Minimal dissipation processes and estimates of limiting possibilities of irreversible thermodynamic systems

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Abstract

Irreversible work of separation and irreversible maximal productivity of heat driven separation are derived in this paper.

1 Finite Time thermodynamics

Finite-time thermodynamics have been developed to provide in-principle limits of performance for processes operating within finite intervals or at a nonzero rate. These performance limits are obtained by solving control problems where these performances are optimized subject to given finite average rates/times of its processes.

The minimal work of separation is one of such in-principle limits that is defined as the minimal amount of work, required for separation of a mixture with the given composition. It plays fundamental role similar to the role of Carnot efficiency in science and engineering. The reversible work of separation was obtained in classical thermodynamics. In this paper we obtain the irreversible work of separation for mechanical, heat driven and chemical potential differential driven separation.

It is known that the productivity of an irreversible heat engine is bounded. In this paper we derive similar bound on the productivity of a heat driven separation process.

2 Irreversible separation

Consider the system (Fig. 1) that includes reservoir, finite capacity output subsystem and the working body. Assume that T,P are time independent. The reservoir's k-component vector of concentrations c_0 is time independent and therefore its vector of chemical potentials μ_0 is also time independent. The out-
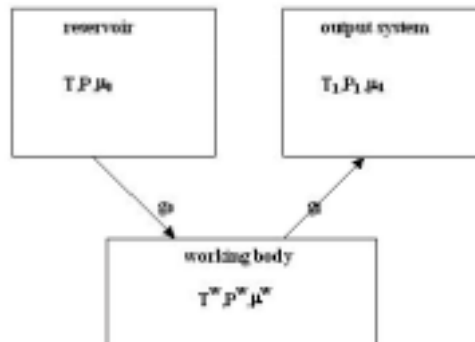

Figure 1: Computational schema for separation system

put subsystem's chemical potential and molar concentration are denoted as $\mu(t)$ and $c(t)$. The working body's chemical potential is denoted as $\mu^w(t)$ and is assumed to be a control variable of the problem. At $t = 0$ the intensive variables of the output subsystem and reservoir are the same. The number of moles in the output subsystem at the initial $N(0) = N_0$ and final $N(\tau)$ time and its compositions $c(0)$ and $c(\tau)$ are given. The mass transfer coefficients between reservoir and the working body and the working body and the output subsystem are finite and fixed. The temperatures and pressures in all subsystems are the same $T = T_1 = T^w$ and $P = P_1 = P^w$. Stodola's formula yields the following expression for the work of separation in isothermal, adiabatic system

$$A = A_0 + T\Delta S. \qquad (1)$$

ΔS here is the entropy production and A_0 is the reversible work of separation that is completely determined by $N(\tau)$, $N(0)$, $c(\tau)$, $c(0)$.

Therefore the problem of minimum work A is re-

duced to the problem of minimal entropy production

$$\Delta S = \frac{1}{T} \int_0^\tau \sum_{i=1}^k [g_{0i}(\mu_{0i} - \mu_{0i}^w) + g_{1i}(\mu_{1i}^w - \mu_{1i})] \, dt =$$

$$= \frac{1}{T} \int_0^\tau \sum_{i=1}^k (g_{i0}\Delta\mu_{0i} + g_{i1}\Delta\mu_{1i}) dt \rightarrow \min \qquad (2)$$

subject to the cyclic conditions

$$\int_0^\tau g_{i0} dt = \int_0^\tau g_{i1} dt \qquad (3)$$

and subject to the conditions of non-negativity of the flows

$$g_{0i} \geq 0, \qquad g_{1i} \geq 0. \qquad (4)$$

The problem of (2)-(4) becomes simpler if the differences of chemical potentials $\Delta\mu_{0i}$ and $\Delta\mu_{1i}$ are unique functions of flows g_{0i} and g_{1i} correspondingly. If the processes are close to equilibrium then these functions are linear.

Assume

$$\Delta\mu_{0i} = \varphi_{0i}(g_{0i}), \qquad \Delta\mu_{1i} = \varphi_{1i}(g_{1i}),$$

then the problem (2)– (4) can be decomposed onto $2k$ problems

$$\Delta S_{ji} = \frac{1}{T} \int_0^\tau \sigma_{ji}(g_{ji}) dt \rightarrow \min \qquad (5)$$

subject to

$$\int_0^\tau g_{ji} dt = \Delta(Nc_i), j = 0, 1, \quad i = 1, 2, ..., k \qquad (6)$$

where $\sigma_{ji} = g_{ji}\varphi_{ji}(g_{ji})$ is the function that determines dissipation.

Problem (5), (6) is the averaged problem of nonlinear programming. Its optimal solution g_{ji}^* [3] is either time independent and equal to

$$g_{ji}^* = g_{1i}^* = \frac{\Delta(Nc_i)}{\tau}, \qquad (7)$$

or switches over the interval $(0, \tau)$ between two so-called basic values. The latter corresponding to the problem where the convex envelope of the function $\sigma_{ji}(g_{ji})$ is lower than the value of this function at g_{ji}^*. Characteristic forms of the function $\sigma_{ji}(g_{ji})$ for the time-independent and for switching regimes are shown in Fig. 2. If the function σ_{ij} is concave then the optimal rate g_{ji} is always constant. Let us calculate the second derivative of σ on g (we omit subscripts for simplicity). If it is positive then the constancy of rate in the optimal process is guaranteed.

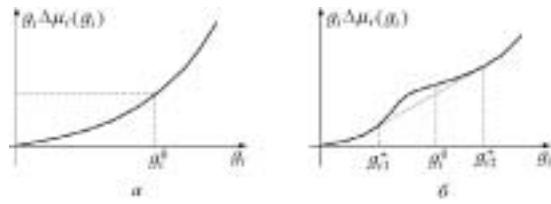$$\sigma''(g) = 2\varphi'(g) + g\varphi''(g) \geq 0. \qquad (8)$$

Figure 2: The dependences of the entropy production on the rate for the constant (a) and switching (b) solutions (g_{i1}^* and g_{i2}^* are the basic values of the rate)

The first term in this expression is always positive because chemical potentials' difference is the driving force of mass transfer and monotonically depends on the flow. For the majority of laws of mass transfer the inequality (8) holds. In particularly, it holds if the flow of mass transfer is proportional to the difference of chemical potentials at any degree $l > 0$. Consider mass transfer flow that depends linearly on the chemical potential difference for all i, j. Then

$$g_{ji} = \alpha_{ji}\Delta\mu_{ji} \Rightarrow \varphi_{ji} = \frac{g_{ji}}{\alpha_{ji}}. \qquad (9)$$

It is clear that the conditions (8) hold and the optimal rates of flows obey equalities (7). Equalities (7) hold for any non-switching solution. The minimal entropy production here is

$$\Delta S^{\min} = \sum_{i,j} \Delta S_{ji}^{\min} = \frac{\tau}{T} \sum_{i,j} \sigma_{ji} \left(\frac{\Delta(Nc_i)}{\tau} \right) \qquad (10)$$

and the minimal work of separation is

$$A_{\min} = A_0 + \tau \sum_{i,j} \sigma_{ji} \left(\frac{\Delta(Nc_i)}{\tau} \right). \qquad (11)$$

The optimal rates are determined by the initial and final states, which allows us to specify the estimate (11).
Onsanger's linear kinetics (9) holds for near equilibrium flows and from (11) it follows here that

$$A_{\min} = A_0 + \frac{1}{\tau} \sum_{i=1}^n \frac{\Delta^2(Nc_i)}{\overline{\alpha_i}}, \qquad (12)$$

where

$$\overline{\alpha_i} = \frac{\alpha_{0i}\alpha_{1i}}{\alpha_{0i} + \alpha_{1i}} \qquad (13)$$

is the equivalent mass transfer coefficient on the i-th component and the minimal entropy production is

$$\sigma_{min} = \frac{1}{T\tau^2} \sum_{i=1}^n \frac{\Delta^2(Nc_i)}{\overline{\alpha_i}}. \qquad (14)$$

The average power of separation is

$$p_{\min} = \frac{A_{\min}}{\tau} = \frac{A_0}{\tau} + \frac{1}{\tau^2} \sum_{i=1}^n \frac{\Delta^2(Nc_i)}{\overline{\alpha_i}}. \qquad (15)$$

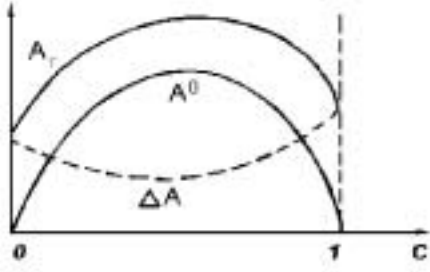**Figure 4:** Separation system with m finite capacity subsystems

Figure 3: The reversible (A^0) and irreversible (A) work of separation of binary mixture as functions of key component's concentration c

$p_0 = \frac{A_0}{\tau}$ is the reversible power of separation.
If

$$N(0) = 0, \quad \Delta(Nc_i) = Nc_i(\tau)$$

then the expressions (12), (15) take the form

$$A_{\min} = A_0 + \frac{N^2}{\tau} \sum_{i=1}^{n} \frac{c_i^2(\tau)}{\overline{\alpha_i}}, \qquad (16)$$

$$p_{\min} = p_0 + g^2 \sum_{i=1}^{n} \frac{c_i^2(\tau)}{\overline{\alpha_i}}, \qquad (17)$$

where

$$2A_0 = NRT \sum_{i=1}^{n} [c_i(\tau) \ln c_i(\tau) - c_i \ln c_i]. \qquad (18)$$

Note that the irreversible estimate of the work of separation (16) does not tend to zero for poor mixtures when concentration of one of the components tend to one (Fig. 3).

If the system includes not one but a number of output subsystems then it is clear that the minimal work of separation is the sum of the minimal works for each subsystem. For j –th subsystem we get

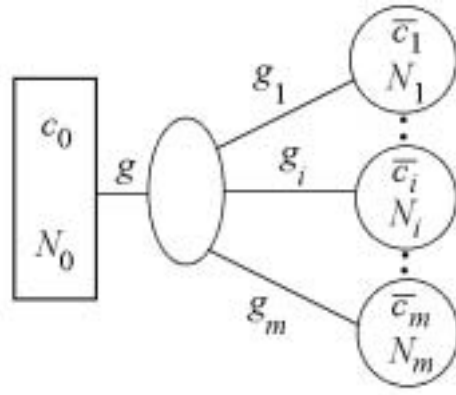$$A_{min} = \sum_{j=1}^{k} A_{min}^{j}, \quad p_{min} = \sum_{j=1}^{k} p_{min}^{j}. \qquad (19)$$

Separation system with m finite capacity subsystems.

Consider a system that is shown in Fig. 4. Its initial state is described by the vector of concentrations c_0, the number of moles of the mixture N_0, and its final state by the number of moles $\overline{N}_j (j = 1..., m)$ in each of the subsystems and their concentrations $\overline{c}_j$. The mass balances must hold here

$$\sum_{j=1}^{m} \overline{N}_j = N_0, \qquad (20)$$

$$\sum_{j=1}^{m} \overline{N}_j \overline{c}_{ji} = N_0 c_{0i}, \quad i = 1, 2, ..., k.$$

The reversible separation of separation here is

$$A_r^0(c_0, \overline{c}) = RT(\sum_{j=1}^{m} N_j \sum_i \overline{c}_{ji} \ln \overline{c}_{ji} - N_0 \sum_i c_{0i} \ln c_{0i}) = $$

$$= A_{r0}^0(c_0, N_0) - \sum_{j=1}^{m} A_{rj}^0(\overline{c}_j, N_j). \qquad (21)$$

The reversible work of separation is equal to the difference of the reversible work of separation for the initial mixture into pure components and this work of separation for the mixture in each of the output subsystems.

We again assume that flows g_j have components g_{ij} proportional to the difference of chemical potential of subsystem and the working body with the coefficient α_{ij}. Here the condition of minimal work of separation corresponds to the condition of flows' constancy

$$g_{ij} = \frac{\overline{N}_j \overline{c}_{ij}}{\tau}, \quad i = 1, 2, \ldots, k, \quad j = 1, \ldots, m, \quad (22)$$

$$\Delta \mu_{ij} = \frac{g_{ij}}{\overline{\alpha}_{ij}}, \quad j = 0, 1, 2, \ldots, m. \qquad (23)$$

Here $\overline{\alpha}_{ij}$ is equivalent mass transfer coefficient calculated using (13), the flow in the j –th output subsystem on the i –th component.

Similarly as it was done above for the system with the reservoir and one finite capacity output subsystem and flows proportional to the final concentrations (22), these concentrations in the output subsystems are time independent and equal to $\overline{c}_j$ correspondingly, and the number of moles $N_j(t)$ depends linearly on time. The power p here is constant

$$p = \frac{RT}{\tau} \sum_{j=1}^{m} \overline{N}_j \sum_i \overline{c}_{ij} \ln \frac{\overline{c}_{ij}}{c_{i0}} + \frac{1}{\tau^2} \sum_{j=1}^{m} N_j^2 \sum_i \overline{c}_{ij}^2 / \overline{\alpha}_{ij}. \tag{24}$$

The minimal work of separation for the mixture with concentrations c_0 into m subsystems with concentrations $\overline{c}_i$ over the time τ is

$$A_r = RT N_0 \sum_{j=1}^{m} \gamma_j \sum_i \overline{c}_{ij} \ln \frac{c_{ij}}{c_{i0}} + \frac{N_0^2}{\tau} \sum_{j=1}^{m} \gamma_j^2 \sum_i \overline{c}_{ij}^2 / \overline{\alpha}_{ij}. \tag{25}$$

Here $\gamma_j = \overline{N}_j / N_0$, $\overline{\alpha}_{ij} = \frac{\alpha_{ij}\alpha_{i0}}{\alpha_{i0}+\alpha_{ij}}$.

The first term here coincides with the reversible work of separation A_r^0 of the mixture of N moles with concentration c_0 into subsystems with number of moles N_j and concentrations $\overline{c}_j$. The second term takes into account irreversibility of the process. A_r decreases monotonically and tends to A_r^0 when process duration τ and mass transfer coefficient $\overline{\alpha}_{ij}$ increases.

Example:

Consider separation of the binary mixture into pure components in the time τ. In this case $N_1 = c_0 N_0$, $N_2 = (1 - c_0)N_0$, where c_0 is the concentration of key component, $\overline{c}_{11} = \overline{c}_{22} = 1$. From the formula (25) we get

$$A_r = -RT N_0 (c_0 \ln c_0 + (1 - c_0) \ln(1 - c_0)) + \tag{26}$$

$$+ \frac{N_0^2}{\tau} \left(\frac{c_0^2}{\overline{\alpha}_{11}} + \frac{(1 - c_0)^2}{\overline{\alpha}_{22}} \right) =$$

$$= A_r^0(c_0) + \frac{N_0^2}{\tau} \left(\frac{c_0^2}{\overline{\alpha}_{11}} + \frac{(1 - c_0)^2}{\overline{\alpha}_{22}} \right)$$

The estimate (26) derived in [8] by solving the problem of optimal separation of the binary mixture in the given time τ in Van-Hoff's thought experiment with movable pistons and semitransparent membrane where $\overline{\alpha}_{11}$ and $\overline{\alpha}_{22}$ are the permeability coefficients on the first and second component. If flows do not depend explicitly on the chemical potentials' differentials, e.g. are proportional to the concentrations' differential, then estimate similar to the obtained above can be constructed by solving the following auxiliary nonlinear programming problem
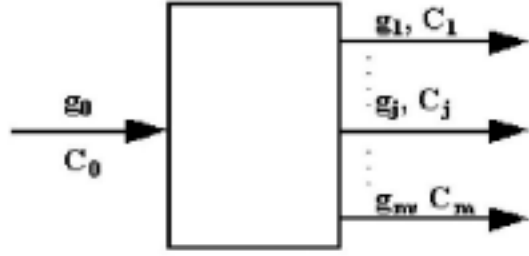

Figure 5: The schema of continuous separation system

$$\Delta\mu_i(P_0^i, P_i) \rightarrow \min_{P_0^i, P_i} \Big/ g_i(P_0^i, P_i) = g_i, \quad i = 1, 2, ... \tag{27}$$

Here (P_0^i, P_i) are partial pressures of the components in contacting subsystems that depend on the chemical potentials' differentials $\Delta\mu_i$. The flow g_i depends on the same differentials. Minimums in these problems are sought for different values of constant $g_i > 0$ and non-positive P_0^i and P_i. We denote the minimal values of the objective in each of these problems $\Delta\mu_i^{\min}(g_i)$ as $\Delta\mu_i^*(g_i)$. This dependence can be used in the estimate (5) of the irreversible work of separation.

Example.

Assume $\Delta\mu = RT \ln(P_0/P)$, $g(P_0, P) = (P_0 - P)/\alpha$, and $0 < P < P_{\max}$. Let us express P_0 in terms of g and P:

$$P_{0i} = \alpha_i g_i + P_i, \quad i = 1, 2.$$

For each g $\Delta\mu = RT \ln(\alpha g/P + 1)$ Attains its minimum for $P = = P_{\max}$, therefore $\Delta\mu_i^*(g_i) = RT \ln(\alpha_i g_i / P_{i\max} + 1)$.

Consider continuous separation system with the input flow with concentration c_0 and m output flows $g_j (j = 1, ..., m)$ with concentrations $c_j = \{c_{ji}\}$ (Fig. 5). Here the temperatures on the input and output flows are assumed to be close to each other.

Equation (24) allows us to estimate the minimal power required for continuous separation in such system

$$p_{\min} = \sum_{j=1}^{m} p_{0j} + g_0^2 \sum_{j=1}^{m} \gamma_j^2 \sum_{i=1}^{n} \frac{c_{ji}^2}{\alpha_{ji}}, \tag{28}$$

where

$$\gamma_j = \frac{g_j}{g_0} \geq 0, \quad \sum_{j=1}^{m} \gamma_j = 1, \tag{29}$$

$$p_{0j} = g_0\gamma_j RT \sum_{i=1}^{n}[c_{ji}\ln c_{ji} - c_{0i}\ln c_{0i}] = \gamma_j M_j(g_0, c_j).$$
$$(30)$$

From mass balance equations

$$\sum_{j=1}^{m}\gamma_j c_{ji} = c_{0i}, \quad i = 1, ..., k-1, \qquad (31)$$

$$\sum_{i=1}^{n}c_{ji} = 1, \quad j = 0, ..., m.$$

The number of conditions (31) is $k-1$, because the concentration of one of the components is determined by the conditions (29).

If the number of flows $m > k$, and their compositions are given, then the removal fractions can be chosen in such a way that the power of separation is minimal subject to constraints (29), (31). The Lagrange function of this problem is

$$L = \sum_{j=1}^{m}\left\{\gamma_j M_j + \gamma_j^2 r_j - \lambda_0\gamma_j - \sum_{i=1}^{k-1}\lambda_i\gamma_j c_{ji}\right\}. \quad (32)$$

here

$$r_j(g_0, c_j) = g_0^2 \sum_{i=1}^{k}\frac{c_{ji}}{\alpha_{ji}}.$$

L is concave function on γ_j, and its conditions of stationarity determine the flows that minimize the power for separation for given flows' compositions

$$\gamma_j^* = \frac{\lambda_0 - M_j + \sum_{i=1}^{k-1}\lambda_i c_{ji}}{2g_0^2, r_j}, \quad j = 1, ..., m. \quad (33)$$

We have k linear equations for λ_0 and λ_i

$$\frac{1}{2g_0^2}\left[\sum_{j=1}^{m}\frac{\lambda_0 - M_j}{r_j}\right. \qquad (34)$$

$$\left. + \sum_{i=1}^{n-1}\lambda_i \sum_{j=1}^{m}\frac{c_{ji}}{r_j}\right] = 1,$$

$$\frac{1}{2g_0^2}\left[\sum_{j=1}^{m}c_{ji}\left(\frac{\lambda_0 - M_j}{r_j} + \frac{1}{r_j}\sum_{i=1}^{n-1}\lambda_i c_{ji}\right)\right] = c_{0i},$$
$$(35)$$

$$i = 1, ..., k-1.$$

Example: Assume $m = 3$, $k = 2$, $g_0 = 1\frac{mol}{s}$, $c_{01} = c_{02} = 0.5$; $c_{11} = 0.9$; $c_{12} = 0.1$; $\overline{\alpha}_{11} = \overline{\alpha}_{12} = 0.04\frac{mol^2}{Js}$; $c_{21} = 0.3$; $c_{22} = 0.7$; $\overline{\alpha}_{21} = \overline{\alpha}_{22} = 0.01\frac{mol^2}{Js}$; $c_{31} = 0.1$; $c_{32} = 0.9$; $\overline{\alpha}_{31} = \overline{\alpha}_{32} = 0.06\frac{mol^2}{Js}$.

From (30) we get $M_1 = 1232$, $M_2 = 1000$, $M_3 = 918.026$, and $r_1 = 25$, $r_2 = 100$, $r_3 = 16.667$.

Equations (34), (35) for λ-multipliers take the form

$$\frac{1}{2}\left[\frac{\lambda_0 - M_1}{r_1} + \frac{\lambda_0 - M_2}{r_2} + \frac{\lambda_0 - M_3}{r_3}+\right.$$

$$\left. +\lambda_1\left(\frac{c_{11}}{r_1} + \frac{c_{21}}{r_2} + \frac{c_{31}}{r_3}\right)\right] = 1,$$

$$\frac{1}{2}\left[c_{11}\left(\frac{\lambda_0 - M_1}{r_1} + \frac{\lambda_1 c_{11}}{r_1}\right)+\right.$$

$$+c_{21}\left(\frac{\lambda_0 - M_2}{r_2} + \frac{\lambda_1 c_{21}}{r_2}\right)+$$

$$\left. +c_{31}\left(\frac{\lambda_0 - M_3}{r_3} + \frac{\lambda_1 c_{31}}{r_3}\right)\right] = c_{01}.$$

We get $\lambda_0 = 829.605$, $\lambda_1 = 403.98$. Their substitution in (33), yields $\gamma_1^* = 0.483$, $\gamma_2^* = 0.068$, $\gamma_3^* = 0.449$ and the corresponding estimate for minimal irreversible power of separation (28) is $p_{\min} = 1081\frac{J}{s}$.

3 Limiting productivity and minimal heat consumption for heat driven separation

In many separation processes heat engine is used to create differential of chemical potential between the working body and the reservoirs (driving force of mass transfer). Here the working body is heated during contact with one reservoir and is cooled during contact with the other reservoir. One can represent heat driven separation system as a transformer of heat into the work of separation that generates power p, consumes heat flow from hot reservoir g_+ and rejects flow g_- to the cold reservoir. Heat transfer coefficients for contacts with the hot and cold reservoir α_+ and α_- are fixed.

It was shown in [5], [6], that the potential of direct transformation of heat to work are limited and the maximal generated power for the working body with the distributed parameters is

$$p_{\max} = \overline{\alpha}(\sqrt{T_+} - \sqrt{T_-})^2. \qquad (36)$$

In this expression $\overline{\alpha} = \frac{\alpha_+\alpha_-}{\alpha_+ + \alpha_-}$ is the equivalent heat transfer coefficient for continuous contact with the reservoirs is $\overline{\alpha} = \frac{\alpha_+\alpha_-}{(\sqrt{\alpha_+}+\sqrt{\alpha_-})^2}$ for sequential contact. The maximal power determines the heat flow consumed from the hot reservoir. Further increase of heat consumption for given values of heat transfer coefficients requires increase of temperature differential between reservoirs and the working body and reduces the power.

The dependence of the used power on the productivity of irreversible separation processes is monotonic (28). Therefore, the limiting productivity of heat driven separation processes corresponds to the maximal-possible power produced by transformation

of heat into work. Further increases of heat consumption q_+ reduces power, and therefore reduces the productivity of separation process.

For the Newton (linear) law of mass transfer and heat-work transformer the dependence of the power on the heat used [7] is

$$q^+(p) = \frac{p}{\eta_p} = \frac{2p}{\left(\frac{p}{\overline{\alpha}T_+} + \eta_k\right) + \sqrt{\left(\frac{p}{\overline{\alpha}T_+} + \eta_k\right)^2 - \frac{4p}{\overline{\alpha}T_+}}}.$$

(37)

here $\eta_k = \frac{T_+ - T_-}{T_+}$ is the Carnot efficiency, T_+ and T_- are the hot and cold reservoir's temperatures and $\overline{\alpha} = \frac{\alpha_+ \alpha_-}{\alpha_+ + \alpha_-}$ is the equivalent heat transfer coefficient.

The minimal heat consumption q_+ as a function of productivity g_0 for heat driven separation can be obtained substituting p with the expression (37) in the right-hand side of (28). The result holds for $0 \leq p \leq p_{\max}$, and therefore for $0 \leq g_0 \leq g_{0\,\max}$. The duration here must not exceed the maximal possible duration.

Substitution of the right-hand side of (36) instead of p in (28) yield the maximal possible productivity of the system (where $\overline{\alpha}$ is chosen according to the type of contact between transformer and reservoir). We denote

$$B = RT \sum_j \gamma_j \sum_i c_{ij} \ln \frac{c_{ij}}{c_{i0}}, \quad D = \sum_j \gamma_j^2 \sum_i \frac{c_{ij}^2}{\alpha_{ij}}.$$

(38)

We obtain

$$p_{\max} = \overline{\alpha}\left(\sqrt{T_+} - \sqrt{T_-}\right)^2 = Bg_{0\,\max} + Dg_{0\,\max}^2,$$

and the limiting productivity is

$$g_{0\,\max} = \frac{-B + \sqrt{B^2 + 4\overline{7}\alpha D(\sqrt{T_+} - \sqrt{T_-})^2}}{2D}$$

(39)

Formulas (38), (39) allow us to estimate the limiting productivity of heat driven separation process for Newton's laws of heat transfer between the working body and reservoirs and mass transfer proportional to the differentials in chemical potentials (mass transfer is close to isothermal with the temperature T).

4 Conclusions

Irreversible work of separation obtained in this paper differs from reversible ones not only quantitatively but also qualitatively. The reversible work of separation for such mixtures tends to zero. For poor mixtures with the concentration of one of the components close to one this work tends to a finite non-zero limit, which depends on the kinetics factors. This results are in qualitative agreement with experimentally observable amounts of energy required for separation of poor mixtures differ by the factor of 10^5 [2].

For heat driven separation processes the novel results obtained in this paper include the estimate of minimal heat consumption as a function of kinetic factors and the thermodynamic limit on the productivity of heat driven separation.

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