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DESIGN OF SLURRY REACTOR FOR INDIRECT LIQUEFACTION APPLICATIONS. QUARTERLY TECHNICAL PROGRESS REPORT, JANUARY 1990--MARCH 1990

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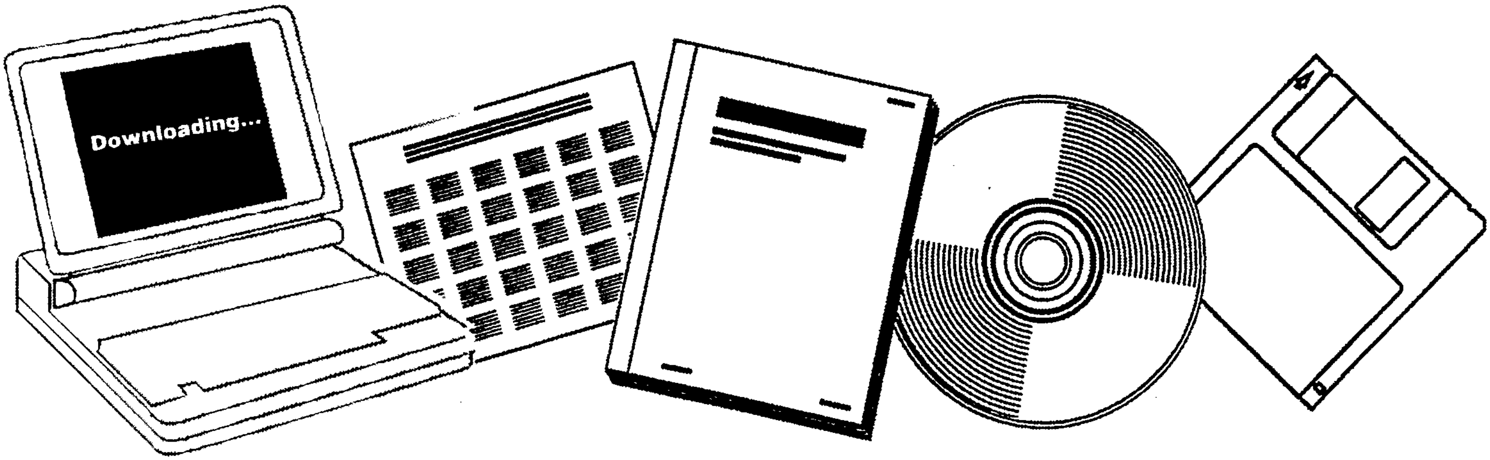

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QUARTERLY TECHNICAL PROGRESS REPORT

January 1990 - March 1990

DESIGN OF SLURRY REACTOR FOR INDIRECT LIQUEFACTION APPLICATIONS

by

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MASTER

Introduction

The objective of this project is to design a conceptual slurry reactor for two indirect liquefaction applictations;production of methanol and production of hydrocarbon fuels via Fischer-Tropsch route. The work will be accomplished by the formulation of reactor models for both the processes and use computer simulation. Process data, kinetic and thermodynamic data, heat and mass transfer data and hydrodynamic data will be used in the mathematical models to describe the slurry reactor for each of the two processes. The cost of current vapor phase reactor systems will be compared with cost estimated for the slurry reactor systems. For the vapor phase systems, upstream and downstream processing equipments may have to be included during cost analysis for a meaningful cost comparison.

Finally, we will point out any inadequacies in the technical database currently available for a commercial design of the slurry reactor and identify research needs to improve upon the slurry reactor design. Assumptions used in the design will be documented.

Project Status

During this quarter, work was directed toward mathematical description and formulation of reactor model and selection of appropriate correlations for estimation of model parameters. Various kinetic models for methanol synthesis reaction are under review.

Reactor Model

The development of mathematical model for a reactor starts with differential mass and energy balance equations. The resulting differential equations can be solved using suitable boundary conditions. The reactor design configuration and its operating conditions usually allow for some simplifying assumptions to be made.

The following assumptions made for the development of FT synthesis reactor model are generally applicable to slurry reactors with fine catalyst particles.

Model Assumptions:

i) The total pressure within the reactor does not vary with axial position, i.e. the influence of hydrostatic head on gas expansion is neglected. This assumption is reasonable in view of the relatively high pressures used in both the processes.

ii) The effectiveness factors for the catalyst particles are taken as unity. Due to the relatively small particle sizes used in the reactors, diffusional limitations should be negligible.

iii) Again, as a consequence of the small particle size, mass and heat transfer resistances between the catalyst and liquid are assumed to be negligible.

iv) The gas and slurry phases are modeled using the axial dispersion model, the most appropriate model for bubble column reactors (Deckwer et al., 1983).

v) The catalyst is not uniformly distributed in the reactor and the sedimentation-dispersion model (Cova, 1966; Kato et al., 1972, Smith et.al, 1985) is used for modeling the catalyst concentration.

vi) The hydrodynamic parameters, namely gas holdup, interfacial area, heat and mass transfer coefficients and dispersion coefficients are assumed to be spatially independent.

vii) A heat balance will be made only on the slurry phase, since the heat capacity of the gas phase is much lower than that of the slurry.

viii) The temperature dependence of the total gas phase concentration $C_G=P/RT$ can be neglected since

$$d(1/T)/dz = -1/T^2(dT/dz) \sim 0$$

This approximation is justified since the temperature gradient is usually small due to liquid-phase back-mixing.

ix) Heat and mass dispersion in the slurry phase are equal and related by Lewis analogy, i.e.

$$D_L = (k_{eff}/\rho\, C_p)_L$$

x) Steady-state conditions prevail

Model Equations:

Based on the above assumptions, the following mass and energy balance were developed for the reactor model. Definitions of the variables, coefficients and dimensionless groups are given in the Notation.

Gas Phase Mass Balance:

The gas phase mass balance with axial dispersion for component i can be written as:

$$\frac{1}{Pe_G}\frac{d^2y_i}{dz^2} - \frac{d(vy_i)}{dz} - (St_{G,i})(y_i - x_i) = 0$$

subject to the following boundary conditions:

$$vy_i - \frac{1}{Pe_G}\frac{dy_i}{dz} = y_{io} \qquad \text{at } z = 0$$

$$\frac{dy_i}{dz} = 0 \qquad \text{at } z = 1$$

The superficial gas velocity (U_G) will vary with the volumetric flowrate of gas along the column. The gas velocity can be determined by writing an overall mass balance: assumptions (i) and (viii) ensure negligible effect of change in pressure and temperature along the column. Thus

$$-\frac{dv}{dz} - \Sigma \, St_{G,i} \, (y_i - x_i) = 0$$

The above equation is subject to the boundary condition

$$v = 1 \qquad \text{at } z = 0$$

Liquid Phase Mass Balance:

The liquid phase mass balance for component i can be written as:

$$\frac{1}{(Pe_L)} \frac{d^2x_i}{dz^2} - \frac{dx_i}{dz} + St_{L,i} \, (y_i - x_i) + \Sigma \, \alpha_{i,k} \epsilon_l C_{cat} r_k (H_i L/PU_L) = 0$$

subject to the following boundary conditions:

$$-\frac{1}{Pe_L} \frac{dx_i}{dz} + x_i = x_{i,o} \qquad \text{at } z = 0$$

$$dx_i/dz = 0 \qquad \text{at } z = 1$$

Solid Phase Mass Balance:

The axial distribution of catalyst is governed by gravitational settling and axial dispersion due to agitation of the catalyst slurry by the gas flow. From assumption (vi) the volume fraction of the liquid is constant along the column length and hence the mass balance on catalyst can be written as:

$$\frac{1}{Pe_s}\frac{d^2w}{dz^2} + \frac{dw}{dz} = 0$$

with boundary conditions:

$$w - 1/Pe_s(dw/dz) = 0 \qquad \text{at } z = 0$$

$$\int_0^1 w\,dz = 1$$

An analytical solution to the above equation is possible and is given below:

$$w = \frac{Pe_s \exp(-Pe_s z)}{[1 - \exp(-Pe_s)]}$$

Heat Balance

For fine catalyst particles, considering the slurry phase to be pseudo-homogenous, we can write the following heat balance:

$$\frac{1}{Pe_H}\frac{d^2\theta}{dz^2} - \frac{d\theta}{dz} - St_H(\theta - \theta_W) + \Sigma \frac{\epsilon_L C_{cat} r_k (-\ H_R)_k L}{\rho_L C_p U_L T_o} = 0$$

subject to the following boundary conditions:

$$\theta - 1/Pe_H(d\theta/dz) = 0 \text{ at } z = 0$$

$$d\theta/dz = 0 \qquad \text{at } z = 1$$

Determination of Model Parameters

An extensive literature search was undertaken to estimate various parameters in the above model equations. Justifications for selecting various correlations are summarized in the following paragraphs, while actual correlations are shown in Appendix A.

Gas Holdup

The gas holdup in slurry bubble columns is affected by a number of factors which include: physical properties and coalescing behavior of liquid, gas and liquid velocities, column diameter, gas distributor design, solids concentration, size and density. The gas holdup also depends on the operating flow regime in the column. The development of a general correlation which takes into account the effect of all these variables is indeed a difficult task. The work done by various researchers is summarized below:

There seems to be general agreement among researchers that the gas holdup and volumetric mass transfer coefficient decrease with increasing solids concentration for particle size larger than 10 micron (Kato et al., 1973; Kara et al., 1982; Koide et al., 1984). For particles smaller than 10 micron, the gas holdup can be higher than the solid free bubble column (Kara et al., 1982; Sada et al., 1986). The effect of suspended particles was generally found to diminish when the superficial gas velocity was higher than 0.1 m/s. Kato et al. (1973) found the gas holdups to decrease with increasing particle size over the size range used for their study (i.e. 60-175 micron). These results show that a correlation for gas holdup in slurry bubble columns should account for the effects of particle size as well as slurry concentration. While most of the literature work for hydrodynamic studies in slurry bubble columns has been carried out in aqueous solutions, Bukur et al. (1989) used Fischer-Tropsch waxes for their study. The data of Bukur et al. (1989) is, therefore more relevant for our purposes.

The correlations for gas holdup in slurry bubble columns have been proposed by Deckwer et al. (1980),Koide et al. (1984), Sada et al. (1986) and Sauer and Hempel (1987). These correlations will be tested using data from Bukur et al. (1989) and pilot plant data from Air Products and Chemicals. The hydrodynamics of solid-free bubble column have been investigated more extensively. Some of the well known correlations for solid-free bubble column (Hughmark, 1967; Akita and Yoshida, 1973; Hikita et al., 1980) will also be tested. Some of these correlations may give better fit under certain operating conditions.

Volumetric Mass Transfer Coefficient

Like gas holdup, volumetric mass transfer coefficient in slurry bubble columns has been found to depend on the concentration, density and size of solids used. The volumetric mass transfer coefficient was found to decrease with increasing solids concentration by most researchers. Correlations for volumetric mass transfer coefficient in slurry bubble columns have been proposed by Koide et al. (1984), Nguyen-tien et al. (1985) and Sauer and Hempel (1987). There is no relevant data available, however, to test the validity of these correlations for our purposes. The correlation of Nguyen-tien et al. (1985) is based on a comprehensive study of gas liquid mass transfer in three-phase contactors. This empirical correlation, however, is based on data obtained for oxygen mass transfer in aqueous solutions. Therefore suitable corrections for physical properties and diffusivities of gases need to be applied. The correlation proposed by Akita and Yoshida (1973) for solid-free bubble column should also give a reasonable estimate of volumetric mass transfer coefficient for slurry bubble columns, since it includes a gas holdup term and also accounts for physical properties of liquid. We would, therefore, provide an option to use either of these of these correlations for the estimation of volumetric mass transfer coefficient.

Liquid Phase Dispersion Coefficient

While liquid phase dispersion in solid-free bubble columns has been investigated extensively, very little work has been done to measure liquid backmixing in slurry bubble columns. However, in the suspension of fine catalyst particles used in Fischer-Tropsch synthesis reactors, there is expected to be little effect of the presence of solids on liquid mixing. The various correlations available in the literature for liquid phase dispersion in bubble column were compared by Wendt et al. (1984). The correlation proposed by Deckwer et al. (1974) provides a good estimate of the liquid phase dispersion coefficient. This correlation will, therefore be used to predict liquid phase dispersion coefficient.

Axial Solids Distribution

Although several groups of researchers have investigated distribution of solids in slurry bubble columns, the work of Bukur et al. (1989), using Fischer-Tropsch waxes, is more relevant for our purposes. Bukur et al. (1989) observed that the axial distribution for the 0-5 μm and 20-44 μm iron oxide and silica particles were uniform for all runs in the continuous mode of operation. For batch mode of operation, the 0-5 μm particles showed only a slight gradient with higher concentration towards the bottom of the column. However, with 20-44 μm particles significant gradients in axial solids distribution profiles were observed in the batch mode of operation. The study of Bukur et al. (1989) also shows that the correlation of Smith and Ruether (19850 gives a good prediction of solids dispersion coefficient in slurry bubble columns.

Gas Phase Dispersion Coefficient

The correlation of Towell and Ackermann (1972) will be used to estimate the gas phase axial dispersion coefficient. In a recent study, Kawagoe et al. (1989) showed that the correlation of Towell and Ackerman (1972) gave a good estimate of the overall gas phase dispersion coefficient in bubble columns.

Heat Transfer Coefficient

In a recent review, Saxena (1989) pointed out the limitations of available literature correlations to predict heat transfer coefficient in slurry bubble columns. His group is expected to propose new and/or refined correlations in the near future, based on their experimental data in slurry bubble columns.

Development of Computer Codes

The model equations for the slurry reactor constitute a set of coupled second-order ordinary non-linear differential equations. These equations are not amenable to an analytical solution and therefore, numerical methods will be used for solution. We plan to use the B-spline collocation technique developed by Ascher et.al. (1981) which from our previous experience has shown to be very efficient for solution of boundary-value problems. Orthogonal collocation techniques are particularly suitable for the solution of boundary value problems and the software package, COLSYS, developed by Ascher et al. (1981) will be used for the numerical solution of these equations. This method is based on spline collocation at Gaussian points using a B-spline basis. Approximate solutions are computed on a sequence of automatically selected meshes until a user-specified set of tolerances is satisfied. This code has been found to be particularly effective for difficult problems (eg., determining effectiveness factors at high values of Thiele modulus (Davis, 1984)).

The computer codes will be developed on a VAX 8700 computer at the University of Pittsburgh under a VMS operating system. Standard FORTRAN 77 will be used for writing the codes since this will ensure transfer to other compatible systems.

Notation:

a : gas-liquid interfacial area, m^{-1}

a_H : interfacial area for heat transfer, m^{-1}

C_{cat} : catalyst concentration, gm/cc slurry

$C_{cat,avg}$: average catalyst concentration, kg/m^3 slurry

$C_{cat,b}$: catalyst concentration at column bottom, kg/m^3 slurry

$C_{G,i}$: gas-phase concentration of component i, $kmol/m^3$

$C_{L,i}$: liquid-phase concentration of component i, $kmol/m^3$

C_G : total gas-phase concentration, $kmol/m^3$

C_p : heat capacity, kJ/kg.K

D_G : gas-phase dispersion coefficient, m^2/s

D_i : diffusivity of component i in the reactor liquid (m^2/s)

D_L : liquid-phase dispersion coefficient, m^2/s

$D_{o,w}$: diffusivity of oxygen in water (m^2/s)

D_S : solid-phase dispersion coefficient, m^2/s

E_L : thermal diffusivity (m^2/s)

h : heat transfer coefficient, $kJ/m^2.s.K$

H_i : Henery's constant for component i ($kgmoles/m^3.atm$)

H_o : static slurry height (m)

H_R : heat of reaction, kJ/kmol

k_{eff} : effective heat dispersion coefficient, kJ/m.s.K

$k_{L,i}$: liquid-side mass transfer coefficient for component i, m/s

k_La : volumetric mass transfer coefficient (s^{-1})

$(k_La)_i$: volumetric mass transfer coefficient for component i (s^{-1})

$(k_La)_o$: volumetric mass transfer coefficient for oxygen in aqueous phase (s_{-1})

L : reactor length, m

m_i : solubility coefficient for component i, $(mol/m^3)_G/(mol/m^3)_L$

P : total pressure, atm

R : universal gas constant, 0.082 m^3-atm/kmole/K

r_k : reaction rate for kth reaction, kmol/kg.s

T : temperature, K

T_o : inlet temperature, K

T_w : reactor wall temperature, K

U_G : gas superficial velocity, m/s

U_{Go} : inlet gas superficial velocity, m/s

U_L : liquid superficial velocity, m/s

U_S : settling velocity of catalyst particles in swarm, m/s

v : dimensionless gas-phase superficial velocity (U_G/U_{GO})

w : dimensionless catalyst concentration

x_i : dimensionless liquid-phase concentration of component i ($C_{L,i}H_i/P$)

y_i : mole fraction of component i in the gas phase

z : dimensionless axial distance (x/L)

Dimensionless numbers

Bo : Bodenstein number (U_SH_c/D_s)

Fr_G : Froude number for gas $(U_G/ gD_c$

Pe_G : gas-phase Peclet number $(U_{Go}L/D_G\epsilon_G)$

Pe_H : Peclet number for heat transfer $(U_LL/E_L\epsilon_L)$

Pe_L : liquid-phase Peclet number $(U_LL/D_L\epsilon_L)$

Pe_S : solid-phase Peclet number $[(U_S - U_L/\epsilon_L)(L/D_S)$

Pe_p : particle Peclet number (U_GD_c/D_s)

Re_G : gas Reynolds number $(U_GD_co_l/\mu_l)$

Re_p : particle Reynolds number $(U_td_po_l/\mu_l)$

St_G : gas-phase Stanton number $(K_{L,i}aL/U_{Go})(RT/H_i)$

St_H : Stanton number for heat transfer (ha_HL/pC_pU_L)

$St_{L,i}$: liquid-phase Stanton number for component i $(K_{L,i}aL/U_L)$

Greek

α_{ik} : stoichiometric coefficient of component i in reaction k

ϵ_G : gas holdup

ϵ_L : liquid holdup

ϵ_s : solids holdup

$\epsilon_{s,sus}$: solids holdup in the slurry

μ_G : viscosity of gas phase (kg/m.s)

μ_L : viscosity of liquid phase (kg/m.s)

ν_L : kinematic liquid viscosity (m^2/s)

ν_{sus} : kinematic viscosity of slurry (m^2/s)

$\nu_{eff,rad}$: radial momentum transfer coefficient (m^2/s)

Θ : dimensionless temperature (T/T_o)

Θ_w : ratio of wall temperature T_w to inlet temperature T_o

ρ_G : density of gas phase (kg/m^3)

ρ_L : density of the liquid phase, kg/m^3

ρ_{sus} : density of pseudo-homogeneous slurry (kg/m^3)

σ : surface tension of liquid (kg/s^2)

Subscripts

L : liquid phase

G : gas phase

i : component i

k : kth reaction

w : wall

o : inlet condition

S : solid phase

H : heat transfer

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Appendix A

Correlations for Model Parameters

Gas Holdup Correlations:

Koide et al. (1984)

$$\frac{\epsilon_G}{(1-\epsilon_G)^4} = \frac{0.277\left(\frac{U_G\mu_L}{\sigma_L}\right)^{0.918}\left(\frac{g\mu_L^4}{\rho_L\sigma_L^3}\right)^{-0.252}}{1+4.35\left(\frac{\epsilon_S}{\rho_S}\right)^{0.748}\left(\frac{\rho_S-\rho_L}{\rho_L}\right)^{0.881}\left(\frac{D_T U_G\rho_L}{\mu_L}\right)^{-0.168}}$$

This correlation is based on a large number of data points.

Sada et al. (1986)

$$[\epsilon_G/(1-\epsilon_G)^3]_{III} = 0.41\ U_t^{\ 1/16}\ \epsilon_{s,sus}^{\ -(0.125\ U_t^{-0.18})}\ \ U_G[\epsilon_G/(1-\epsilon_G)^4]_{II}$$

here

$U_t = g(\rho_p - \rho_l)d_p^2/(18\mu_l)$

units of U_t and U_G are in cm/s

Sauer and Hempel (1987)

$$[\epsilon_G/(1-\epsilon_G)] = 0.0277\ [U_G/(\nu_{sus}\ gU_G)]^{0.844}\ [\nu_{sus}/\nu_{eff,rad}]^{-0.136}\ [C_{cat,avg}/C_{cat,b}]^{0.0392}$$

$\nu_{sus} = \nu_l\ (1 + 2.5\epsilon_s + 10.05\epsilon_s^2 + 2.73\text{x}10^{-3}\exp(16.6\epsilon_s))$

$\nu_{eff,rad} = 0.011\ D_c(gD_c)^{0.5}\ (U_G^3/\nu_{sus}g)^{1/8}$

$C_{cat,avg}/C_{cat,b} = [1 - \exp(-Bo)]/Bo$

$Bo = U_sH_o/D_s$

This correlation accounts for the effect of suspension viscosity on gas holdup in slurry bubble column.

Deckwer et al. (1980)

$$\epsilon_G = 0.053\ U_G^{1.1}$$

This correlation is based on the data obtained with molten wax slurry system applied in the FT synthesis.

Hughmark (1967)

$$\epsilon_G = [2 + (0.35/U_G)(\rho_L\ \sigma/72)^{1/3}]^{-1}$$

Smith and Ruether (1985) observed that incorporation of the slurry density rather than the liquid density in the above equation reduced the residual error for slurry bubble columns.

Akita and Yoshida (1973)

$$[\epsilon_G/(1-\epsilon_G)^4] = 0.2(gD_c^2\rho_L)^{1/8}\ (gD_c^3/\nu_L^2)^{1/12}\ U_G/(gD_c)^{0.5}$$

This correlation is based on the data obtained with a solid-free bubble column. For a slurry bubble column this correlation may be applicable only for suspension of very fine particles (< 10 μm) at high gas velocities (> 0.1 m/s).

Hikita et al. (1980)

$$\epsilon_G = 0.672(U_G\mu_L/\sigma)^{0.578}\ (\mu_L^4 g/c_L\sigma^3)^{-0.131}(\rho_G/\rho_L)^{0.062}\ (\mu_G/\mu_L)^{0.107}$$

This correlation is also based on the data obtained from solid-free bubble column, its applicability may, therefore, be limited to fine particles suspension.

Correlations for Volumetric Mass transfer Coefficient:

Nguyen-tien et al. (1985)

$$k_L a = 0.39\ (1 - \epsilon_{s,sus}/0.58) U_G^{0.67}$$

This correlation is based on data obtained for oxygen mass transfer in aqueous solutions. For other systems $k_L a$ can be corrected as below:

$$(k_L a)_i = (k_L a)_o (D_i/D_{o,w})^{2/3}\ (\mu_w/\mu_L)^{0.3}$$

Akita and Yoshida (1973)

$$(k_L a D_c^2/D_l) = 0.6\ \epsilon_G^{1.1}\ (\mu_l/\rho_L D_l)^{0.5}\ (g D_c^2 \rho_L/\sigma)^{0.62}\ (g D_c^3 \rho_L^2/\mu_L^2)^{0.31}$$

This correlation is based on the data obtained from solid-free bubble columns. Its applicability for slurry bubble columns may be limited to dilute suspension of fine particles.

Correlations for Liquid Phase Dispersion:

Deckwer et al. (1974)

$$D_L = 2.7\ D_c^{1.4}\ U_G^{0.3}$$

Correlations for Axial Solids Dispersion Coefficient

Smith and Ruether (1985)

$$Pe_p = 9.6(Fr_G{}^6/Re_G)^{0.114} + 0.019Re_p{}^{1.1}$$

$$Re_p = Ar/18 \qquad \text{if } Re_p < 0.5$$

$$Re_p = (Ar/13.9)^{0.7} \qquad \text{if } Re_p > 0.5$$

where

$$Ar = \frac{\rho_L(\rho_{cat} - \rho_L)g}{\mu_L{}^2} d_p{}^3$$

and

$$Re_p = \frac{U_t d_p \rho_L}{\mu_L}$$

Smith and Ruether (1985) have suggested the following equation for hindered settling velocity in particle swarm.

$$U_s = 1.1\ U_G{}^{0.026}\ U_t{}^{0.8}\ \epsilon_{s,sus}{}^{3.5}$$

Correlation for Gas Phase Dispersion Coefficient

Towell and Ackerman (1972)

$$D_G = 20\ D_c{}^2 U_G$$

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