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# Dispersion—An Entropy Generator of Diffusion

In multicomponent gaseous flows with chemical reactions, diffusion is usually considered to be a single-source entropy generator. Introducing the concept of dispersion, where the atom balance is dominant rather than the molecular, shows in an ideal setting that, even when there is diffusion, the dispersion part of entropy generation can be reduced to zero by proper choice of species flow velocities. Fuel cells and methane reformers are employed as examples to illustrate these concepts.

#### Introduction

Is it possible to have diffusion without having entropy generation in a multicomponent flow, with chemical reactions? In mechanics, examples used to illustrate reversible movement of a mass on a frictionless plane, show that that energy may be borrowed from an outside source to accelerate the mass. Then the energy is returned to the source to decelerate the mass; all the while no entropy generation occurs. In chemical equilibrium, a reaction may occur in one direction, while nearby, the reverse reaction is occurring resulting in no entropy generation. Can these principles be applied to complex multicomponent flow processes?

Real world (macroscopic) processes are always irreversible, but it is useful to examine theoretical cases with the objective of determining the minimum entropy generation for a process as a benchmark for the practical case. In this paper, the focus is on diffusion and how it contributes to entropy generation. It is shown that a (theoretical) fuel cell can be operated, by proper manipulation of the flow rates of the chemical species in the direction of the fuel stream bulk flow, so as to produce vanishingly small entropy generation. A new concept in the study of diffusion under chemical equilibrium is proposed in which kinetics has been removed. Its entropy generation can be reduced to zero.

In a real fuel cell, set conditions for operation yield a unique thermodynamic path for the cell, and this is a large entropygenerating path. What that path is, is not of interest in this paper. Here, with the aforementioned minimization objective in mind, several sources of irreversibility other than diffusion are removed from consideration. The theoretical fuel cell (hightemperature methane/air solid oxide, SOFC) is assumed to operate isothermally ( $T = 1000^{\circ}$ C, 1832°F), at atmospheric pressure with no (total) pressure drop throughout the cell, and with no interaction with the cell walls (frictionless). The fluids are treated as ideal gases. The cell is considered one-dimensional in the direction of the bulk flow of the gases, and no electrochemical losses (polarization losses) such as electrical resistance or activation losses in the electrolyte is allowed. No kinetic energy or gravitational effects will be included in the energy balance. The only entropy-generating mechanism considered is diffusion in the direction of the bulk flow.

The concept of *dispersion* is introduced, a special form of diffusion that produces irreversibility. Suppose methane is partially combusted, brought to chemical and thermal equilibrium at high temperature, and then fed to a pipe at a uniform rate. In the steady-state setting, the amount of carbon per second passing is the same at every point along the length of the pipe,

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even if diffusion occurs. A combustion product, say CO, may be allowed to "slow down" compared to the bulk flow, but other carbon-containing species such as CH4 or CO2 must compensate. It is important to understand that the process flow can remain in chemical equilibrium, with constant partial pressures  $P_i$ , while the molar flow rates of the species are being manipulated. Since the chemical composition remains constant in the pipe, the atom ratios C:H:O remain constant, i.e., there is no 'dispersion of the atoms," but there can be diffusion. It will be shown that it is dispersion at the atom level that causes entropy generation, and not just relative flow rates of the species at the molecular level. In the pipe, one chemical species stream can borrow energy from another without entropy generation due to dispersion. These concepts will be extended and verified later in the fuel cell case where the chemical composition is forced to vary.

Finally, the case of a methane reformer, which must generate entropy, is considered, and it is compared to the fuel cell case.

### The Isothermal Fuel Cell Case

In this section, the equations for entropy generation are developed for the isothermal, constant pressure SOFC. Implicitly in Gaggioli and Dunbar (1993) and explicitly in Call (1996), the assumption is made that the volumetric flow rates of the chemical species in the fuel stream are equal. This is not assumed here, but their methods are adapted. Figure 1 shows a sketch of a single cell, while Fig. 2 gives the control volume (CV) parameters: T = 1273 K (1832°F), P = 101.325 kPa (14.7 psi). The fuel is syngas, consisting of 1 mol/s CH<sub>4</sub> (7.92 lbmol/ h) and 1 mol/s H<sub>2</sub>O previously reformed and brought to chemical equilibrium at T and P (heat needed to do this is not part of the CV). The air (stochiometric) is modeled as 2 mol/s (O<sub>2</sub>) + 3.76 $N_2$ ) at T and P (again heat needed is not part of the control volume). The parameter x (not a linear distance) from 0 to 1 represents the fraction of oxygen in the air that is consumed. POW is the (electrical) power generated by the cell (positive) and  $\dot{O}$  is the heat into the cell needed to keep the cell at a constant temperature (negative when heat flows out). It is assumed that only six products, labeled 1-6, are formed on the fuel side and these are in the order: CH<sub>4</sub>, CO, CO<sub>2</sub>, H<sub>2</sub>,  $H_2O$ ,  $O_2$ . Also, assume that only  $O_2$  and  $N_2$  exist in the air stream, labeled 7 and 8, respectively.

Enthalpy balance for the CV:

$$\sum_{j=1}^{8} n'_{j} h_{j} + \dot{Q} = \text{POW} + \sum_{i=1}^{8} n'_{i} h_{i}$$
(1)

Entropy balance for the CV:

$$\sum_{j=1}^{8} n'_{j} s_{j} + \frac{\dot{Q}}{T} + S'_{\text{gen}} = \sum_{i=1}^{8} n'_{i} s_{i}$$
(2)

Eliminate  $\dot{Q}$  to obtain



Fig. 1 Westinghouse solid oxide cell located at http://www.metc.doe .gov/projfact/power/fc/west\_so.html

POW + 
$$TS'_{gen} = \sum_{j} n'_{j}(h_{j} - Ts_{j}) - \sum_{i} n'_{i}(h_{i} - Ts_{i})$$
  
=  $-\Delta G'$  (3)

There is a reversible isothermal thermodynamic path between the input and output states. In that case, POW is the total power, and since  $S'_{gen} = 0$ , it follows that the total power that can be extracted from the cell is  $-\Delta G'$ .

The task now is to find explicit expressions in terms of the  $n'_i$ 's and the  $P_i$ 's for POW and  $TS'_{gen}$ . The  $n'_i$ 's and the  $P_i$ 's are in turn functions of the parameter x, i.e., depend on the particular thermodynamic path chosen. The general entropy generation equations for multicomponent fluid systems are given in Teng et al. (1996), but those are in terms of a Eulerian frame of reference. The needed form will be derived here independently, using the material and pressure constraints.

The two pressure constraints are

$$\sum_{i=1}^{6} P_i = P = P_7 + P_8 \qquad (4a, b)$$

Material balance is given by the matrix equation

$$\begin{bmatrix} n_{1}'\\ n_{2}'\\ n_{3}'\\ n_{3}'\\ n_{4}'\\ n_{5}'\\ n_{6}'\\ n_{7}'\\ n_{8}' \end{bmatrix} = \begin{bmatrix} 1\\ 0\\ 0\\ 0\\ 1\\ 1\\ 0\\ 2\\ 7.52 \end{bmatrix} + \begin{bmatrix} 0 & -1 & 0 & 0\\ 0 & 1 & -1 & 0\\ 0 & 0 & 0 & 0\\ 0 & 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} x\\ \epsilon_{1}\\ \epsilon_{2}\\ \epsilon_{3} \end{bmatrix}$$

## Nomenclature .

- $\mathscr{A}$  = activity, J/mol g = molar Gibbs free energy h - Ts,
- J/molG' = total molar Gibbs free energy
- flow  $\sum n'_i g_i$ , W
- $h_i = \text{molar enthalpy}, \text{J/mol}$
- $n_i' = \text{molar flow, mol/s}$
- P = total pressure, Pa
- $P_i$  = partial pressure, Pa POW = power, W
  - $\dot{Q}$  = heat flow, W
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R =ideal gas constant, J/mol-K

- s = molar entropy, J/mol-K
- $S'_{gen} = entropy generation, W/K$
- T =temperature, K
- $V_i' =$  volumetric flow rate of *i*th component, m<sup>3</sup>/s
- x = parameter, fraction of oxygen consumed
- $\epsilon_i$  = parameter,  $i = 1, \ldots, 3$
- $\rho_i = \text{molar density, mol/m}^3$
- $\tau = P_1 + P_2 + P_3, \text{ Pa}$



Fig. 2 Inputs and outputs to the control volume in the fuel cell case at an arbitrary value of the fuel utilization x

Equations (5g) and (5h) say that  $n'_{7}$  and  $n'_{8}$  are already unique functions of x since there are no chemical reactions in the air stream (except for diffusion of O<sup>-2</sup> ions across the solid electrolyte to the fuel side, done reversibly), only depletion of O<sub>2</sub>. Since there are three atom constraints (carbon, hydrogen, and oxygen) on the fuel side, and there are six chemical species, the six molar flow rates can be written in terms of three parameters (the  $\epsilon$ 's) and of course x. A standard parameterization for the first seven rows of the four columns of the large matrix is determined by the stochiometric coefficients of the four reactions  $2O_{2,\text{suir}} \rightarrow 2O_{2,\text{fuel}}$ ;  $CH_4 + H_2O \leftrightarrow CO + 3H_2$ ; CO + $H_2O \leftrightarrow CO_2 + H_2$ ;  $0.5O_2 + H_2 \leftrightarrow H_2O$ . The last three are in equilibrium while the first is put into equilibrium by a counter voltage. The  $\epsilon$ 's are functions of x and depend on the thermodynamic path chosen.

Next, compute the differential of Eqs. (3), (4), (5), and substitute the differentials  $n'_i dg_i = -n'_i s_i dT + (n'_i / \rho_i) dP_i = V'_i dP_i$ 

$$dPOW + TdS'_{gen} = -\sum_{i}^{\circ} n'_{i} dg_{i} - \sum_{i}^{\circ} g_{i} dn'_{i}$$
$$= -\sum_{2}^{6} (V'_{i} - V'_{1}) dP_{i} - (V'_{7} - V'_{8}) dP_{7}$$
$$+ \mathcal{A}_{x} dx + \mathcal{A}_{1} d\epsilon_{1} + \mathcal{A}_{2} d\epsilon_{2} + \mathcal{A}_{3} d\epsilon_{3} \quad (6)$$

where the activities are  $\mathscr{A}_x = -(2g_6 - 2g_7), \ \mathscr{A}_1 = -(g_2 + 3g_4 - g_1 - g_5), \ \mathscr{A}_2 = -(g_3 + g_4 - g_2 - g_5), \ \mathscr{A}_3 = -(g_5 - g_4 - 0.5g_6).$  With the previous assumption that irreversibilities associated with chemical reactions are negligible  $(\mathscr{A}_1 = \mathscr{A}_2 = (5a-h) - \mathscr{A}_3 = 0)$ 

## Subscripts

- $1-8 = CH_4, CO, CO_2, H_2, H_2O, O_{2, fuel}, O_{2, air}, N_2$ 
  - eq = at chemical equilibrium
  - i = chemical species at exit of CV
  - j = chemical species at entrance

## Superscripts

Act = actual

\* = classical case

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Fig. 3 Molar flow rates (fuel side) versus fuel utilization for the ideal isothermal fuel cell with no diffusion

$$dPOW = \mathscr{A}_x dx \tag{7a}$$

$$TdS'_{gen} = -\sum_{2}^{0} (V'_{i} - V'_{1})dP_{i} - (V'_{7} - V'_{8})dP_{7} \quad (7b)$$

The Classical Case of a Reversible Fuel Cell. In the classic reversible case it is assumed not only that the fuel side is in chemical equilibrium at each value of x, but also that there is no relative diffusion on either the fuel or the air side, i.e.,

$$\mathcal{A}_{1} = \mathcal{A}_{2} = \mathcal{A}_{3} = 0$$

$$V'_{1} = V'_{2} = V'_{3} = V'_{4} = V'_{5} = V'_{6}$$

$$V'_{7} = V'_{8} \qquad (8a-i)$$

This yields  $dS'_{gen} = 0$  from Eq. (7b). Since there are 19 unknown functions of x (8  $P_i$ 's and 8  $n'_i$ 's, 3  $\epsilon_i$ 's) and 19 equations (Eqs. (4), (5), and (8)), the system can be solved. Figures 3 and 4 show the results of the  $n'_i$ 's and the  $P_i$ 's as functions of x. These solutions will be denoted by  $n_i^*$ ,  $P_i^*$ ,  $\epsilon_i^*$ . This is the same case as in Call (1996).

The Case of "Reversible" Diffusion. There is another way, at least mathematically, in which  $dS'_{gen}$  can be made zero for all values of x. Again, constant pressure (Eqs. (4)), material balance (Eqs. (5a-h)), chemical equilibrium (Eqs. (8a-c)), and Eq. (8i) are assumed.

Now take differentials of Eqs. (8a-c), and with  $dg_i = (RT/$  $P_i$ )  $dP_i$  and  $P_iV'_i = n'_iRT$ , eliminate  $dP_2$ ,  $dP_3$ , and  $dP_4$  as well as the  $n_i$ 's from Eq. (7b) to obtain (the  $\epsilon_i$ 's will cancel!)

$$TdS'_{gen} = -\frac{RT}{2\tau} \left\{ (2P_1 + 6P_2 + 6P_3 - 2P_4 - 2P_5) \frac{dP_5}{P_5} + (4xP_1 + (4x - 3)P_2 + (4x - 4)P_3 + P_4 - 2P_6) \frac{dP_6}{P_6} \right\}$$
(9)

where  $\tau = P_1 + P_2 + P_3$ . The coefficients of  $dP_5$  and  $dP_6$  in Eq. (9) can be related to physical phenomena. The ratio (O:C)<sup>Act</sup> of oxygen atoms to carbon atoms in the fuel side, at any x, is determined by the partial pressures of the oxygen and carbon containing species. On the other hand, the classical case ratio (O:C)\* is determined by the input molar flow rates (1 mol/s CH<sub>4</sub>, 1 mol/s H<sub>2</sub>O, and  $2x \text{ mol/s } O_2$ ; the composition can be computed from these numbers as there is no diffusion; similarly, for the actual hydrogen/carbon ratio (H:C)<sup>Act</sup> and the classical ratio (H:C)\*. The formulas for the four ratios where  $\tau = P_1 + P_2 + P_3$  are

$$(O:C)^{Act} = (P_2 + 2P_3 + P_5 + 2P_6)/\tau$$
$$(O:C)^* = (4x + 1)/1$$
$$(H:C)^{Act} = (4P_1 + 2P_4 + 2P_5)/\tau$$
$$(H:C)^* = 6/1 \qquad (10a-d)$$

Substitution of Eqs. (10a-d) into Eq. (9) yields

$$TdS'_{gen} = \frac{RT}{2} \left\{ ((\text{H:C})^{\text{Act}} - (\text{H:C})^*) \frac{dP_5}{P_5} + ([(\text{O:C}) - 0.5(\text{H:C})]^{\text{Act}} - [(\text{O:C}) - 0.5(\text{H:C})]^*) \frac{dP_6}{P_6} \right\}$$
(11)

Define no dispersion (of atoms) to mean  $(O:C)^{Act} = (O:C)^*$ and  $(H:C)^{Act} = (H:C)^*$ , and *dispersion* if these two equations do not hold. It is clear that  $dS'_{gen} = 0$  exactly when there is no dispersion, i.e., when the following two equations hold:

$$3(P_1 + P_2 + P_3) = 2P_1 + P_4 + P_5$$
  
+ 
$$4x(P_1 + P_2 + P_3) = -P_1 + P_3 + P_5 + 2P_6 \quad (12a, b)$$

Note that there are enough equations in the case of no dispersion to determine the  $P_i$ 's for i = 1, ..., 6 as functions of x, using Eqs. (4a), (8a-c), and (12a, b). For i = 7, 8, the  $n_i$ 's are given by Eqs. (5g, h) and the  $P_i$ 's can be determined by Eqs. (4b), (8i), and  $P_i V'_i = n'_i RT$ , and these values are the same in the classical case, viz.,  $n'_i$ ,  $P_i^*$ . Even though in this nondispersive case there is a unique solution for the  $P_i$ 's, there is not a *unique* solution for the remaining  $n_i'$ 's, i = 1, ..., 6 (in fact there are three degrees of freedom). In contrast, there is a unique solution for the  $n_i'$ 's in the classical case. The next section shows the connection between the two cases.

Comparison of the Classical and the Nondispersive Cases. The difference between the two cases is in solving for the  $n'_i$ 's,  $i = 1, \ldots, 6$ . In the no dispersion case, the five constraints (8d-h) are removed and the two constraints (12a, b) are added, a net loss of three equations. The precise connection that shows what these three equations are is given by the following algebraic result.



Fig. 4 Partial pressures (fuel side) versus fuel utilization for the ideal isothermal fuel cell with no diffusion

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Table 1 Fuel side ratios of component molar flows n/n/\* (= V/V/\*) comparing a fuel cell with no dispersion to the classic fuel cell case of no diffusion, for values of fuel utilization x

x	CH <sub>4</sub>	CO	CO <sub>2</sub>	H <sub>2</sub>	H <sub>2</sub> O	O <sub>2</sub>
0.0	1.0001	0.9970	2.1854	1.001	0.7602	0.99
0.1	1.0001	0.9971	1.0381	1.001	0.9919	0.99
0.2	1.0001	0.9973	1.0161	1.001	0.9964	0.99
0.3	1.0001	0.9974	1.0089	1.001	0.9979	0.99
0.4	1.0001	0.9975	1.0055	1.001	0.9987	0.99
0.5	1.0001	0.9976	1.0035	1.001	0.9991	0.99
0.6	1.0001	0.9977	1.0022	1.001	0.9994	0.99
0.7	1.0001	0.9979	1.0013	1.001	0.9996	0.99
0.8	1.0001	0.9980	1.0007	1.001	0.9998	0.99
0.9	1.0001	0.9981	1.0003	1.001	0.9999	0.99

**Lemma.** The following three sets of equations are equivalent, under the assumptions  $P_iV'_i = n'_iRT$  for i = 1, ..., 6, and Eqs. (5a-f)

$$V'_1 = V'_2 = V'_3 = V'_4 = V'_5 = V'_6$$
 (8*d*-*h*)

$$n'_i(P_1 + P_2 + P_3) = P_i \quad i = 1, ..., 6 \quad (13a-f)$$

$$3(P_1 + P_2 + P_3) = 2P_1 + P_4 + P_5$$
  

$$4x(P_1 + P_2 + P_3) = -P_1 + P_3 + P_5 + 2P_6$$
  

$$\epsilon_1(P_1 + P_2 + P_3) = P_2 + P_3$$
  

$$\epsilon_2(P_1 + P_2 + P_3) = P_3$$
  

$$\epsilon_3(P_1 + P_2 + P_3) = P_5 + P_3 - P_1 \qquad (14a-e)$$

Note that Eqs. (14a, b) are the same as the nondispersion Eqs. (12a, b). Thus, the unique solution for the  $P_i$ 's for i = 1, ..., 6 in the nondispersion case is just the  $P_i^*$ 's of the classical case. It is clear that the classical case is just a special case of the nondispersion case, in which the three constraints (14c-e)have been added. In the nondispersion case which is more general, the three  $\epsilon_i$ 's can be chosen arbitrarily and, since the coefficients of the  $d\epsilon_i$ 's in deriving Eq. (7b) were zero, there will be no entropy generation due to diffusion or chemical reaction.

An example is shown in Table 1 where three of the  $n'_i$ 's  $(n'_1, n'_4, \text{ and } n'_6)$  have been chosen to be a little different from the  $n'_i$ \*'s.

*Remarks.* (i) One of Eqs. (13) is redundant. (ii) The physical significance of the form of Eqs. (14c - e) by themselves is not known. (iii) It appears that the concept of dispersion is very general, that Eq. (11) is independent of the number of species and reactions that are assumed to represent the dynamics of the flow. (iv) It is not the case that any three of the  $n_i$ 's can be chosen arbitrarily, only the  $\epsilon_i$ 's,  $i = 1, \ldots, 3$ . For example,  $n_1', n_2'$ , and  $n_3'$  cannot be changed arbitrarily since their sum equals 1.

#### The Isothermal Methane Reformer Case

The setup and equations for a methane reformer is similar to the fuel cell. The fuel input flows are as before, but no reforming has started. There are six reaction products as before, 1–6. There is no air and no power POW, electrical or otherwise. There is no x, but the output state is assumed variable in which the final flow rates  $n_i$ 's and partial pressures  $P_i$ 's for i = 1, ..., 6 can be manipulated. As before, all other sources of entropy generation (temperature gradient, friction, kinetic energies, etc.) have been removed from consideration.

The resulting equations are obtained from the fuel cell case by setting POW = 0 and limiting the summations to the first 6 terms in Eqs. (1), (2), and (3). The entropy generation  $S'_{gen}$ depends only on the input (fixed) and output states and not on the thermodynamic path inside the reformer. Equations (4b) and (5g, h) are dropped. The differentials in Eq. (6) are then thought of as changes in the thermodynamic properties along (differentiable) paths from one possible output to another possible output. Thus, the physical position of the outputs (the exit of the reformer) is not changing.

The modified Eq. (7b) becomes

$$TdS'_{gen} = -\sum_{2}^{6} (V'_{i} - V'_{1})dP_{i}$$
(15)

Suppose now that the output is the unique (classical) case of not only chemical equilibrium, Eqs. (8a-c), but also no diffusion, Eqs. (8d-h). Then the numerical values for the  $n'_i$ 's and  $P_i$ 's, i = 1, ..., 6 can be computed (in fact, they are just  $n'_i$ \*(0) and  $P_i^*(0)$  of the classical fuel cell case), and thus  $S'_{gen} = -\Delta G'/T$  as well. Now, move along a path of output states starting from this equilibrium, always remaining in chemical equilibrium, allowing diffusion, but no dispersion, Eqs. (12a, b) with x = 0. Then the chemical composition of these outputs are unchanging (Lemma with x = 0) as is  $S'_{gen}$ , but the  $n'_i$ 's can vary with three degrees of freedom as in the no-dispersion fuel cell case.

## The Kinetic Part of Diffusion

If the reader is alarmed that the second law of thermodynamics appears to be violated, keep in mind that the terms in Eq. (1) due to kinetic energy were dropped. If included, Eq. (3) would have a positive term  $-\Delta$ (KineticPower) on the RHS, and this would be allocated to  $TS'_{gen}$ . This term, insignificant to begin with, can be made arbitrarily small (on a basis per mole of reactants) compared to any disperse effects by taking the cross sectional area arbitrarily large.

For those who prefer the static environment, the reformer case, for example, can be redone in the following way. Imagine a fixed amount of inputs in a controlled space containing six receptacles (balloons?). Each receptacle is allowed to contain only one chemical specie, to vary in volume, and to carry out chemical reactions with the other five. The sum of the six pressures is assumed to be P.

#### **Future Considerations**

The unusual methodology used here of adding an independent variable x simplifies the analysis, but is one-dimensional in its nature. A three-dimensional analysis should be made using standard (Eulerian) methods to see if a "no-dispersion" concept holds more generally, even in the nonequilibrium thermodynamics setting. This could include nonideal gas properties with nonideal flow. Experimental work could be done to see to what extent the C:H:O ratios vary in the flow of real SOFCs, and whether these numbers can be manipulated by controlled diffusion of additional fuel.

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Thermodynamic properties and graphs are from the Electronic Equation Solver (EES), while the sketches are from CorelDRAW!

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