α = thickness/diameter for disk
ρ = fluid density
ρ_s = solid density
λ = heat of fusion
ϕ = diffusion factor in Equation (6)

Subscripts

i = interface
b = bulk
f, b = freezing point of bulk solution
f, i = freezing point of solution at interface

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On a Conjecture of Aris: Proof and Remarks

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In this paper a conjecture of Aris is proved to be correct, that of all catalyst particles of fixed volume for isothermal first-order chemical reactions the spherical particle has the lowest effectiveness factor. The method of proof uses a process of symmetrization. Some conjectures are also made about other chemical reactions. The method of proof is also valid for two-dimensional domain (infinite cylinder).

The purpose of this paper is to explore the effect of particle shape on effectiveness factors and to point out some features of the problem which to the authors' knowledge have not been dealt with, and in particular to investigate certain extremal properties related to catalyst shape. In retrospect it seems that the results are obvious, but they did come as a surprise first. To review the situation briefly, effectiveness factor plots which appear in textbooks (*1*, *3*) usually are concerned with spheres, infinite cylinders, and infinite slabs. If for a first-order isothermal reaction the effectiveness factor η is plotted against ϕ_T, the Thiele parameter, where

$$\phi_T = R\sqrt{\frac{k}{D}}$$

and where R is the radius of the sphere or the cylinder or the half thickness of a slab, then for the same value of ϕ_T the effectiveness factor for the sphere is greater than that for the cylinder and that for the latter is greater than that for the slab. It has also been shown by Aris (*2*) that the curves may be brought together asymptotically by defining a modified Thiele parameter

$$\phi_A = \frac{V_p}{S}\sqrt{\frac{k}{D}}$$

and, furthermore, that η as a function of ϕ_A may be represented for the three shapes mentioned above with rea-

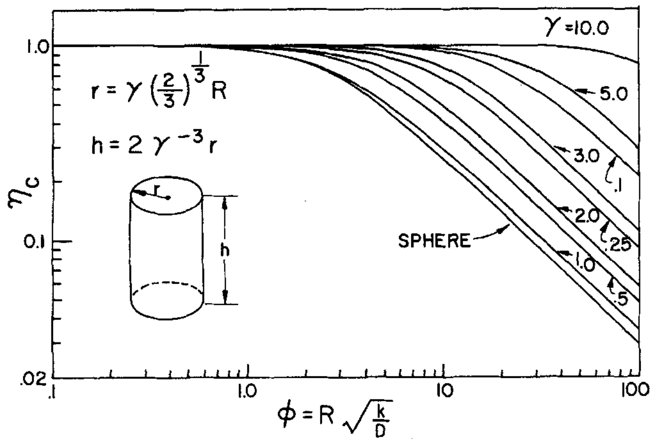

Fig. 1. Plot of effectiveness factor η_c for a cylinder vs. ϕ where R is the radius of a sphere having the same volume. (Note that this is not ϕ_c of the text.) $\gamma = 1$ is for the minimum cylinder.

sonable accuracy by a single curve. Aris in the same paper also gave a formula for the effectiveness factor for a finite cylinder, although to the authors' knowledge there are no computations in the literature of effectiveness factors for finite bodies other than spheres. In order to examine the problem in more detail let us consider a simply connected catalyst particle of fixed volume V but with, for the moment, an arbitrary shape and corresponding superficial external surface S and suppose a single chemical reaction $\sum_{i=1}^{n} A_i a_i = 0$ takes place in the pores. Then if f_i is the reaction rate based on A_i, the following equations result for the steady state:

$$\text{in } V \begin{cases} D_i \nabla^2 A_i + \rho_s S_g f_i = 0 \quad , \quad i = 1, 2, \ldots, n \\ k_T \nabla^2 T + (-\Delta H_i)\, \rho_s S_g f_i = 0 \end{cases}$$

If it is assumed that the surface concentrations and temperature are constant, then

$$\text{on } S \begin{cases} A_i = A_{is} \; , \quad i = 1, 2, \ldots, n \\ T = T_s \end{cases}$$

Since $a_i^{-1} f_i = f$, the intrinsic rate, we obtain

$$\text{in } V \begin{cases} D_i \nabla^2 A_i + \rho_s S_g a_i f = 0 \quad , \quad i = 1, 2, \ldots, n \\ k_T \nabla^2 T + (-\Delta H)\, \rho_s S_g f = 0 \end{cases}$$

where ΔH corresponds to the intrinsic rate.

Appropriate manipulation of these equations produces the equations

$$\nabla^2 \omega_{ij} = 0 \qquad \text{in } V$$

$$\omega_{ij} = \omega_{ijs} \qquad \text{on } S$$

and

$$\nabla^2 \nu_i = 0 \qquad \text{in } V$$

$$\nu_i = \nu_{is} \qquad \text{on } S$$

where

$$\omega_{ij} = \frac{D_i A_i}{a_i} - \frac{D_j A_j}{a_j}$$

$$\nu_i = \frac{D_i A_i}{a_i} - \frac{k_T T}{(-\Delta H)}$$

with the obvious definition for ω_{ijs} and ν_{is}. From the maximum principle the solution to these equations is obvious:

$$\omega_{ij} = \omega_{ijs} \qquad \text{in } V$$

$$\nu_i = \nu_{is} \qquad \text{in } V$$

It follows that if the particle is isothermal, that is, temperature gradients within the particle are negligible, any species may be chosen as dependent variable, say $A_i = u$ and

$$D \nabla^2 u + F(u) = 0 \qquad \text{in } V$$

$$u = u_s \qquad \text{on } S$$

where $F(u)$ depends on the surface concentration of all species and the surface temperature. In case the heat effects within the particle are not negligible, one equation is sufficient also:

$$k_T \nabla^2 T + G(T) = 0 \qquad \text{in } V$$

$$T = T_s \qquad \text{on } S$$

From the above relations it is apparent that

$$\frac{D_i}{a_i} \iint_S \text{grad } A_i \cdot \mathbf{n}\, dS = \frac{D_j}{a_j} \iint_S \text{grad } A_j \cdot \mathbf{n}\, ds = \frac{k_T}{-\Delta H} \iint_S \text{grad } T \cdot \mathbf{n}\, ds$$

where the integrals are over the surface and $\mathbf{n}$ is the outward normal to S. The effectiveness factor is then defined to be

$$\eta = \frac{-D_j}{\rho_s S_g V f_{js}} \iint_S \text{grad } A_j \cdot \mathbf{n}\, dS = \frac{-D_j}{\rho_s S_g V f_{js}} \iint_S \frac{\partial A_j}{\partial n}\, dS$$

where f_{js} is the rate of production of A_j evaluated at the surface conditions.

Now it is clear that the value of η depends on a number of parameters explicitly. It also depends on the shape of the particle, but this dependence is less clear since calculations have only been made for one real shape, the sphere, and two idealized shapes, the infinite cylinder and the infinite slab; even here the comparisons have not been between shapes having the same volume but rather with shapes having *one* dimension the same. The question naturally arises as to that shape which will produce a maximum effectiveness factor for a fixed volume of catalyst. Clearly the infinite cylinder and slab must be excluded since the physical and mathematical problems make no sense in the context of a fixed volume.

EXPLORATION OF EXTREMA FOR A SIMPLE CASE

In this section we will consider the simple isothermal first-order reaction $A \rightarrow B$. The effectiveness factor for a particle of arbitrary shape is

$$\eta = \frac{D}{\rho_s S_g V k_s A_s} \iint_S \text{grad A} \cdot \mathbf{n}\, dS$$

and we ask whether there is a shape which will maximize η. Now from the divergence theorem

$$D \iint_S \text{grad A} \cdot \mathbf{n}\, dS = D \iiint_V \nabla^2 A\, dV = k \iiint_V A\, dV$$

where the latter follows from the equation $D \nabla^2 A - kA = 0$ in V. Hence, if the effectiveness factor is to be a maximum, the particle volume must be distributed so that the integral of the concentration throughout the volume is a maximum. It is intuitively clear that the particle must be snowflakelike (without holes) or like a spoked wheel without a rim since it is only for thin sections that the average concentration will be a maximum. One can obtain easily the average concentration for a thin slab of half width a as

$$\overline{A} = \overline{A}_s \frac{\tanh \sqrt{\frac{k}{D}}\, a}{\sqrt{\frac{k}{D}}\, a}$$

and this is monotonically decreasing with a. Hence a body containing thin sections will have a greater effectiveness factor than any other with the same volume. This creates an interesting problem, for the sphere has the thickest section possible, and therefore it seems that of all bodies of the same volume the sphere should have the least effectiveness factor. This perhaps would have been obvious had effectiveness factors for any other closed shape been calculated.

Before proceeding we will consider effectiveness factors for finite cylinders and rectangular parallelopipeds of various aspect ratios. These may be obtained by a straightforward solution of the appropriate partial differential equations to be

$$\eta_c(\text{cylinder}) = 1 - \frac{32\phi_c^2}{\pi^2} \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \frac{1}{(2m+1)^2 \lambda_n [\lambda_n + \alpha^2 (2m+1)^2 \pi^2 + \phi_c^2]}$$

$$\eta_r(\text{rect.}) = 1 - \frac{512\phi_r^2}{\pi^8} \sum_{m=1}^{\infty} \sum_{n=1}^{\infty} \sum_{p=1}^{\infty} \frac{1}{(2m+1)^2 (2n+1)^2 (2p+1)^2} \times \frac{1}{(2m+1)^2 + \beta^2 (2n+1)^2 + \delta^2 (2p+1)^2 + \frac{\phi_r^2}{\pi^2}}$$

where λ_n is determined from $J_o(\sqrt{\lambda_n}) = 0$, and α, β, δ are the appropriate aspect ratios. These effectiveness factors are plotted in Figures 1, 2, and 3. In Figure 1, η_c is plotted vs. ϕ for different values of the ratio $2r/h = \gamma^3$, where r the radius of the cylinder is related to a sphere having the same volume by the relation $r = \gamma R^3 \sqrt{2}/3$. Thus at a given value of the parameter ϕ the corresponding η_c's are all for the same volume of particle. The sphere is the lowest curve, and $\gamma = 1$ corresponds to the cylinder

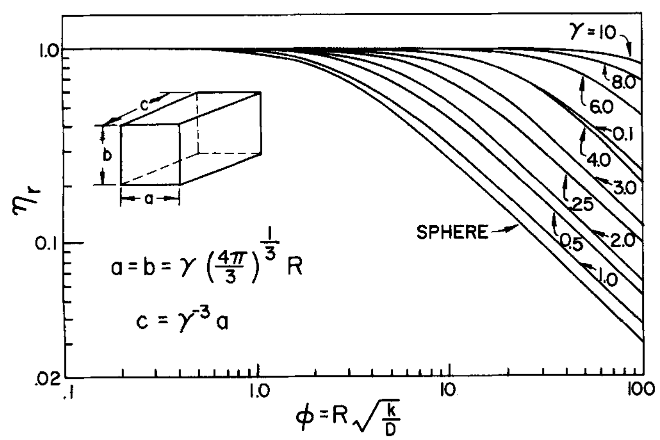

Fig. 2. Plot of effectiveness factor η_r for a rectangular parallelepiped having a square face vs. ϕ where R is the radius of a sphere having the same volume. (Note this is not ϕ_r of the text.) $\gamma = 1$ is for a cube.

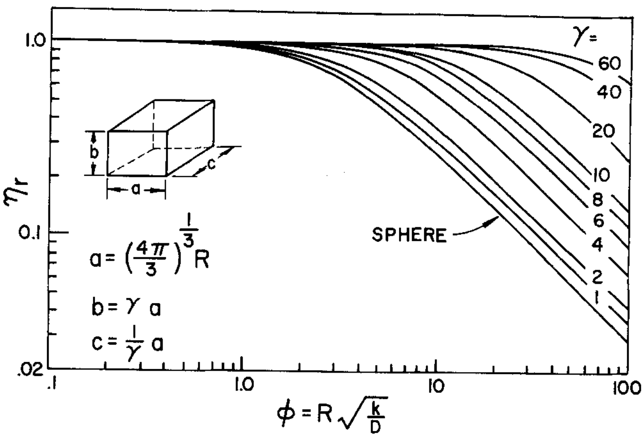

Fig. 3. Plot of effectiveness factor η_r for a rectangular parallelepiped having one fixed dimension (the radius of a sphere having the same volume) vs. ϕ where R is a sphere having the same volume. (Note this is not ϕ_r of the text.) $\gamma = 1$ is for a cube.

with maximum surface for a fixed volume. The effectiveness factors then tend to increase as the cylinder gets flatter or longer. Figures 2 and 3 are similar except for particles of rectangular shape. Figure 2 is drawn for the case where a cross section is always a square, and hence the particles approach flat slabs in one limit and long square rectangular parallelopipeds at the other. Again the sphere has the lowest effectiveness factor and the two limiting shapes the highest. Figure 3 is a similar plot in which one dimension of the parallelopiped is held fixed at the radius of the equivalent sphere and the other two dimensions are allowed to vary but at fixed volume.

In the appendix of this work the following theorem is proved.

Theorem: Given a simply connected region B bounded by a surface Σ and with a fixed volume V. Suppose u is the solution of

$$\nabla^2 u = \lambda u \quad \text{in } B \tag{1}$$

$$u = 1 \quad \text{on } \Sigma \tag{2}$$

then the quantity

$$\iint_{\Sigma} \frac{\partial u}{\partial n} dS$$

has a minimum value for the sphere (in three dimensions and the circle in two dimensions).

In Equation (1) $\lambda = k/D$, and the problem has been normalized so that the surface concentration is unity. We will assume that λ is a constant and physically it is always greater than zero. The analysis which follows permits λ to be a function of position but always positive. The proof of this theorem is accomplished by a mathematical process known as *symmetrization* invented by Jacob Steiner in 1836 and generalized and elaborated on by many (see reference 4). The theorem above is similar to others but not identical and will be proved in the appendix through a series of lemmas.

Hence it follows that the sphere for a simple irreversible isothermal chemical reaction has the least effectiveness factor of all three dimensional simply connected domains of fixed volume. For cylindrical catalyst particles of long axis the one with circular cross section would have a smaller effectiveness factor than any other cylinder of the same cross-sectional area since the theorem above may be modified for two-dimensional domain. For the reversible reaction first order in each direction and isothermal, that is

$$A_1 \underset{k_2}{\overset{k_1}{\rightleftharpoons}} A_2$$

the appropriate equations are

$$\nabla^2 u - \left(\frac{k_1}{D_{A1}} + \frac{k_2}{D_{A2}}\right) u = -k_2 \left[\frac{1}{D_{A1}} \frac{A_{2s}}{A_{1s}} + \frac{1}{D_{A2}}\right] \quad \text{in } B$$

$$u = 1 \quad , \quad \text{on } \Sigma$$

where $u = A_1/A_{1s}$ or

$$\nabla^2 u - \lambda u = -q \quad \text{in } B$$

$$u = 1 \quad \text{on } \Sigma$$

with

$$\lambda = \frac{k_1}{D_{A1}} + \frac{k_2}{D_{A2}}$$

$$q = \frac{k_2}{D_{A1}} \frac{A_{2s}}{A_{1s}} + \frac{k_2}{D_{A2}}$$

Now if $k_1 > k_2$ and $A_{1s} \geqq A_{2s}$, then this problem reduces to

$$\nabla^2 w = \lambda w \quad \text{in } B$$

$$w = 1 \quad \text{on } \Sigma$$

where

$$\frac{u - q/\lambda}{u - q/\lambda} = w$$

and $q/\lambda < 1$. It follows that for this reaction the same analysis is valid so that the effectiveness factor for the sphere and circular cylinder are the least for all particles of fixed volume.

SOME CONJECTURES ON PARTICLE SHAPE

What has been shown above for simple, first-order isothermal reactions probably holds for single, isothermal, one-step reactions in general for the difficulty resides in the fact that there is not sufficient superficial area for reactant to enter the particle. It seems apparent also that for endothermic reactions the spherical shape might not be desirable since the transport of heat into the particle is inhibited by the shape. For a single exothermic reaction the spherical shape might be the best since in this case the temperature difference between the center and the surface would be the largest and the greatest enhancement of the reaction rate would result, if we assume that equilibrium considerations do not come into play. In the case of simultaneous and consecutive reactions the situation will certainly become more complicated. For the system $A \rightarrow B \rightarrow C$, where the first reaction is exothermic and B is the desired product, the spherical shape is probably the poorest since the temperature difference, being the greatest, might activate the second reaction thereby reducing the yield of B.

It would be interesting and it is important to solve these problems in a general way, but this does not seem to be possible except by extensive computations.

NOTATION

- a = half width of an infinite slab
- A = chemical species and its concentration
- A = i^{th} chemical species
- a_i = stoichiometric coefficient of A_i
- B = domain of catalyst particle
- D = diffusion coefficient
- D_i = diffusion coefficient
- D_{A_1}, D_{A_2} = diffusion coefficients of A_1 and A_2
- f = intrinsic reaction rate = $f_i\, a_i^{-1}$
- f_i = reaction rate, moles of A_i formed per unit time percent area
- f_{is} = reaction rate at the particle surface
- G = grad u
- k = first-order reaction velocity constant (per unit of volume)
- k_1 = reaction rate for A_1 (per unit of volume)
- k_2 = reaction rate for A_2 (per unit of volume)
- k_T = thermal conductivity
- $\mathbf{n}$ = unit vector normal to Σ (outward)
- n = distance along outward normal
- O = spherical domain
- $P(\rho)$ = function defined in text
- r = radius of cylinder
- R = radius of sphere or equivalent sphere
- S = surface area of B
- S_g = surface area per gram of catalyst
- T = temperature
- T_s = surface temperature
- u = dimensionless concentration
- V, V_p = volume of particles
- $V(\rho)$ = volume inside $B(\rho)$

Greek Letters

- α = r/h ratio of radius to height of cylinder
- β = a/b ratio of two sides of rectangular parallelepiped
- γ = cube root of $2r/h$
- δ = a/c ratio of two sides of rectangular parallelepiped
- η = effectiveness factor
- η_c = effectiveness factor for a cylinder
- η_r = effectiveness factor for a rectangular parallelepiped
- λ = k/D
- ϕ_c = Thiele parameter for cylinder based on radius
- ϕ_r = Thiele parameter for rectangular parallelepiped based on one side (a)
- ϕ_A = modified Thiele parameter based on V_p/S
- ϕ = Thiele parameter for a sphere or equivalent sphere
- ρ = level surface value of u
- ρ_s = density of catalyst particle
- v_i, v_{is} defined in text
- ω_i, ω_{is} defined in text
- ΔH = heat of reaction
- ∂O = surface boundary sphere

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APPENDIX

Lemma 1: For any simply connected region B

$$\iint_{\Sigma} \frac{\partial u}{\partial n}\, ds = \text{Min} \iiint_{B} [\lambda f^2 + (\nabla f)^2]\, dV \qquad (3)$$

where f is an arbitrary function satisfying

$$0 \leqq f \leqq 1 \quad \text{in } B \qquad (4)$$

$$f = 1 \text{ on } \Sigma$$

and $\nabla^2 u = \lambda u$ in B and $u = 1$ on Σ.

Proof: Since $u = 1$ on Σ

$$\iint_{\Sigma} \frac{\partial u}{\partial n}\, ds = \iint_{\Sigma} u \frac{\partial u}{\partial n}\, ds =$$

$$\iiint_B [u\nabla^2 u + (\nabla u)^2]\, dV = \iiint_B [\lambda u^2 + (\nabla u)^2]\, dV \quad (5)$$

Now define $h = f - u$ so that $h = 0$ on Σ and then

$$\iiint_B [\lambda f^2 + (\nabla f)^2]\, dV = \iiint_B [\lambda u^2 + 2\lambda uh + \lambda h^2 + (\nabla u)^2 + 2\nabla u \cdot \nabla h + (\nabla h)^2]\, dV \quad (6)$$

From Green's theorem

$$\iiint_B \nabla h \cdot \nabla u = \iint_\Sigma h \frac{\partial u}{\partial n} - \iiint_B h\nabla^2 u\, dV$$

and since $h = 0$ on Σ and $\nabla^2 u = \lambda u$ in B

$$\iiint_B \nabla h \cdot \nabla u\, dV = -\iiint_B \lambda hu\, dV$$

and therefore (6) becomes

$$\iiint [\lambda f^2 + (\nabla f)^2]\, dV = \iiint_B [\lambda u^2 + (\nabla u)^2 + \lambda h^2 + (\nabla h)^2]\, dV \geqq \iiint_B [\lambda u^2 + (\nabla u)^2]\, dV \quad (7)$$

From (5) and (7) one obtains

$$\iint_\Sigma \frac{\partial u}{\partial n} dS = \text{Min} \iiint_B [\lambda f^2 + (\nabla f)^2]\, dV \quad (8)$$

since h is arbitrary save $h = 0$ on Σ.

Lemma 2: If one can construct a function $\bar{u}$ in a sphere O where $\bar{u} = 1$ on the surface ∂O of the sphere with O having the same volume as B, such that

$$\iiint_O [\lambda \bar{u}^2 + (\nabla \bar{u})^2]\, dV \leqq \iiint_B [\lambda u^2 + (\nabla u)^2]\, dV$$

and where u is the solution of $\nabla^2 u = \lambda u$ in B, $u = 1$ on Σ, then

$$\iint_{\partial O} \frac{dv}{dn} ds \leqq \iint_\Sigma \frac{\partial u}{\partial n} dS \quad (10)$$

where v is the solution of $\nabla^2 v = \lambda v$ in O and $v = 1$ on ∂O

Proof: From Equation (8)

$$\iint_\Sigma \frac{\partial u}{\partial n} dS = \iiint_B [\lambda u^2 + (\nabla u)^2]\, dV \quad (11)$$

$$\iint_{\partial O} \frac{\partial v}{\partial n} dS \leqq \iiint_O [\lambda \bar{u}^2 + (\nabla \bar{u})^2]\, dV \quad (12)$$

Direct substitution of Equations (11) and (12) in Equation (9) proves the lemma.

Lemma 3: A function $\bar{u}$ which satisfies condition (9) may be constructed by spherical symmetrization.

Proof: In the body B we can construct level surfaces of u. These surfaces $\Sigma(\rho)$ are $u = \rho$, $0 \leqq \rho \leqq 1$. The volume inside $\Sigma(\rho)$ is $V(\rho)$. The domain between two neighboring level surfaces $u = \rho$ and $u = \rho + d\rho$ will be called $\Delta B(\rho)$.

The function $\bar{u}(\bar{\rho})$ will be chosen such that the volume closed by any level surface in the sphere and the body B will be the same. The volume of the sphere and the body are defined by the level surface $\bar{u} = u = 1$, and hence owing to the construction the sphere has the same volume as the body B. The function $\bar{u}$ will be constructed so that

$$\bar{u}(\bar{\rho}) = \rho \quad (13)$$

where $\bar{\rho}$ satisfies the condition

$$\frac{4}{3}\pi \bar{\rho}^3 = V(\rho)$$

Define a function

$$G = \text{grad } u \cdot \mathbf{n} = \frac{d\rho}{dn} = |\text{grad } u|$$

since grad u and $\mathbf{n}$ are both normal to the level surfaces of u. Now

$$\iiint_{\Delta B(\rho)} (\nabla u)^2\, dV = \iint_{\Sigma(\rho)} (\nabla u)^2\, d\sigma\, dn = \iint_{\Sigma(\rho)} G^2\, d\sigma\, dn$$

$$= \iint_{\Sigma(\rho)} G \frac{d\rho}{dn} d\sigma\, dn = d\rho \iint_{\Sigma(\rho)} G\, d\sigma = P(\rho)\, d\rho$$

or

$$P(\rho) = \iint_{\Sigma(\rho)} G\, d\sigma \quad (14)$$

The difference in the volumes of the neighboring domains is $V(\rho + d\rho) - V(\rho) = V'(\rho)\, d\rho$

$$= \iiint_{\Delta B(\rho)} dV = \iint_{\Sigma(\rho)} d\sigma\, dn = \iint_{\Sigma(\rho)} d\sigma \frac{d\rho}{G}$$

or

$$V'(\rho) = \iint_{\Sigma(\rho)} \frac{d\sigma}{G} \quad (15)$$

From Schwarz inequality

$$\left(\iint_{\Sigma(\rho)} G\, d\sigma\right)\left(\iint_{\Sigma(\rho)} \frac{d\sigma}{G}\right) \geqq \left(\iint_{\Sigma(\rho)} d\sigma\right)^2 = [\text{surface of } \Sigma(\rho)]^2 \quad (16)$$

The isoperimetric inequality for simply connected bodies is

$$S \geqq 4\pi \left(\frac{3V}{4\pi}\right)^{2/3} = A\, V^{2/3}$$

$$A = (4\pi)^{1/3}\, 3^{2/3} \quad (17)$$

where the equality holds only for a sphere. From (14), (15), (16), and (17)

$$P(\rho)\, V'(\rho) \geqq A^2\, V(\rho)^{4/3}$$

or

$$P(\rho) \geqq \frac{A^2\, V(\rho)^{4/3}}{V'(\rho)}$$

and equality holds only for a sphere, where $V(\rho)$ is the volume of the level surface $u = \rho$.

Hence

$$\iiint_{B(1)} (\nabla u)^2\, dV = \int_{0<\rho\leqq 1} P(\rho)\, d > \int_{0<\rho\leqq 1} A^2 \frac{V(\rho)^{4/3}}{V'(\rho)} d\rho \quad (18)$$

Now the spherical symmetrization is such that in the sphere and in B $V(\rho)$ and $V'(\rho)$ are the same. Thus it follows from (15) that

$$\iiint_{B(1)} (\nabla u)^2\, dV \geqq \iiint_{O(1)} (\nabla \bar{u})^2\, dV \quad (19)$$

Also

$$\iiint_{B(1)} u^2\, dV = \iiint_{B(1)} \rho^2\, dV(\rho) = \int_{0<\rho\leqq 1} \rho^2\, V'(\rho)\, d\rho$$

since ρ and $V'(\rho)$ are the same for $B(\rho)$ and $O(\rho)$

$$\iiint_{B(1)} u^2\, dV = \iiint_{O(1)} \bar{u}^2\, dV \quad (20)$$

From (19) and (20) we obtain

$$\iiint_{B(1)} [(\nabla u)^2 + \lambda u^2]\, dv \geqq \iiint_{O(1)} [(\nabla \bar{u})^2 + \lambda \bar{u}^2]\, dV$$

The theorem then follows from Lemmas 2 and 3.

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