive understanding of irreversible processes. He formulated the second law in terms of inequalities that apply to irreversible processes, providing a framework for analyzing systems far from equilibrium [14, 15].

This makes the theory more applicable to real-world processes, such as turbulence, rapid deformation of materials, and other nonlinear dynamics where classical thermodynamics fails to provide an adequate description. Clausius–Planck inequality, which states that between two equilibrium states A and B

$$\Delta S \geq \int_A^B \frac{dQ}{T} \tag{3}$$

In rational thermodynamics, inequality (1) is written as

$$\frac{d}{dt}\int_{V(t)} \rho s dV \geq -\int_{\Sigma(t)} \frac{1}{T} q \cdot n d\Sigma + \int_{V(t)} \rho \frac{r}{T} dV \tag{4}$$

$$\rho \dot{s} + \nabla \cdot \frac{q}{T} - \rho \frac{r}{T} \geq 0 \tag{5}$$

Considering the Helmholtz free energy $\phi \left(= u - Ts\right)$ and eliminating r between the energy balance equation (1) and the inequality (4) leads to

$$-\rho(\phi + sT) - P{:}V - (1/q)\cdot\nabla T \geq 0 \tag{6}$$

This inequality, for a one-component uncharged system, is known as the *Clausius–Duhem* or the *fundamental inequality.*

## 2.10. Entropy Production

Entropy production remain a key concept in rational irreversible thermodynamics. Truesdell emphasized the concept of entropy production as a measure of irreversibility in non-equilibrium processes. He introduced the notion that entropy production always occurs in real processes and that the second law imposes constraints on the direction and magnitude of entropy production. Entropy production arises due to irreversible processes such as heat transfer, viscous dissipation, and chemical reactions.

In nonequilibrium thermodynamics, the entropy production rate σ must be non-negative:

$$\sigma = \frac{1}{T}\left(-\sum_i J_i \, \nabla\mu_i + \frac{q}{T}\nabla T + AR\right) \geq 0 \tag{7}$$

Where Ji·∇μi represents entropy production due to mass diffusion, qT·∇T represents entropy production due to heat conduction, AR represents entropy production due to chemical reactions.

Since irreversible processes always increase entropy, for spontaneous chemical reactions: AR>0 which means that reactions proceed in the direction that increases entropy until equilibrium is reached (A=0) In chemical kinetics and reaction engineering, RIT helps describe how chemical potentials drive reactions and how these reactions generate entropy. This is crucial for understanding the thermodynamics of nonequilibrium chemical systems. Entropy production must be non-negative in irreversible processes.

## 2.11. Constitutive Relations

Rational irreversible thermodynamics describes the behavior of materials and fluids using constitutive relations, which relate the fluxes of mass, energy, and momentum to the gradients of relevant thermodynamic variables [16]. These relations capture the response of materials to applied forces and gradients under non-equilibrium conditions. Ingo Müller's work explores the constitutive relations that define how materials respond to external forces, temperature changes, and other environmental factors in both elastic and inelastic regimes [17]. These relations are used to describe materials that undergo irreversible deformations (plasticity) or exhibit complex behaviors such as viscoelasticity (where materials show both solid and fluid-like responses depending on time and temperature).

## 2.12. Onsager's Reciprocity Relations

Onsager’s relations apply to linear processes near equilibrium, Rational Irreversible Thermodynamics often deals with non-linear and far-from-equilibrium processes. The constitutive relations in RIT are derived in a way that holds for a broader range of conditions than just near-equilibrium situations. Onsager's reciprocity relations are fundamental in rational irreversible thermodynamics. They establish relationships between the coefficients appearing in the constitutive equations, reflecting the symmetry of irreversible processes. These relations provide constraints on the coefficients and ensure the consistency of the thermodynamic description. By integrating entropy principles, material frame-indifference, and internal state variables, RIT refines and extends Onsager’s framework to accommodate nonlinear, memory-dependent, and non-equilibrium transport processes [18, 19].

For systems exhibiting nonlinear behavior, RIT extends Onsager’s framework beyond the linear regime

$$J_i = \sum_j L_{ij} X_j + \sum_{j,k} M_{ijk} X_j X_k + \cdots \tag{8}$$

Where $M_{ijk}$ are higher-order Onsager-like coefficients that allow for nonlinear transport phenomena. These terms become significant in strongly driven systems like turbulence, phase transitions, and biological transport.

## 2.13. Entropy Production Inequalities

Day (1977) [20], Rational irreversible thermodynamics often formulates the second law of thermodynamics in terms of entropy production inequalities. The entropy production inequality in RIT is a strict requirement that governs all irreversible processes. It is mathematically expressed through the Clausius-Duhem inequality, which ensures that the total entropy production in any thermodynamic process is non-negative. Specific processes, such as heat conduction, diffusion, viscous flow, and electromagnetic interactions, must all satisfy this inequality These inequalities impose constraints on the direction and magnitude of entropy production in non-equilibrium processes, providing a thermodynamic criterion for the feasibility and irreversibility of such processes.

### 2.14. Material Frame Indifference

Truesdell introduced the principle of material frame indifference, which states that the laws of thermodynamics should be independent of the choice of reference frame moving with the material. It helps ensure that the formulations remain invariant under transformations such as rotations and translations. This principle applies to all physical quantities, such as stress, heat flux, and strain, ensuring that they transform appropriately under rigid body transformations (rotations and translations). This principle allows for a consistent formulation of thermodynamic laws in both Lagrangian and Eulerian descriptions of fluid flow and solid deformation [21-22].

### 2.15. Generalization of Classical Thermodynamics

Truesdell's work contributed to the generalization of classical thermodynamics to encompass non-equilibrium phenomena. By formulating the second law in terms of entropy production inequalities and introducing principles such as material frame indifference, he provided a more comprehensive and rigorous framework for studying irreversible processes.

### 2.16. Impact on Rational Thermodynamics

Truesdell's formulations laid the foundation for the development of rational thermodynamics, which seeks to describe and analyze thermodynamic processes using principles of continuum mechanics and mathematical modeling. His contributions have influenced various fields, including fluid dynamics, solid mechanics, and chemical engineering By applying these concepts and assumptions, rational irreversible thermodynamics provides a systematic and rigorous framework for analyzing non-equilibrium processes and systems, incorporating both classical thermodynamic principles and modern concepts from continuum mechanics and mathematical modeling. It has diverse applications in fields such as fluid dynamics, solid mechanics, chemical engineering, and materials science.

## 3. Critical remarks

i) This theory gives a specific framework for providing the details of non-equilibrium processes and systems.

ii) It provides a systematic approach for studying irreversible phenomena based on thermodynamic principles.

iii) This theory also supports in understanding and predicting the behavior of complex systems far from equilibrium.

iv) It has diverse applications across various fields, including chemical engineering, physics, biology, and materials science.

v) RIT focuses a systematic approach based on thermodynamic principles, while CIT emphasizes on empirical relations and EIT extends CIT by incorporating non-equilibrium situations, non-linear effects, and more complex transport phenomena.

vi) Absolute temperature and entropy are considered primitive concepts. They are introduced a priori in order to ensure the coherence of the theory and do not have a precise physical interpretation.

## 4. Limitations of Rational Irreversible Thermodynamics

### 4.1. Simplified assumptions

Some of the assumptions made in rational irreversible thermodynamics may not be valid in all situations, leading to inaccuracies in predictions. For example, in the rapid expansion of a gas in an explosion, the quasi-static assumption breaks down [23].

### 4.2. Local equilibrium assumption

The validity of the local equilibrium assumption depends on the specific system and the nature of the processes involved. In some cases, such as in highly turbulent flows or rapidly changing systems, the assumption of local equilibrium may break down, requiring more advanced modeling techniques. In reality, systems may not always achieve or maintain local equilibrium, especially in highly dynamic or non-uniform systems. In fast processes where the system doesn't have enough time to relax to a local equilibrium state, this assumption can break down, limiting the validity of RIT in such cases. For example, combustion processes or turbulent fluid flow where local equilibrium may not be reached due to rapid changes in temperature and pressure gradients.

Linear response assumption fails in some cases, systems may exhibit non-linear behavior, especially under extreme conditions or when dealing with highly non-equilibrium processes. Non-linear behavior refers to situations where the relationships between fluxes (e.g., heat, mass, momentum) and their corresponding driving forces (e.g., temperature gradients, concentration gradients, stress) are not proportional, leading to more complex system dynamics. For example, the behavior of certain materials under high stress or temperature gradients may deviate significantly from linearity.

### 4.3. Homogeneous system assumption

In reality, many systems are heterogeneous, with spatial variations in temperature, pressure, and other properties. For instance, in chemical reactions occurring in porous catalysts or in biological systems, spatial variations in concentrations and temperature gradients can be significant.

### 4.4. Neglect of fluctuations

Fluctuations can play a significant role, especially in small systems. For example, fluctuations in particle number or energy can be important in systems with a small number of particles, such as colloidal suspensions or biological cells. In microfluidic channels or nanotechnology devices, the system sizes are on the order of microns or nanometers, which means the number of particles involved is small. This leads to significant statistical fluctuations in properties like concentration, pressure, or velocity.RIT describes systems using averaged, macroscopic quantities, assuming these fluctuations are negligible. However, in these small systems, fluctuations in particle number and energy can have a significant effect on transport processes, such as diffusion or heat conduction, making RIT's predictions inaccurate.

### 4.5. Memory Effects

Memory effects can introduce significant complexity into mathematical models and equations, especially in systems with long relaxation times or intricate temporal dependencies. It may be challenging to implement in practical engineering applications, particularly for systems with limited data availability.

### 4.6. Limited predictive power

While rational irreversible thermodynamics provides valuable insights, its predictive power may be limited in highly complex systems or situations with strong non-linearities.

### 4.7. Difficulty in experimental verification

Experimental verification of non-equilibrium thermodynamic predictions can be challenging due to the complexity of many systems and processes involved. For example, biological systems such as cells or ecosystems, industrial processes like chemical reactions in reactors, or atmospheric dynamics are all examples of complex systems where non-equilibrium thermodynamics may be applicable. Understanding and controlling all the variables in such systems to isolate specific non-equilibrium effects can be extremely challenging.

Similarly in a chemical reaction within a porous catalyst, there could be significant spatial variations in temperature and concentration, leading to complex reaction kinetics.

Experimentally controlling and observing non-equilibrium phenomena with high precision and accuracy can be challenging. External conditions must be carefully controlled, and measurements must be made with high sensitivity and resolution to detect subtle non-equilibrium effects. In many chemical systems, particularly at the microscopic scale (e.g., in solutions or gas reactions), molecular fluctuations lead to deviations from average behavior. Experiments often reveal stochastic variations in reaction rates, concentrations, or energy distributions that RIT fails to capture. RIT’s linear approximations cannot handle the complex, non-linear kinetics of multi-step or enzyme-catalyzed reactions [24-26]. Rational extended thermodynamics (RET) overcomes these limitations by extending the thermodynamic state space and incorporating higher-order fluxes, hyperbolic transport equations, and non-equilibrium moments [24-26].

## 5. Conclusion

Rational irreversible thermodynamics extends the scope of classical thermodynamics into the domain of irreversible phenomena by following the principles of entropy production, thermodynamic fluxes and forces, and constitutive relations. We know that RIT provides a valuable framework for studying non-equilibrium processes but inaccuracies in predictions may arise when the assumptions are violated in real-world systems. It is important to critically evaluate its assumptions, limitations, and applicability to specific systems and phenomena. Further research and development are required to overcome these limitations and improve the prediction and practical utility of non-equilibrium thermodynamic models.

Rational Irreversible Thermodynamics continues to evolve as a cornerstone of modern thermodynamics, combining mathematical accuracy with practical versatility. Its ability to unify diverse physical phenomena under a single theoretical framework enhance its importance in both fundamental science and engineering applications. This study highlights the need for ongoing interdisciplinary research to address emerging challenges and extend the boundaries of this powerful theoretical tool. Overcoming the limitations which we have discussed in the previous section usually involves integrating other theoretical approaches, employing advanced computational methods or refining existing models. For example:

1. In heat conduction at very small scales (e.g., nanoscale materials), non-local effects are significant. EIT can model heat transport in such materials more accurately by including terms that account for spatial variations in temperature gradients. [27, 28]
2. In turbulent fluid flow (Lumley 1983) [29] the system is far from equilibrium and linear models fail to capture the complexity of turbulence. Using nonlinear thermodynamics along with computational fluid dynamics (CFD) simulations can provide a more accurate description.
3. In thermoelectric materials, the heat flux and electrical flux are coupled. A coupled transport model that simultaneously considers thermal and electrical conductivities can more accurately predict the material's behavior.
4. The validity of frame-indifference in viscoelastic materials has been discussed by Bird and de Gennes (1983) [30]. In viscoelastic materials, the stress response depends on the history of strain. Time-dependent models, such as those using memory functions or hereditary integrals, can capture this behavior more accurately.
5. In small biological systems, such as cellular processes, thermal fluctuations can have a significant impact. Fluctuation-dissipation theorem and stochastic thermodynamics can model these fluctuations and their effects on the system [31].
6. In quantum dots or superconducting materials, quantum effects dominate. Quantum thermodynamics can model the energy exchange processes and coherence phenomena in these systems [32, 33].

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