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SYNTHESIS OF ECOFRIENDLY DETERGENTS FROM VEGETABLE BASED POLYMER
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Rational Irreversible Thermodynamics – A Review

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Abstract-

The basic hypotheses underlying rational thermodynamics have been studied and reviewed in this paper. Also critical remarks on RIT have been discussed with comparison to other irreversible theories like Classical Irreversible Thermodynamics and Extended irreversible thermodynamics.

I. Introduction:

This formalism was essentially developed by Coleman (1964)³, Truesdell (1984)⁴, and Noll (1974)⁵ and follows a line of thought drastically different from CIT. Its main objective is to provide a method for deriving constitutive equations. The basic hypotheses underlying rational thermodynamics can be summarized as follows.

1. 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.
2. It is assumed that materials have a memory, i.e. the behaviour of a system at a given instant of time is determined not only by the values of the characteristic parameters at the present time, but also by their past history. The local equilibrium hypothesis is no longer assumed, since the knowledge of the values of the parameters at the present time is not enough to specify unambiguously the behaviour of the system.
3. The general expressions previously formulated for the balance of mass, momentum, and energy are however retained. Nevertheless, there are two essential nuances. 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

$$\rho \dot{u} = -\nabla \cdot q - P : \nabla v + \rho r \quad (1)$$

r is generally referred to as the power supplied or lost by radiation. The second crucial point is that the body forces F and the radiation term r are not given a priori as a function of

position and time but are computed from the laws of momentum and internal energy respectively¹.

4. Another capital point is the mathematical formulation of the second law of thermodynamics, which serves essentially as a restriction on the form of the constitutive equations. The starting relation is the Clausius–Planck inequality, which states that between two equilibrium states A and B one, has

$$\Delta S \geq \int_A^B \frac{dQ}{T} \quad (2)$$

In rational thermodynamics, inequality (2) is written as

$$\frac{d}{dt} \int_{V(t)} \rho s dV \geq - \int_{\Sigma(t)} \frac{1}{T} q \cdot n d\Sigma + \int_{(y)_t} \rho \frac{r}{T} dV \quad (3)$$

or, in local form, as

$$\rho \dot{s} + \nabla \cdot \frac{q}{T} - \rho \frac{r}{T} \geq 0 \quad (4)$$

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

$$-\rho(\dot{\phi} + s\dot{T}) - P : V - \frac{1}{T} q \cdot \nabla T \geq 0 \quad (5)$$

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

An important problem is certainly the selection of the constitutive independent variables. This choice is subordinated to the type of material one deals with. In hydrodynamics, it is customary to take as variables the density, velocity, and temperature fields. It is also known that the balance laws and Clausius–Duhem inequality introduce complementary variables, such as the internal energy, the heat flux, the pressure tensor, and the entropy. The latter are expressed in terms of the former by means of constitutive equations. By an *admissible process* is meant a solution of the balance laws when the constitutive relations are taken into account and the Clausius–Duhem inequality holds².

II. Axiom of Memory

If it is admitted that the present is influenced not only by the present state but also by the past history, the constitutive relations² will depend on the whole history of the independent variables. If $\varphi(t)$ designates an arbitrary function of time, say the temperature or the strain

tensor, its history up to the time t is defined by $\varphi(t-t')$ with $0 \leq t' < \infty$. The axiom of memory asserts that the behaviour of the system is completely determined by the history of the set of selected independent variables. This means that the free energy, the entropy, the heat flux and the stress tensor, for instance, will be expressed as functionals of the history of the independent variables. Considering the problem of heat conduction in a rigid isotropic material, an example of constitutive equation with memory is Fourier's generalized law

$$q(t) = \int_{-\infty}^t \lambda(t-t') \nabla T(t') dt' \quad (6)$$

where $\lambda(t-t')$ is the memory kernel. When this expression is substituted in the energy balance, one obtains an integro-differential equation for the temperature field, after use is made of $\dot{u} = c\dot{T}$ with c the specific heat capacity. If the memory kernel takes the form of an exponential like $(-\lambda/\tau) \exp[-(t-t')/\tau]$, the time derivative of (8) is given by

$$\tau \dot{q} = -q - \lambda \nabla T \quad (7)$$

which is the same Cattaneo equation as in EIT. It is important to realize that this result has been obtained by considering only the temperature as single state variable. This is a characteristic of RT where the state space is generally restricted to the classical variables, i.e. mass, velocity (or deformation), and temperature, while the fluxes are expressed in terms of integral constitutive equations containing the whole history of the independent variables. Instead of assuming that q depends on the whole history of temperature field, in practical applications it is assumed that q is a function of ∇T and its higher order time derivatives. If the memory is very short in time, one may restrict this sequence to a limited number of terms. But even in this case, RT offers an interesting formalism which departs radically from that of classical irreversible thermodynamics.

III. Axiom of Equipresence

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 inequality². 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 relations².

IV. Axiom of Local Action

It is admitted that a material particle is only influenced by its immediate neighbourhood and that it is insensitive to what happens at distant points. Practically, it means that second and higher-order space derivatives are excluded from the constitutive relations. Higher spatial gradients have however been included in some developments of the theory on non-local actions².

V. Critical Remarks

Rational thermodynamics has not been free of criticisms, such as:

1. Temperature and entropy remain undefined objects. For example, it is not possible to check whether or not the temperature measured by a thermocouple corresponds to the temperature T used in rational thermodynamics. Concerning the entropy, no measure is given for determining its actual functional dependence, either by experiment or by calculation from a physical model. Furthermore, it has been demonstrated by Day (1977)⁶, who examined the problem of the temperature distribution in a rigid heat conductor with memory, that the value of the entropy is not unique.
2. The fundamental inequality (5) used in rational thermodynamics is not, particularly the Clausius inequality. Actually it is given by (2) and connects two equilibrium states. In rational thermodynamics, Clausius' expression is generalised to arbitrary non-equilibrium states. When dealing with the Clausius–Duhem inequality in rational thermodynamics, it must be understood that the existence of a specific entropy that satisfies the fundamental inequality (5) has been postulated. The latter also implies that the entropy flux is given by the heat flux divided by the temperature, a result only valid in the vicinity of equilibrium, as shown in the kinetic theory of gases.
3. Likewise, rational thermodynamics predicts unphysical properties in some classes of rheological materials. If it is admitted that the Rivlin–Ericksen model provides a good description of rheological bodies, then the signs of some material coefficients, as given by the rational approach, are found to be in contradiction with experimental data.
4. Although the principle of material frame-indifference has revealed itself as a useful tool in establishing constitutive equations in continuum mechanics, it has recently been stressed that the two requirements of the principle, namely form invariance and frame independence, are not satisfied in several disciplines, such as classical mechanics, kinetic theory of gases, turbulence, rheology, and molecular hydrodynamics. In kinetic theory, it has been shown that the Burnett constitutive relations are frame-dependent (Muller 1972⁷; Edelen and McLennan

1973⁸); the origin of the frame-dependence lies in the Coriolis force of the rotating frame. A similar problem arises in turbulence theory (Lumley 1983)⁹; there is an ample experimental confirmation that turbulence, in a non-inertial frame, is quite different from turbulence in an inertial frame, owing to the dependence of the turbulent viscosity on the angular velocity of the reference frame. The validity of frame-indifference in viscoelastic materials has been discussed by Bird and de Gennes (1983)¹⁰. It was concluded that inertial forces can contribute to the material functions of viscoelastic media and that frame indifference is useful only whenever inertial effects are negligible. Another example of violation of the principle of material frame-indifference is provided by the phenomenological coefficients L_c of CIT.

When measured in a rotating frame, the L_c are known to depend on the angular velocity, as pointed out earlier. Hoover et al. (1981)¹¹ performed a molecular dynamics simulation for a fluid modelled by two-dimensional rotating disks: they found an angular component for the heat flux, in contradiction with the material Frame-indifference. All these observations have cast serious doubts about the general validity of the principle of material frame-indifference (Murdoch 1983¹²; Ryskin 1988¹³).

In RT, the presence of the source terms F and r is required to guarantee that the state variables and their time derivatives can be varied independently. The quantities F and r are selected in order that the momentum and energy equations are identically satisfied. In normal circumstances, the situation is reversed: F and r are known from the start and take well-defined values. This limits seriously the domain of applicability of the theory but this difficulty can be circumvented by using a technique proposed by I.S. Liu (1972)¹⁴ and based on the introduction of Lagrange multipliers.

5. From a practical point of view, the constitutive equations, when they are written in their general form involving functionals dependent on the whole history of the variables, are not easily tractable and generally require the knowledge of too vast an amount of information.

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Synthesis of Ecofriendly Detergents from Vegetable Based Polymer

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ABSTRACT

Polymer based on sorbitol, maize starch, acids, and anhydrides has been synthesized and used successfully in detergent compositions. In the present work a small quantity of oxalic acid and citric acid along with major quantity of maize starch and sugar solution has been used in synthesis of polymer. The overall idea is to develop a polymeric using higher quantity of maize starch and substantial quantity of sugar along with oxalic acid and citric acid. These polymers are ecofriendly and based totally on vegetable products. The synthesized polymers have been analyzed for physicochemical characteristics like acid value, saponification value, HLB ratio, and cleaning efficiency. Based on the physicochemical observations, some selected polymers have been used in the preparation of powder detergent and liquid detergent. The acid slurry and Alpha Olefin Sulphonate based on crude petroleum have been replaced to some extent by these starch-sugar based polymers. The preparation of these polymers is simple and has inherent character of biodegradability. These novel polymers are economically viable and suitable for commercial production. They are cheaper than petroleum based products and can be recommended for commercial exploration.

Keywords: Polymer, Sorbitol, Starch, Ecofriendly, Biodegradability, etc.

INTRODUCTION:-

The polymers containing carbohydrates are potentially process-able and biodegradable and biocompatible polymers. The present research work encompasses the polymer synthesis from carbohydrates.

Synthesis of carbohydrate polymers based on sugar, sorbitol, maize starch, acid anhydrides (phthalic and maleic) and small quantity of oxalic acid and citric acid. The oxalic acid and citric acid have been specially used to study their effect on foam, detergency and stain removing characteristics. In this synthesis various catalysts like HCl, sodium bisulphite, sodium bisulphate and commercial tween 80/20, have been used.

The detergents made out of acid slurry causes harm to aquatic flora and fauna. Acid slurry has a petroleum origin. The detergents of petroleum origin is responsible for river foaming and eutrophication. By using biodegradable polymers in detergent formulations the above mentioned problems of water pollution can be minimized to a greater extent.

EXPERIMENTALS:-

The synthesis of polymers was carried out in a glass reactor. The reactor consists of two parts. Lower part of the reactor is a round bottom vessel with very wide mouth. The upper part of the reactor is its lid, having four necks with standard joints.

Motor driven stirrer was inserted in the reactor through the central neck, while another neck was used for thermometer. A Condenser was fitted with the reactor through the third neck. And the further neck was used for dropping the chemicals in to the reactor. The reactor was heated by an electric heating mantle having special arrangement for smooth control of the temperature of the reactor. A regulator controlled the speed of the stirrer. The reaction vessel and its lid were tied together with help of clamps.

In our earlier experiments we have successfully prepared polymer based on natural products of vegetable origin like maize starch, sorbitol, vegetable oils, and glycerin in synthesis of polymer along with acid anhydride like maleic anhydride and phthalic anhydride. These novel polymers have been successfully used in formulation liquid, powder and cake detergents as practical or total replacement of acid slurry of petroleum origin.

Now in the present work we wish to incorporate sugar which is abundantly available throughout the world and small quantity of oxalic acid and citric acid.

Table No.1:- Composition of Carbohydrate Polymers

Ingredients	Batch B1	Batch B2	Batch B3
Sorbitol (100%)	39.8	-	24
Maleic anhydride	3.83	-	4.0
Phthalic anhydride	1.53	4.1	1.6
Sugar (100%)	13.33	41.2	24.0
Maize Starch	7.66	-	30
Benzoic acid	0.8	-	-
Oxalic acid	3.83	4.1	4.0
Citric acid	3.83	-	4.0
glycerol	-	32.9	-
Water	25.61	17.7	9.8
Sodium bisulphate	-	1.5	1.5
Sodium bisulphite	-	0.5	0.5
Tween 80/20	30	-	-
HCL	0.5	-	-

Note:

1. All data is reported for 100% solids.
2. Actually Sorbitol is used as 70% solids & Sugar is also used as 70% solids. HCL (1%) is used as catalyst in B1, B2 & B3.
3. Sodium bisulphate (1.5%) and sodium bisulphate (0.5) has been used as catalyst in all batches.

Table No. 2:- Cooking Schedule of Polymers

Sr.no.	Heating period	Batch B1	Batch B2	Batch B3
1	After ½ hr	80°C	80°C	80°C
2	After 1 hr	130°C	110°C	110°C
3	After 1 ½hr	130°C	130°C	130°C
4	After 2 hr	130°C	130c	130°C
5	After 2 ½ hr	130°C	130°C	130°C
6	After 3 hr	130°C	130°C	130°C

Table No. 3:- %YIELD AND BATCH SIZE

	Batch B 1	Batch B2	Batch B3
Yield	90.2	92.36	94.08
Batch size	1	1	1

Neutralization of Polymers:-

Weighted amount of polymer are taken whose acid value was known to us. 30% solution of KOH used to neutralize the polymer so that acid value reached to zero. During neutralization, temperature of the polymer & 30% KOH solution are maintained at 60⁰ C. 30% KOH was added in about 15 min. with constant stirring.

Analysis of Polymers

Physicochemical analysis & Spectroscopic analysis for prepared polymers are undertaken. For physicochemical properties the standard techniques have been used to evaluate the following constants.

Table 4:- Physico- Chemical Analysis of Novel Carbohydrate Polymers

Sr. no.	Test	Batch B1	Batch B 2	BatchB3
1	% Solid	91.09	69.23	96.5
2	Acid value	63	32.25	61.47
3	Viscosity (in sec.)	340	312	514
4	Color	Dark brown	Dark brown	Dark brown
5	Consistency	Thick	Thick	Thick
6	Solubility	Water	Water	Water
7	PH. value	3.92	3.25	4.83
8	Molecular weight	5417.5	4885.6	5213.7
9	Ester value	488	219.55	171.12
10	Epoxy value	12	24	16

Table No.5:- Stain Removing Characteristics of Various Polymers on cotton cloth

Sr. no.	medium	Batch B1	Batch B2	Batch B3	Comm.1	Comm.2
1	Soil	91.90	93.06	92.65	94.12	94.56
2	Tea	86.48	78.37	85.32	88.23	88.54
3	Coffee	88.57	80	87.54	89.63	90.32
4	Palak	92.5	80	92.51	93.12	93.24

Preparation of Powder Detergent:-

The various ingredients and the composition of detergents powder are as shown in the formulations. The said ingredients in the powdered form are weighed and mixed thoroughly in a tray. Then add liquid ingredients like linear alkyl benzene sulphonate, Alpha olefin sulphonate and neutralized resin. Whole mass is then homogenized thoroughly. This mixture then poured in a homogenizer pot. After mixing, the homogeneous mass thus obtained is taken out in a try and kept out in open air for drying. After complete drying, the solid mass thus formed is grind again in a mixer to get homogenized detergent powder.

Table No.6:- POWDER DETERGENT BASED ON THESE POLYMERS

S.N.	INGREDIENTS	PD1	PD2	PD3	PD4
1	Our Polymer (B)	10.52	18.86	10.30	14.81
2	Acid Slurry	1.04	-	1.02	2.40
3	A.O.S.	1.04	-	1.02	2.40
4	S.L.S.	5.26	4.71	5.15	4.81
5	Dolomite	31.57	28.30	30.91	28.92
6	Sodium carbonate	31.57	28.30	30.91	28.92
7	S.T.P.P.	5.26	4.71	5.15	4.81
8	Urea	3.15	2.83	3.08	4.81
9	Sodium Sulphate	5.26	4.71	5.15	2.89
10	E.D.T.A.	0.20	0.18	0.20	0.192
11	Robin Blue	2.0	2.0	2.0	2.0
12	Whitener	3.13	5.40	5.11	3.04
	TOTAL	100	100	100	100

*PD1, PD2, PD3, PD4 –All samples are based on our polymer (B)

Table No.7:- Analysis of Powder Detergents at 1.0% conc.

Sr.No.	Powder detergent	Foam volume in (cm ³) foam stability after				density	Surface tension Dynes/cm
		0 min.	5 min.	7 min.	10 min.		
1	PD1	900	900	85	850	0.9985	30.93
2	PD2	950	950	950	950	0.9958	36.28
3	PD3	1000	950	950	900	0.9985	33.10
4	PD4	1000	950	900	900	0.9981	35.58
5	CD1	1000	1000	950	950	0.9996	62.11
6	CD2	1000	1000	950	950	1.02	62.35

Table No 8:-Effect of Powder Detergent on %Detergency Soil Stain on polyester, Terri cot, and cotton cloth samples

Sr.No.	Powder detergent	Conc. %	POLYSTER		TERICOT		COTTON	
			RW	detergency	RW	detergency	RW	detergency
1	PD1	0.1	94	91.17	89	83.82	95	92.53
2		0.25	96	94.11	91	86.76	95	92.53
3		0.5	97	95.58	95	92.64	97	95.52
4		1	98	97.05	95	92.64	98	95.58
1	PD2	0.1	96	94.11	87	80.88	95	92.53
2		0.25	96	94.11	90	85.29	97	95.52
3		0.5	98	97.05	93	89.70	98	95.58
4		1	99	98.52	96	94.11	99	97.05
1	PD3	0.1	91	86.76	90	85.29	96	92.64
2		0.25	94	91.17	91	86.76	97	95.52
3		0.5	96	94.11	95	92.64	97	95.52
4		1	97	95.58	96	94.11	98	95.58
1	PD4	0.1	88	82.35	87	80.88	91	85.29
2		0.25	90	85.29	89	83.82	92	86.76
3		0.5	91	86.76	90	85.29	93	88.23
4		1	92	88.23	91	86.76	94	89.70
1	CD1	0.1	92	88.23	94	91.17	92	86.76
2		0.25	94	91.17	96	94.11	94	89.70
3		0.5	96	94.11	97	95.58	95	92.53
4		1	97	95.58	98	97.05	96	92.64
1	CD2	0.1	94	91.17	90	85.29	88	82.08
2		0.25	95	92.64	93	89.70	90	85.07
3		0.5	97	95.58	96	94.11	93	89.55
4		1	98	97.05	99	98.52	96	92.64

Results and Discussion

Powder Detergents

The composition of various formulations based on Sugar, Starch & Sorbitol Resin are given in Table 6 Composition of Selected Powder Detergent.

The special features of these formulations are

1. They are using very limited amount of STPP i.e. only 5.21% therefore problem of Polluting rivers and lakes is reduced considerably
2. The soil & Stain removing capacity is many times comparable and better than commercial product.
3. No special additives like enzyme have been used.
4. The cost is moderate and reasonable.

CONCLUSION:-

- a. Sorbitol, sugar and starch based surfactants can be used successfully for formulations of powder detergents. A combination of small amount of organic acids like oxalic acid, phthalic anhydride and citric acid in polymer give excellent properties for detergent formulations. Particularly use of oxalic and citric acid helps in improving detergency.
- b. Conventional activities can be replaced by these novel polymers to the extent of 50 to 90% without affecting detergency. The foam is normally adversely affected in many compositions. For particular use (High foaming, medium foaming & foamless) detergents can be selected depending on commercial requirement.
- c. Sugar and starch can be used polymeric surfactants in combination with sorbitol, without affecting adversely the property of detergents.
- d. We can reduce the amount of petroleum based actives to the extent of 50-80% this is very important achievement. As we are formulating a product which is vegetable based ecofriendly and biodegradable.
- e. The use of 1% HCL as catalyst is giving positive results. The use of sodium bisulphate (1.5%) and sodium bisulphite (0.5%) give good color and homogeneity to product.
- f. The following samples have been identifies as excellent. PD1, PD3, and PD4.

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