Atmospheric Environment Pergamon Press 1971. Vol. 5, pp. 571–578. Printed in Great Britain.

RESIDENCE TIME OF PARTICLES IN URBAN AIR*

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(*First received* 3 *June* 1970 *and in final form* 18 *November* 1970)

Abstract—A preliminary set of experiments to determine particle residence time in the dry atmosphere was conducted. Using a membrane filter exposed in the horizontal plane and another membrane filter for sampling the ground level concentration of suspended particulate matter, ground level concentration, and flux of atmospheric aerosol were measured. Assuming a ceiling height of 2 km the mean residence time of atmospheric aerosol was calculated. The preliminary results indicate that the mean residence time of submicron particles in the absence of precipitation is on the order of 10^2–10^3 h. Particles in the range of 1–10 μm have residence times on the order of 10–100 h.

INTRODUCTION

IN ORDER to maintain a balance between the input of both natural and artificial particulate contaminants of the atmosphere and the concentrations which exist at any given place and time, there must be a very significant removal of particles from the atmosphere. The mechanisms of removal of particulate matter from the atmosphere include diffusion, sedimentation, capture by cloud droplets and scrubbing by precipitation.

If capture by cloud droplets and the scrubbing effect of rain are considered together as the process of "wet removal" and sedimentation is considered "dry removal", one may express the total removal as the sum of these three components, i.e. diffusion, wet removal and dry removal. The net diffusion of particles out of a control volume of atmosphere may be neglected as being several orders of magnitude smaller than the other mechanisms of removal. Of the remaining two mechanisms, in terms of efficiency and the total particulate bulk cleared from the atmosphere in a short period of time, the more important is that of "wet removal". The enormous quantity of water which cycles continuously in the atmsophere is responsible for the bulk of particulate removal, but the dry removal process essentially determines the life of aerosols in the air. This apparently contradictory assertion can be defended as follows. After a significant precipitation, there is a drastic reduction in the particulate concentration of the lower atmosphere. The build up and equilibrium concentrations attained between two subsequent intervals of precipitation is governed solely by the dry removal process provided that the input conditions do not alter significantly. The latter is, in most cases, a reasonable assumption in an air basin. Hence, if one assumes that a steady state is reached for addition and removal of particulate matter to and from the atmosphere, during periods between precipitation it is possible to derive an expression for the mean residence time of suspended particulate matter injected into the air.

The determination and basic understanding of dry removal of particles from the atmosphere has been at best scanty. JUNGE (1963) calculated the residence time

* This investigation was supported by Public Health Service Research Grant No. AP 00431-05, National Center for Air Pollution Control.

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of particulate matter based on their fall velocities and assumed a 5 km ceiling for the uniform mixing depth above the earth. His results were not experimentally verified. McDonald (McDONALD, 1961) attempted to approximate such a residence time using available values of concentration of suspended particulate matter and dustfall. The result he obtained for the mean residence time of atmospheric particles was on the order of 10 h. This result was based on data which by its very nature, led to calculations of residence time based on wet and dry removal processes. This result applied to the entire size spectrum of airborne particles. It is clear that the removal of airborne particles from the atmosphere depends on aerodynamic particle size. This result, therefore, leaves much to be desired. It does not permit one to associate a residence time with a particle of given size after it is injected into the atmosphere.

Chamberlain (CHAMBERLAIN, 1966) made measurements in the field and in a wind tunnel of the transport of Lycopodium spores of approximately 30 μm dia. and 1.175 g cm^{-3} density to grass and other surfaces. Wind tunnel experiments were also performed with particles as small as 0.08 μm. For release at a height of 10 m, it was estimated that half the spores could travel a distance of 10 km.

Because of the scarcity of data, and in order to obtain a method to compare the relative removal efficiencies and magnitudes of wet and dry removal we performed a preliminary set of experiments to determine particle residence time in the atmosphere. This residence time was calculated from the particle flux to the ground and the ground level concentration of particles segregated into size groups.

THEORETICAL CONSIDERATIONS

Consider a vertical distribution of particle concentration $C(h)$ with boundary conditions $C(0) = C_0$ and $C(H) = C_H$ at heights 0 and H above ground. From the studies on the vertical distribution of aerosol concentration in the atmosphere (HAMILTON, 1966; AHLQUIST and CHARLSON, 1968) it is reasonable to assume that this concentration function is monotonic non-increasing and $C_H << C_0$ (FIG. 1). We can also consider a monotonic non-increasing particle flux $J(h)$, orthogonal to a horizontal surface element on the ground, with boundary conditions $J(0) = J_0$ and $J(H) = 0$. Furthermore, it is assumed that a steady state is established over the entire region under consideration and $C(h)$, $C^{-1}(h)$ and $J(h)$ are integrable. Then,

$$J(h) = C(h)\, \bar{U}_R(h) \tag{1}$$

at any given height h and the particle removal velocity $\bar{U}_R(h)$ is in the direction of the flux. We can calculate the arithmetic mean particle residence time, τ, by assuming that the only contribution to particle removal on the vertical axis is described by $\bar{U}_R$, the mean particle velocity.

$$\tau = H/2\, \bar{U}_R \tag{2}$$

But

$$\bar{U}_R = \frac{1}{H}\int_0^H U_R(h)\, dh = \frac{1}{H}\int_0^H \frac{J(h)}{C(h)}\, dh. \tag{3}$$

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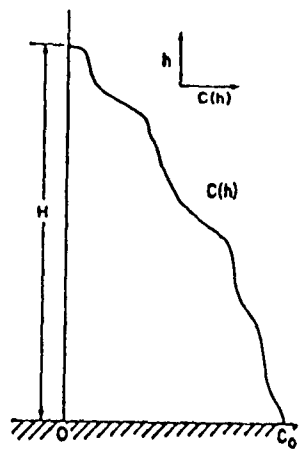

FIG. 1. Variation of particle concentration between ground level and height H.

When the ground level concentration C_0 and ground level flux J_0 are measured it is possible to determine a limiting value for $\bar{U}_R$. Let the normalizing function ψ be:

$$\psi = 1 - h/H, \quad 0 \leq \psi \leq 1 \tag{4}$$

and define the distribution functions $g(\psi)$ and $f(\psi)$ for $J(\psi)$ and $C(\psi)$ respectively.

$$C(\psi) = C_0 f(\psi) \qquad 0 \leq f(\psi) \leq 1 \tag{5}$$

$$J(\psi) = J_0 g(\psi) \qquad 0 \leq g(\psi) \leq 1. \tag{6}$$

Now we consider equation (3):

$$\bar{U}_R = \frac{1}{H}\int_0^H \frac{J(h)}{C(h)}\,dh = \frac{J_0}{C_0}\int_0^1 \frac{g(\psi)}{f(\psi)}\,d\psi \tag{7}$$

The value of the integral above may be investigated using the mean value theorem of integral calculus:

$$\int_0^1 \frac{g(\psi)}{f(\psi)}\,d\psi = \frac{1}{f(x)}\int_0^1 g(\psi)\,d\psi, \quad 0 \leq x \leq 1. \tag{8}$$

By the definition of the function $g(\psi)$:

$$\int_0^1 g(\psi)\,d\psi \equiv 1. \tag{9}$$

And similarly since $f(\psi)$ is normalized:

$$0 < f(\psi) \leq 1. \tag{10}$$

Hence the right hand side of equation (8) is:

$$\frac{1}{f(x)} \int_0^1 g(\psi)\, d\psi \geq 1. \tag{11}$$

And therefore using equation (11):

$$\int_0^1 \frac{g(\psi)}{f(\psi)} \geq 1. \tag{12}$$

Therefore, from equation (7)

$$U_R \leq U_0. \tag{13}$$

Therefore from equation (2)

$$\tau \geq H/2\, U_0 = C_0 H/2\, J_0. \tag{14}$$

Equation (14) indicates that the mean particle residence time calculated using measured ground level particle concentration and flux represents the lower bound of mean particle residence time, i.e. all values of mean particle residence time should exceed or be equal to this value. Of course, this result is only applicable within the framework of the assumptions made here.

In order to extend this argument to cover a larger class of cases we can investigate the effects of local deviations from monotonic behavior. It is clearly evident that if the average value of $C(h)$ does not exceed C_0 and the behavior of $J(h)$ is as assumed the conclusions arrived at above will still be valid. The experimental values available (HAMILTON, 1966; AHLQUIST and CHARLSON, 1968) on the vertical distribution of atmospheric aerosol seems to justify the acceptance of this limiting condition provided that $J(h)$ behaves as assumed. Now we can investigate the strength of the monotonic behavior assumption of $J(h)$. Suppose due to the increase in local eddy diffusivity $J(h)$ between h_1 and h_2 is increased in a manner that the monotonic balance is disturbed. Then we expect a local build up of aerosol at a level below h_1, say h_0. The removal velocity between h_1 and h_0 being the same these will be increased in concentration and hence there will be an increase in the flux in that direction, increasing the flux all the way to the ground level. This, of course, will again establish the monotonic behavior of the flux. Because the sampling time in the studies to be described is relatively long (1–4 h) it is expected that these deviations will be smoothed and on the average a quasi equilibrium may be assumed. These arguments will, of course, be invalidated if the atmospheric stability is such that there is a very high ceiling (above the assumed height) with strong fumigation and/or local sources have a strong influence. By a judicious choice of sampling site and time these exceptional cases may be excluded from the investigation and the validity of the results would not be affected.

The upper bound of the mean particle residence time may be determined as follows. The particle removal velocity is composed of several components [equation (15)].

$$U_R = U_t + U_d + U_x \tag{15}$$

where U_t = Particle terminal settling velocity

U_d = [illegible] velocity

U_x = Particle removal velocity due to reaction and any other effects.

It is assumed that U_t is always in the direction of U_R, but that other removal velocity components may be in opposing directions for short durations of time. In the exploratory study reported here, averages over appreciable time periods were of interest, i.e., hours. Hence it is reasonable to assume that:

$$U_R \geqslant U_t \tag{16}$$

Thus, the range of residence time, as calculated from C_0 and J_0, and the limiting values based on U_t, are given by equation (17).

$$\frac{H}{2}\frac{1}{U_t} \geqslant \tau \geqslant \frac{C_0}{J_0}\frac{H}{2}. \tag{17}$$

EXPERIMENTAL METHODS

In each test, a membrane filter (Millipore HA) was exposed to collect settled particles for four hours in the horizontal plane on the roof of the Graduate School of Public Health, University of Pittsburgh. The ground level concentration of suspended particulate matter was measured by simultaneously sampling at 6.5 l.min^{-1} adjacent to the filter with another membrane filter (Millipore HA) oriented in the vertical plane for one randomly selected hour within the four hour interval. In order to minimize the effects of cross-wind a leak tight filter holder (GUSSMAN *et al.*, 1962) was modified by gluing to the filter holder face a 12 cm dia., $\frac{1}{4}$ in. thick plate with a 7.5 mm i.d. hole in its center. This geometry of the collecting filter will not introduce sampling velocity distortions for 40 μm dia. particles in winds up to 40 km h^{-1}, according to calculations by Davies (DAVIES, 1968). After the sample was collected, particles on each filter were microscopically sized and counted, using two magnifications and overlapping the calculated concentrations. The magnifications used were 20× ocular, 1.25 optovar in both cases, 25× and 63 objective for larger and smaller fractions, respectively.

The ground level measurements were not made due to the expected distortion from the nearby buildings. However, for the purpose of the study, measurements at the roof level should be sufficiently accurate to estimate the residence time of the particles.

The sampling periods were selected when there was no precipitation for at least three days prior to the sampling period and during the sampling period relative humidity was less than 70 per cent.

VERIFICATION OF SAMPLING METHOD

In order to investigate the effect of sampling surface on the flux measurements five independent tests at a different locality were conducted. On four corners of a square of approximately 1 m a Whatman 40 filter paper, a glass slide and two Millipore HA filters diagonally opposing each other were placed. These surfaces were exposed to the atmospheric dust for a duration of four hours and the deposited particles were sized microscopically. Due to the difficulty in assessing the size distribution on Whatman paper this verification was performed on particles larger than 2.0 μm only. One of the millipore filters was assumed to be a correct evaluation and the others were compared to that filter. The results show that the deviations are well within experimental error and a trend does not seem to exist between sampling media (TABLE 1). The use of membrane filters is well justified in terms of the ease in the particle size assessment and handling. Although glass slides are easy to use in conjunction with the microscope, handling and cleaning presents some difficulties.

TABLE 1. COMPARISON OF DIFFERENT COLLECTOR SURFACES IN THE MEASUREMENT OF FLUX

Collector surface	Test No.	Comparative flux measurement (Membrane filter I taken as 1.00) Particle diameter 2.0–4.8 μm	4.8–9.6 μm	> 9.6 μm
Membrane II	1	0.98	1.05	0.90
	2	1.08	0.96	1.00
	3	1.12	1.00	1.02
	4	0.90	0.96	1.09
	5	0.95	0.99	1.02
Whatman 40	1	1.09	0.98	1.10
	2	1.12	1.00	1.09
	3	0.90	1.11	0.90
	4	0.91	1.05	0.92
	5	1.10	1.02	0.91
Glass slide	1	1.00	0.90	0.90
	2	0.95	0.98	0.87
	3	0.92	1.09	0.90
	4	1.08	1.01	1.09
	5	1.13	0.95	1.10

RESULTS

Five preliminary tests were performed. Test data were reduced in the following way:

(1) From microscopic sizing and counting of particles on each filter a ground level particle concentration and a ground level particle flux were defined for each particle size group.

(2) The particle size data were then reduced to particle aerodynamic equivalent size, d_e, using a mean particle shape factor of 1.29 (STEIN *et al.*, 1969).

(3) From C_0 and J_0, U_R (0) and τ were calculated by assuming that the maximum aerosol height, H, was 2000 m above the ground level.

(4) $U_R(0)$ was then compared to particle terminal settling velocity in calm air, U_t, based on calculated values of d_e.

The results of these procedures are shown in FIGS. 2–4.

FIG. 2. Re

DISCUSSION OF RESULTS

The preliminary results indicate that the mean residence time of submicron particles in the absence of precipitation is on the order of 10^2–10^3 h. Particles in the aerodynamic equivalent diameter range 10 μm to 1 μm have residence times of 10–100 h, respectively. These times are long compared to those reported by McDonald (MCDONALD, 1961); but compare favorably with Junge's results (JUNGE, 1963). McDonald utilized National Air Sampling Network data to calculate residence time. These data included the removal effect by rainfall. As expected, the effect of precipitation appears to be large.

The consistent lack of particles larger than approximately 20 μm in statistically significant numbers indicates that the mean residence time of these particles is on the order of, or less than, the transport time necessary for their arrival at the sampling site. Furthermore, for particles greater than 20 μm, the upward draft would have to be

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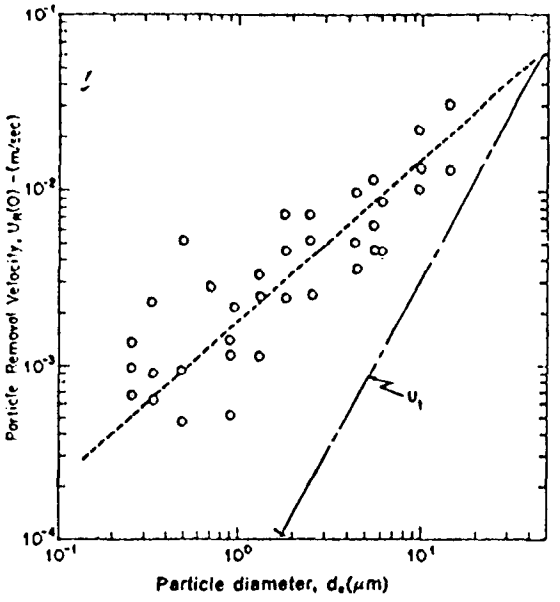

FIG. 2. Relationship between calculated particle removal velocity and particle size (d_e), calculated from measured particle projected area diameters.

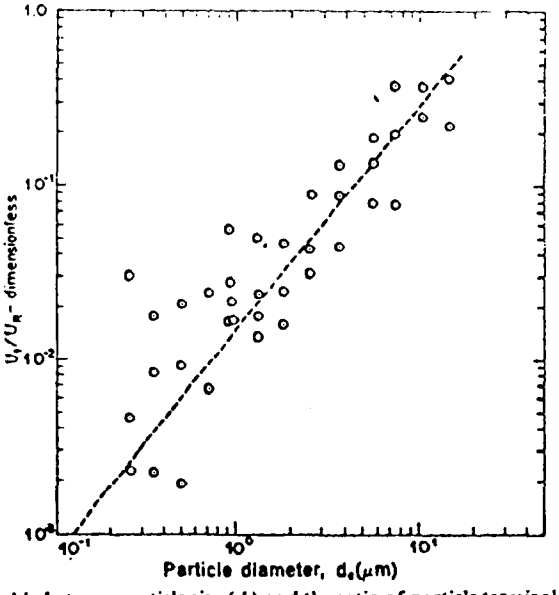

FIG. 3. Relationship between particle size (d_e) and the ratio of particle terminal settling velocity to particle removal velocity.

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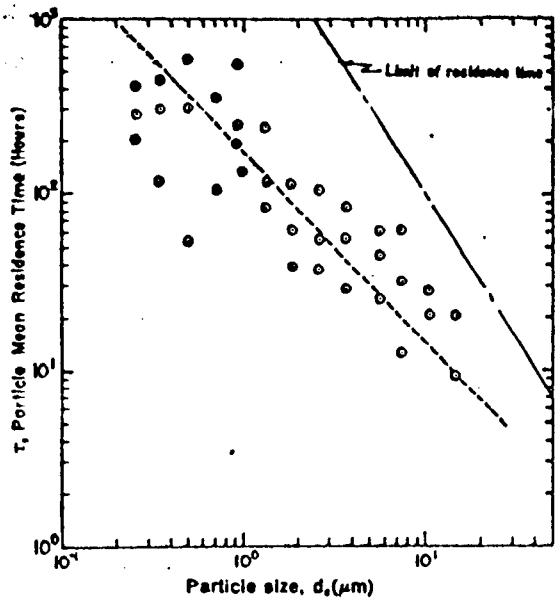

FIG. 4. Relationship between particle mean residence time and particle size.

about 0.1 km h^{-1} in order to distribute them up to the assumed ceiling of 2 km above the surface. This will lead to a further complexity of calculating the residence time of the large particles.

Because the concentration of particles in precipitation was not determined in this study, we cannot compare the calculated dry and wet removal particle velocities.

The results discussed above are based on a model with severe restrictions and assumptions pertaining to the atmospheric ceiling height for particle dispersion and to the nature of the distribution of particles within this layer. However, the results offer a first order approximation to the residence time of particles injected into an urban atmosphere.

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RESIDENCE TIME OF PARTICLES IN URBAN AIR*

THE RESIDENCE time of 0.1 to 1.0 μm radius particles in urban air seems to be on the order of one day based on several years experience in measuring atmospheric turbidity by optical means. This qualitative conclusion arises from observing the diurnal variation in turbidity which normally increases during the day but decreases during the night. The increase is due to two factors: (1) the direct production of particles by human activities and by turbulent lifting of natural particles, and (2) by production of particles by photochemical reactions of trace gases in the air. Both of these processes are at least reduced at night. Since the turbidity decreases then, there must be a removal mechanism, at least as rapid in the dark as in the light; possibly more rapid, due to reduction in atmospheric turbulence. ESMEN and CORN correctly called this mechanism "dry removal". The observed changes in turbidity overnight in conditions of "dry removal" only lead to a mean lifetime for this size range of particles of 17–39 h. The day-time mean lifetime may be slightly longer due to meteorological parameters, so that an estimate of 10^1–10^2 h would seem warranted. The difference between this result and the 10^2–10^3 h given by ESMEN and CORN may arise from two reasons: (1) the assumption of steady state made by them, and (2) the low collection and optical detection efficiencies of these small particles.

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AUTHORS' REPLY

Although atmospheric turbidity measurements show a diurnal variation, we fail to see a contradiction between our results and the diurnal variation in turbidity. This can be shown by a few simple calculations. The turbidity is defined as (DAVIES, 1966):

$$K = n a E l \tag{1}$$

where

n = number of particles
a = particle projected area proportional to particle diameter squared
E = particle extinction coefficient
l = thickness of light path.

We now let the turbidity at nightfall be $K(0)$, and investigate the value of $K(t)$ after a time t, assuming no generation and greatly reduced mixing. This can be accomplished by investigating the behavior of an exponential decay model for $K(t)$. Our results indicate that

$$\tau \simeq 150/d \tag{2}$$

where

d = particle aerodynamic diameter in microns
τ = mean residence time in hours.

From the exponential model we can calculate the $n(d,t)$ variation of particle count

$$n(d, t) = n_0(d) \exp(-t/\tau) = \exp\left(-\frac{td}{150}\right). \tag{3}$$

From equations (1)–(3) we get

$$\frac{K(0)}{K(t)} = \exp\left(\frac{td}{150}\right). \tag{4}$$

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Integrating this value between $d = 0$ and $d = 20$

$$\frac{\overline{K(0)}}{\overline{K(t)}} = \frac{1}{20}\int_0^{20} \exp\left(\frac{tx}{150}\right) dx = \frac{150}{20t}\left[\exp\left(\frac{20t}{150}\right) - 1\right]. \quad (5)$$

To make the calculations easy we can take $t = 7.5$ h. Hence

$$\overline{K(0)} = 1{\cdot}7\ \overline{K(7{\cdot}5)} \quad (6)$$

This indicates a 70 per cent reduction in turbidity. Considering the other removal mechanisms, partial generation and further complicating atmospheric effects, this figure is certainly reasonable. On the other hand, if we take τ approximately 1/4 of the values reported here, as suggested by FISCHER we will obtain (by similar calculations)

$$K(0) = 16\ \overline{K(7.5)} \quad (7)$$

This value seems excessive. Therefore, we feel that our results are at least a good first approximation to the true value of the dry removal residence time under equilibrium conditions.

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TECHNOLOGY

Twenty million tons a year of particulate pollutants are emitted into the atmosphere from U.S. stationary industrial sources, according to a comprehensive 15-month study by Midwest Research Institute (MRI), Kansas City, for the Environmental Protection Agency (EPA). MRI's study—to be released by EPA to the public this spring—lists 18 major industrial categories with more than 15,000 tons of particulate emission per year, and finds the largest source to be electric utility and industrial coal-fired power plants (5.7 million tons yearly). Among other industries cited are iron and steel (fourth, 1.4 million tons per year), paper and wood processing (sixth, 580,000 tons), primary nonferrous metals (eighth, 470,000 tons), and fertilizer processing (10th, 328,000 tons). The study, a 600-page handbook, compiles recent information on particulate emissions—including their physical and chemical natures—and on control equipment.

Unleaded gasolines may not exhibit the disadvantages claimed by critics, according to Harold R. Taliaferro and Dr. Lawrence T. Wright of American Oil Co., and Russell C. Mallatt of Standard Oil Co. (Ind.). Claims have been made that use of higher amounts of aromatics in unleaded fuels to increase octane rating might increase hydrocarbon emissions and worsen smog problems. However, Mr. Taliaferro told the 137th annual meeting of the American Association for the Advancement of Science in Chicago last week, analyses of the new unleaded gasolines show that they are similar in aromatic content to the current leaded gasolines. Exhaust-valve recession can occur with some engines, but it is minimized by adding a phosphorus-containing additive. Unleaded gasolines cost 2 to 3 cents per gallon more than leaded regulars, but studies show that the unleaded fuels give maintenance savings of 2 to 5 cents per gallon.

Controlled thermonuclear fusion as a source of electric power may be a step closer to reality with successful testing of a 13-ton, 6-foot-diameter, superconducting magnet at Lawrence Radiation Laboratory, Livermore, Calif. The magnet—largest and most powerful of its kind—has already reached a field strength of 55 kilogauss at its windings and 13.5 kilogauss at the center of its magnetic field, according to Dr. Carl Henning, Dr. Charles Damm, and Dr. Richard Post. The Lawrence scientists expect the magnet to reach design field strength of 75 kilogauss at the windings and 20 kilogauss at the center in the near future. The magnet will be used to capture the nuclei of light atoms heated to about 300° K. and to "contain" them while they fuse (C&EN, Oct. 5, 1970, page 32).

Dynachem Corp. has developed a photoresist for printed circuit board production that develops in—and is stripped by—aqueous solutions. The Santa Fe Springs, Calif., firm supplies the negative working photopolymer—called Laminar A—in roll form, sandwiched between protective covers of polyethylene and polyester, much like Du Pont's Riston (C&EN, Nov. 30, 1970, page 48). By eliminating use of organic solvents in the developing and resist-removal steps, Laminar A promises easier, safer handling, reduced pollution, and lower costs, according to Dynachem.

A new programable electronic calculator for engineers and scientists has been introduced by Wang Laboratories, Inc., Tewksbury, Mass. The Wang model 500 fits into the middle of calculator and computer ranges, and expands to handle many computer jobs, but its price starts at $2700, Wang says.