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#### Abstract

The continuous sulfur coating of urea to produce controlled-release fertilizer was studied by TVA in a $300-\mathrm{lb}-\mathrm{per}-\mathrm{hr}$ pilot plant. Molten sulfur and a molten wax sealant (containing coal tar microbicide to prevent degradation in soil) were sprayed in successive layers onto a rolling bed of heated fertilizer. The waxed material was cooled and dusted with conditioner to obtain a free-flowing product. A typical coated product contained $17 \%$ sulfur, $3 \%$ wax, $0.2 \%$ coal tar, and $1.8 \%$ conditioner. The


substrate dissolution rate in water at $100^{\circ} \mathrm{F}$ was $15 \%$ in 1 week and $0.5 \%$ per day during the second week. The operating conditions that represent departures from previously reported batch laboratory work were: continuous operation; use of several spray nozzles rather than one; and higher atomizing air pressure in the sulfur spray nozzles. Estimated costs should permit use on long-term crops in some areas.

Increasing the efficiency of fertilizers, particularly nitrogenous fertilizers, by controlling the rate of release of nutrients to match plant requirements, has been studied by many investigators for nearly 30 years. The hope of reducing or eliminating nutrient losses due to leaching, chemical decomposition, soil fixation, and luxury consumption has sustained this long study. The fact that no such controlledrelease materials economically suitable for general farm use are yet commercially available attests to the difficulty of the task of combining technical, agronomic, and economic feasibility in a single product. TVA is engaged in the development of one approach to controlled release, that of covering individual fertilizer particles with a permeable coating. The results of initial tests made batchwise in laboratory equipment have been published (Blouin and Rindt, 1967a,b; Rindt et al., 1968). The process as described in these publications consisted of preheating the fertilizer substrate, spraying the rolling substrate first with molten sulfur and then with molten wax in a rotary drum, and then cooling the product and coating it with a conditioning agent. The more important factors defined in this laboratory-scale study were as follows. (A) Neither wax (up to $15 \%$ ) nor sulfur (up to $40 \%$ ) alone formed effective coatings; only sulfur coatings sealed with a wax film were effective. The wax film sealed the microscopic cracks and pores in the sulfur, thus greatly reducing the rate of penetration by soil moisture. (B) The wax sealant must have some mobility, even at ambient temperatures; a microcrystalline petroleum wax having an oil content of about $10 \%$ and a melting point of about $165^{\circ} \mathrm{F}$ was the best of many tested. About $3 \%$ wax (product weight basis) was found to be optimum. (C) Preheating the fertilizer substrate to $150-170^{\circ} \mathrm{F}$ prior to spray-coating with sulfur gave a smooth, glasslike coating; lower degrees of preheat gave rough, less effective coatings. (D) Soil microorganisms attacked the wax sealant, thus destroying coating effectiveness; coal tar added to the wax proved to be the most economical microbicide for preventing this attack. About $0.5 \%$ coal tar (product weight basis) was satisfactory.

The present report covers the work in a small continuous pilot plant. Experience with the small unit $(300-1 \mathrm{~b} / \mathrm{hr}$ capacity) provided design information for a 1 -ton-per-hr unit that is expected to be in operaton at TVA in 1971.

[^0]Products from the small unit are being tested in the United States and 22 foreign countries.

## PILOT PLANT STUDY

Description of Pilot Plant. A schematic flowsheet of the pilot plant is shown in Figure 1. The fertilizer particles (substrate) were fed to a 3 - ft diameter by $4-\mathrm{ft}$ long rotary drum, which was divided into three compartments with retaining rings. The first compartment ( $1-\mathrm{ft}$ long) was the preheating section where the substrate was heated to about $160^{\circ} \mathrm{F}$ with jets of hot air ( $280^{\circ} \mathrm{F}$ ). Molten sulfur ( $300-310^{\circ} \mathrm{F}$ ) was applied to the preheated substrate in the next compartment ( $2-\mathrm{ft}$ long) by means of three constant-bleed, airatomizing spray nozzles; the sulfur sprays were directed against the surface of the rolling substrate bed. The sulfur fused into a smooth, glassy shell around each individual substrate particle. Details of the operation and adjustment of the spray nozzles are given later. The sulfur-coated particles then flowed to the third compartment (1-ft long) where they were coated with molten wax sealant containing about $8 \%$ coal tar (microbicide). The wax was distributed by means of a small manifold having four outlet tubes. At the elevated temperatures involved ( $150-160^{\circ} \mathrm{F}$ substrate, $200-220^{\circ} \mathrm{F}$ wax) the oil-like wax was distributed evenly throughout the rolling bed of sulfur-coated particles. The coal tar was pumped at room temperature to the wax distributor, where it mixed with the wax just prior to application.
The waxed material was then discharged into a second rotary drum (2-ft diameter $\times 3$-ft long) in which the rolling bed of material was cooled to about $100^{\circ} \mathrm{F}$ and then conditioned (about $2 \%$ clay or diatomaceous earth) to offset the tackiness of the wax sealant. Cooling prior to conditioning was required to prevent the conditioner from embedding in the hot wax, thus becoming ineffective. Following conditioning, the product was screened on a 4 -mesh screen to remove the small amount of oversize, and then bagged.

Each feed stream-substrate, sulfur, wax, microbicide, and conditioner-was metered into the system with volumetric feeders and weigh tanks (disk feeders for solids; positive displacement, remote head diaphragm pumps for liquids). Temperature recorder controllers were provided for heated feed streams and for hot air ( $300^{\circ} \mathrm{F}$ ) to the spray nozzles and preheat section. A photograph of the pilot plant is shown in Figure 2.
In the early phases of the plant operation, difficulties were


Figure 1. Schematic flow diagram of sulfur-coated urea pilot plant


Figure 2. Sulfur coating pilot plant
encountered with freezing of sulfur in the small feed lines and stoppage of both sulfur and wax pumps. It was found that complete jacketing and heavy insulation of the sulfur lines alleviated the freezing problem. Pump stoppage was virtually eliminated by installing $100-$ mesh stainless steel screens in the pump inlet lines to keep trash out of the check valves.
Both the coating drum and the cooler-conditioner drum were adequately vented with a duct-blower system to prevent escape of sulfur mist, wax-coal tar fumes, and conditioner to the operating area.

Description of Substrates, Coating Materials, and Products. Substrates. A list of the types of fertilizer substrate that have been coated in the pilot plant is given in Table I. Also included are some additional fertilizer substrates that were coated in the small-scale work and which could undoubtedly be utilized in the pilot plant.

Urea in various forms-spherodized, pan-, and pugmillgranulated, and air-prilled-was the predominant substrate in this work, primarily because there appears to be a greater agronomic need for controlled-release nitrogen, and the high initial nutrient content of urea ( $46 \% \mathrm{~N}$ ) permits the use of less coating per unit of nitrogen than would ammonium nitrate or ammonium sulfate. In addition to the air-prilled urea shown in Table I, somewhat larger prills, obtained by screening on 10 - or 12 -mesh screens, were used to demonstrate the effect of particle size. Screen analyses of these two materials are given in the tabulation below; the rapid screening did not remove all of the undersize.

| Screen | Tyler mesh, \% |  |  |
| :---: | :---: | :---: | ---: |
| used | $-6+10$ | $-10+12$ | -12 |
| 10 mesh | 67 | 28 | 5 |
| 12 mesh | 36 | 50 | 14 |

Other fertilizer substrates tested in the pilot plant were granular flotation and coarse recrystallized potassium chloride and granular ammonium phosphate nitrate ( $30-10-0$ ).

In the case of the ammonium phosphate nitrate, the coatings were ineffective because of breakage due to the crystal phase change of the ammonium nitrate at about $90^{\circ} \mathrm{F}$ with resulting expansion. The same objections would apply to pure ammonium nitrate; in addition, the hazards involved in applying sulfur and wax to ammonium nitrate would be too great. Potassium chloride presented no chemical or phase change problems.

Coating Materials. The elemental sulfur used in the pilot-plant work was commercial bright, run-of-mine sulfur.


Figure 3. Cross-sectional photomicrographs of sulfur-coated urea particles

Earlier work (Rindt et al., 1968), however, had shown that both the bright and dark grades of commercially available sulfur could be used interchangeably. Plasticizers (extenders) were not used because it was found that they offered no significant advantage over unplasticized sulfur.
The sealant used in all pilot-plant work was a commercially available microcrystalline wax having an oil content. (ASTM) of about 8 to $10 \%$ and a melting point of $167^{\circ} \mathrm{F}$. The material is a byproduct of petroleum refining.
The microbicide used to combat soil microorganism attack of the wax sealant was a commercially available coal tar that was in the form of a viscous liquid at ambient temperature. Originally about $16 \%$ coal tar (based on coal tar-wax mix) was added to the wax, but later about $8 \%$ was found to be adequate.

Diatomaceous earth was used almost exclusively as the conditioning agent in the pilot plant work. However, this was only because it was the most readily available conditioner. Others that have been found to be effective include kaolin clay, talc, and vermiculite.

Product Description. As is apparent from the process description, the sulfur coating is a three-component, laminated coating consisting of first a layer of elemental sulfur, then a thin film of microcrystalline wax, and finally a layer of conditioner. A cross-sectional photomicrograph of typical coated products is shown in Figure 3.

Table I. Fertilizer Substrates Used in Sulfur-Coating Process

Substrate dissolution in water at $100^{\circ} \mathrm{F}$, wt \% in 5 or 7 days

| $33-0-0$ | 27 |
| :--- | :--- |
| $36-0-0$ | 12 |
| $34-0-0$ | 23 |
| $36-0-0$ | 20 |
| $39-0-0$ | 16 |
| $0-0-37$ | 23 |
| $0-0-37$ | 15 |
| $28-4-0$ | 4 |
| $18-6-7$ | 3 |
| $18-20-0$ | 5 |

${ }^{a}$ Granulated in rotary drum. ${ }^{b}$ Crystalline (21-24-0).


Figure 4. Effect of time on substrate dissolution in water at $100^{\circ} \mathrm{F}$

The experimental range of contents (weight percent of total product) of the various components was as follows: sulfur, $10-25 \%$; wax, $3 \%$; microbicide, $0.25-0.50 \%$; and conditioner, $1.5-2.0 \%$. As a result, the range of nitrogen contents of coated urea prills or granules was about 31 to $39 \%$ N.

Laboratory Evaluation of Products. In order to compare the characteristics of products made by different coating procedures, laboratory analytical procedures were developed to determine both the amount of coating and its effectiveness. For coated urea, the coating was taken to be the water-insoluble portion of the sample. It was determined by crushing a $10-\mathrm{g}$ sample and letting it stand in 175 ml of water at room temperature for 24 hr . The insolubles were filtered, washed, dried, and weighed. For substrates that contain waterinsoluble material, an appropriate correction would be made. The dissolution period can be reduced to about 2 hr by maintaining the sample at a temperature of $150^{\circ} \mathrm{F}$.

The relative effectiveness of the coating in controlling the rate of substrate dissolution was determined by immersion of a $50-\mathrm{g}$ sample in 250 ml of water at $100^{\circ} \mathrm{F}$ for specified periods of time. The amounts of substrate in solution were determined by the specific gravity (pycnometer) of the solution; calibration of specific gravity by chemical analysis was necessary for each type of substrate. Data from two typical samples of coated urea are plotted in Figure 4.

The rate of dissolution decreased rapidly with time. The relatively high dissolution in the initial 7 -day period is a measure of the proportion of nitrogen tha: is readily soluble as a result of imperfect coating; the 7 - to 14 -day period, or longer, is used to evaluate the long-term dissolution rates in a relative manner among various materials.

The effect of the temperature of the water used in the dissolution tests on the dissolution rate is shown in Table II.

As might be expected, the amount of substrate dissolved in a given time period increased as the water temperature increased from 32 to $100^{\circ} \mathrm{F}$; the increase was almost twofold for $14-$ day periods. In the routine laboratory evaluation procedure, $100^{\circ} \mathrm{F}$ was used to accelerate the tests; periods of 7 and 14 days generally were used.

General Operating Conditions. In the initial pilot plant work, only one sulfur spray nozzle was used. It was apparent that the product quality did not approach that of small batch coating. It was reasoned that the very short retention time under the spray in the continuous unit (see below) did not permit an even distribution of the sulfur; consequently, a second spray nozzle was added, and later a third. In each case an improvement in distribution of sulfur and in coating quality was noted (see discussion under "Effects of Principal Process Variables"). The number of sprays was not increased further because of space 1 mitations in the coating drum.

The retention times of the fertilizer feed in the various sections of the coating drum were based on exploratory tests in a much smaller drum. The pilot plant drum was sized to give an overall retention time of about 6 min at a $300-\mathrm{lb} / \mathrm{hr}$ production rate with a $2.5 \%$ loading factor ( $2-\mathrm{in}$. bed depth). This loading was maintained by retention ring adjustments, even at higher feed rates, because a significantly deeper bed led to fines segregation in the relatively quiet center or "eye" of the rolling bed, thus giving uneven distribution of the sulfur on the substrate. Although the drum speed could be varied, it was held at 12 rpm for maximum rolling action and mixing of the bed without excessive carryover of the feed.

Only very small flights (1-in. triangular, wooden) could be used to minimize fines segregation; deeper flights would have given better mixing but would have become coated with sulfur.

Temperatures of the various feed streams that were found to give the best results in the small-scale work were retained in the pilot plant. The various temperature levels used in virtually all pilot plant work were as follows.

| Material | Temperature <br> range, ${ }^{\circ} \mathbf{F}$ |
| :--- | ---: |
| Substrate | $155-165$ |
| Sulfur | $295-300$ |
| Preheat air | $280-300$ |
| Atomizing air | $300-305$ |
| Wax (plus coal tar) | $200-220$ |
| Coal tar | Room |
| Product cooler discharge | $90-100$ |

Effects of Principal Process Variables. Coating Weight (Thickness). The rate of diffusion of the solubilized substrate through the coating is a function of the thickness of the laminated coating; thicker coatings have lower diffusion rates; assuming that the "quality" of the coatings is the same; i.e., the same coating conditions are used in each case. For a given substrate, the coating thickness is a function of the weight percent coating. Substrate dissolution

Table II. Effect of Water Temperature on Substrate Dissolution Rate

| Sample | Total coating, wt \% | Substrate dissolution, wt \%, in water at |  |  |  |  |  |  |  |  |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  |  | $32.4{ }^{\circ} \mathrm{F}$ |  |  | $81^{\circ} \mathrm{F}$ |  |  | $100^{\circ} \mathrm{F}$ |  |  |
|  |  | 7 day | 14 day | Daily | 7 day | 14 day | Daily | 7 day | 14 day | Daily |
| 1 | 23 | 8.4 | 12.2 | 0.5 | 12.8 | 17.3 | 0.6 | 17.6 | 21.3 | 0.5 |
| 2 | 23 | 8.0 | 13.4 | 0.8 | 14.1 | 17.8 | 0.5 | 18.1 | 21.1 | 0.4 |
| 3 | 23 | 6.0 | 11.9 | 0.8 | 12.2 | 16.1 | 0.6 | 17.2 | 20.2 | 0.4 |

Table III. Effect of Atomizing Air Pressure on Product
Dissolution Rate

| Average total coating, wt \% | Substrate dissolution in water at $100^{\circ} \mathrm{F}$, wt $\%^{a}$ |  |  |  |  |
| :---: | :---: | :---: | :---: | :---: | :---: |
|  | Atomizing air pressure, $\mathrm{psig}^{\text {b }}$ |  |  |  |  |
|  | 30 | 45 | 60 | 80 | 100 |
| Urea prills (95\%-6+14 Mesh) |  |  |  |  |  |
| 23 | 70 | 60 | 40 |  |  |
| 28 | 57 | 42 | 31 |  |  |
| 31 | 39 | 28 | 27 |  |  |

Spherodized urea granules ( $99 \%-6+10$ Mesh)

| 20 | 16 | 6 | 4 |
| :--- | ---: | ---: | ---: |
| 23 | 9 | 3 | 3 |

${ }^{a}$ In 5 days for prills and 7 days for granules. ${ }^{b}$ Three spray nozzles.
rates, both initial ( 7 day) and daily, are correlated with weight percent coating, rather than coating thickness, as a matter of simplification.
The variation in dissolution rate with coating weight is shown in Figure 5. The substrate was spherodized urea (-6 +10 mesh), which was the most commonly used feed in the pilot plant work because of the beneficial effects of the large, smooth granules on the coating quality.

The average initial 7 -day dissolution rate increased from only $6 \%$ with a $30 \%$ coating to over $60 \%$ with a $12 \%$ coating. In the same way, the average daily ( $7-14$ days) dissolution rates varied from about $0.3 \%$ per day to $1.4 \%$ per day; the daily rate (average of the second 7 days) tended to vary less with coating weight than did the initial ( 7 -day) rate.

Substrate Particle Size. Because of the spherical geometry involved, the surface:volume ratio of small particles is much greater than that of large particles. As a result, for a given coating weight (percent of product weight), the coatings on the small particles are much thinner and less effective (higher dissolution rate) than those on larger particles. This effect of particle size on substrate dissolution at equal coating weights is shown in Figure 6.

Over the narrow coating weight range of $27-30 \%$, the rate of substrate dissolution ( 10 days at $100^{\circ} \mathrm{F}$ ) increased from 4 to $27 \%$ as the particle size range decreased from $99 \%$ plus 6 mesh to $75 \%$ minus 10 mesh.

In addition to the particle size effect, the quality of particle surface also had an influence on coating effectiveness. It was found that rough-surfaced, pan-granulated urea with about the same particle size as that for the spherodized urea granules ( $-6+10$ mesh) required a $30 \%$ coating to achieve the same dissolution rate ( $14 \%$ in 7 days) as that from a $22 \%$ coating on the smooth spherodized urea granules.
In coating potassium chloride, the cubical shape of granular flotation material and the small particle size of recrystallized material necessitated heavier coatings than were required with spherodized urea.

Sulfur Spray Conditions. The single or the interacting effects of the following variables were investigated in the pilot plant: atomizing air pressure; wing tip jet:central jet air volume ratio; number of spray nozzles; cone cs. fan spray pattern; distance from nozzle to substrate bed; spray nozzle discharge rate, lb sulfur/(hr)(nozzle); average production rate. The effects of atomizing air pressure (pressure drop across sulfur spray nozzle jets) over the range 30 to 100 psig are shown in Table III.


Figure 5. Effect of coating weight on dissolution rates


Figure 6. Effect of substrate particle size on dissolution rate

With the coated prills, coating quality improved at all coating weights as the atomizing air pressure was increased from 30 to 60 psig , as evidenced by the reduction in dissolution rate from the range $39-70 \%$ to a range of $27-40 \%$. With granules, the improvement in coating quality due to pressure increase from 60 to 80 psig was more pronounced; the dissolution rates of the products decreased about two-thirds. Further increase in air pressure to 100 psig did not result in significant additional improvement.
The sulfur spray nozzles used in this work were constantbleed, external-mix sprays having diametrically opposed wing tip air jets and a central air jet concentric with the sulfur jet. The volume of air going to each type of jet was governed by two internal replaceable orifices. The ratios of these air volumes had a significant effect on coating effectiveness


Figure 7. Coating weight vs. dissolution rate as affected by various operating conditions
(dissolution rate), as shown in the tabulation below. Atomizing air pressure was 55 to 60 psig.

| Air orifice diameter, in. |  | Total coating, wt \% | Substrate dissolution in water at $100^{\circ} \mathrm{F}$, wt \% |  |
| :---: | :---: | :---: | :---: | :---: |
| Wing | Center |  |  |  |
| jet | jet |  | 7 days | 14 days |
| 0.04 | 0.04 | 28 | 26 | 31 |
| 0.04 | 0.06 | 28 | 9 | 10 |
| 0.06 | 0.04 | 29 | 5 | 6 |
| 0.06 | 0.06 | 28 | 8 | 9 |

The most effective configuration was the $0.06-\mathrm{in}$. diameter wing jet orifice with the 0.04 -in. diameter central jet orifice, since this combination yielded a product with a 7 -day dissolution rate of only $5 \%$ as compared with $8 \%$ or greater for the other possible combinations. This configuration was used in most of the work reported here.

It should be pertinent here to discuss a spray nozzle modification that proved beneficial in reducing nozzle plugging. The original nozzle contained an air-actuated, spring-loaded
valve stem, the purpose of which was to close the sulfur tip if air pressure failed. Plugging was frequent due to sticking of the valve stem and to air leakage into the sulfur tip. This problem was virtually eliminated by locking the stem in the open position with a Teflon collar, which also prevented air leakage into the sulfur tip.
The remainder of the sulfur spray variables listed at the beginning of this section were studied in a series of tests using spherodized urea granules as substrate feed. The number of spray nozzles was varied from one to three, two spray patterns (cone and fan) were used, the spray distance was varied from 5 to 9 in., the sulfur spray rate range was 15 to 130 lb per hr per nozzle, and the production rate was 200 to 470 lb per hr. The effects of these variables on the coating process were measured by determining coating effectiveness (dissolution rates).
The results are summarized in Figure 7, in which the total coating weight is plotted against the 7 -day dissolution rate in a series of curves representing six combinations of the variables above.

The most effective products (curves 1 and 2 ; lowest dissolution rates at a given coating weight) were obtained with: multiple spray nozzles-two or three nozzles gave a greater statistical chance of full coating than did a single nozzle; 5 - to 6 -in. spray distance ( 9 in . apparently allowed the sulfur spray to cool and partially solidify before striking the bed); low to moderate sulfur spray rates of up to 65 lb per hr per nozzle.
Physical Properties of Products. Two properties of the product that have a practical effect on its acceptability are its storage characteristics and its resistance to degradation by normal handling and distribution. Typical products have been tested to evaluate these properties.

Bagged storage tests were made covering periods of up to 3 months. The material (coated spherodized urea) was stored in plastic-lined bags under a stack pressure of 3.5 psi (equivalent to that at the bottom of a $20-\mathrm{bag}$ stack of $50-\mathrm{lb}$ bags) at ambient temperatures. At the end of the period, there was a light bag set but no caking or lumping. However, the dissolution rate increased significantly in this period. The data are given in Table IV.

The 14 -day dissolution rate of the heavily coated material (test 1) increased about twofold, from $2.6 \%$ to $5.4 \%$ in 3 months, which is a large percentage increase but hardly significant in terms of absolute coating effectiveness. However, the 3 -month increases in 14-day dissolution rates from lightly coated regular and forestry-grade urea (tests 2 and 3) were significant in terms of absolute coating effectiveness; the rates increased from 33 to $48 \%$ and from 26 to $46 \%$, respectively. One-month increases in 14-day dissolution losses were considerably lower than the 3 -month increases.

Table IV. Change in Dissolution Rate of Various Sulfur-Coated Fertilizers during 3 Months' Bag Storage

| $\begin{aligned} & \text { Test } \\ & \text { no. } \end{aligned}$ |  | Total coating, ${ }^{a}$ wt \% | Substrate dissolution in $100^{\circ} \mathrm{F}$ water, wt $\%$ |  |  |  |  |  |  |  |  |  |  |  |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  |  |  | As made |  |  | 1-month storage ${ }^{\text {b }}$ |  |  | 3-month storage ${ }^{\text {b }}$ |  |  | Difference in as made and 3 months |  |  |
|  | Coating |  | 7 days | 4 days | Daily | 7 days | 14 days | Daily | 7 days | 14 days | Daily | 7 days | 14 days | Daily |
| Regular spherodized urea ( $-6+10$ mesh) |  |  |  |  |  |  |  |  |  |  |  |  |  |  |
| 1 | Heavy | 29.6 | 2.1 | 2.6 | 0.07 | 3.0 | 4.4 | 0.20 | 4.7 | 5.4 | 0.10 | 2.6 | 2.8 | 0.03 |
| 2 | Light | 16.1 | 27.5 | 32.7 | 0.74 | 33.8 | 41.4 | 1.09 | 40.0 | 47.6 | 1.09 | 12.5 | 14.9 | 0.35 |
| Forestry-grade spherodized urea ( $-3+6$ mesh) |  |  |  |  |  |  |  |  |  |  |  |  |  |  |
| 3 | Light | 13.6 | 20.3 | 26.1 | 0.83 | 25.8 | 33.6 | 1.11 | 34.7 | 45.7 | 1.57 | 14.4 | 19.6 | 0.74 |

Additional studies of these changes in release characteristics are being made to more specifically define the mechanism involved as well as the means of avoiding or minimizing the changes.
It should be emphasized that these increases in dissolution rates are the maximum to be expected (bottom of conventional 20 -bag stack) and that the average increases should be much less, since there is little change with time under nonpressure storage conditions (top of stack).

Handling tests were made also. Coated spherodized urea granules were: (1) tumbled in a rotary drum for varying time periods; (2) dropped 35 ft onto a steel surface; or (3) fed several times through rotary plate or auger drill distributors; and (4) fed through a spinning plate broadcast spreader. The dissolution rates before and after treatment were measures of the effects of the treatments. The results are given in Table V.
Tumbling for 5 or 12 min actually improved the coatings; the initial (7-day) dissolution was originally $2.7 \%$, but decreased to $2.4 \%$ after 5 min of tumbling and to $2.3 \%$ after 12 min . Dropping the material increased the dissolution only slightly from 5.6 to $7.7 \%$.
The plate and screw drills caused measurable but not serious breakage of the coatings. The 7 -day dissolution increased from 3.6 to $6.4 \%$ with the plate and to $5.7 \%$ with the screw drill.

Spinner plate broadcast spreaders turn at very high speed compared with a plate drill. Some are impact types in which the fertilizer flows directly into the path of rotating vertical turbine blades which impact on the particles to broadcast them. In another type, however, the fertilizer feeds into the center of the plate, where the speed ( $\mathrm{ft} / \mathrm{sec}$ ) is much less than at the periphery; therefore, the impact is much less. In this type spreader, the 7 -day dissolution rate increased only 1 to 2 percentage points, even at maximum rpm (1000). Tests will be made with the impact-type to determine maximum allowable plate speed.
It was concluded that normal storage and handling procedures would not result in serious degradation of the products. It is possible that some restriction might have to be placed on the allowable plate rpm of the impact-type spreader.

The product ( $-6-+10$-mesh spherodized urea) was also tested for effects of exposure to humid conditions as compared with similarly exposed uncoated urea. The coatings ranged in weight from 16 to $21 \%$, with dissolution in water at $100^{\circ} \mathrm{F}$ of 4 to $21 \%$ of the substrate in 7 days. The laboratory exposure tests were made at $100^{\circ} \mathrm{F}$ and $90 \%$ relative humidity in a chamber with the air circulated by a fan. Test fertilizers were exposed in open-top glass cylinders $6.8-\mathrm{cm}$ in diameter by 20 cm deep. Moisture absorption was measured after periods of exposure of 24 and 48 hr . Depth of moisture penetration (into the cylinder) was observed as a slight change n color of the material. When the fertilizer was removed from the cylinder, it was evaluated for flowability.

Moisture absorption of the coated materials was only 5 to $12 \%$ of that of similarly exposed uncoated material; moisture penetration was about $25 \%$ of that of uncoated urea. All of the coated materials were free flowing, whereas the uncoated urea was hard caked.

## ECONOMICS

It is recognized that most new products normally go through a low-volume, high-cost developmental stage. However, in the estimated costs given here, it is assumed that a relatively large market for commercial farm use of bulk sulfur-coated

Table V. Effect of Handling Sulfur-Coated Urea ${ }^{a}$ on Its Dissolution Rate

| Type | Degree | Total coating, wt \% | Substrate dissolution in water at $100^{\circ} \mathrm{F}$, wt $\%$ |  |  |
| :---: | :---: | :---: | :---: | :---: | :---: |
|  |  |  | 7 days | 14 days | Daily |
| Simulated field application by drilling |  |  |  |  |  |
| Control |  | 25.3 | 3.6 | 4.4 | 0.1 |
| Plate drill | 1 pass | 25.3 | 5.7 | 6.4 | 0.1 |
| Plate drill | 2 passes | 25.3 | 6.4 | 7.4 | 0.1 |
| Screw drill | 1 pass | 25.3 | 5.2 | 6.0 | 0.1 |
| Screw drill | 2 passes | 25.3 | 5.7 | 6.2 | 0.1 |
| Tumbling in rotary drum |  |  |  |  |  |
| Control |  | 25.3 | 2.7 | 4.0 | 0.2 |
| Tumbling | 5 min | 25.3 | 2.4 | 3.3 | 0.1 |
| Tumbling | 12 min | 25.3 | 2.3 | 3.0 | 0.1 |
| Drop test |  |  |  |  |  |
| Control |  | 27.4 | 5.6 | 7.5 | 0.3 |
| Drop | $\begin{aligned} & 35 \mathrm{ft} \text { to } \\ & \text { steel } \end{aligned}$ | 27.4 | 7.7 | 9.1 | 0.2 |
| Spinning plate spreader, center feed |  |  |  |  |  |
| 450 rpm | Control | 28.2 | 6.8 | 8.8 | 0.3 |
|  | 1 pass | 28.2 | 8.8 | 10.9 | 0.3 |
| 1000 rpm | Control | 27.8 | 7.8 | 9.7 | 0.3 |
|  | 1 pass | 27.8 | 9.3 | 11.6 | 0.3 |

${ }^{a}$ Spherodized granular urea, $-6+10$ mesh.

Table VI. Estimated Production Cost Plus Return on Investment (Coating-Only Basis)
Plant Capacity: 500 tons/day; Total Capital Investment: \$1,700,000. Product Grade: 36-0-0-17 (S)

| Production cost | Tons/ton product | Cost, \$/ton |  | \$/ton product |  |
| :---: | :---: | :---: | :---: | :---: | :---: |
| Raw materials |  |  |  |  |  |
| Sulfur | 0.170 | 28 |  | 4.80 |  |
| Wax | 0.030 | 100 |  | 3.00 |  |
| Microbicide | 0.0025 | 80 |  | 0.20 |  |
| Conditioner | 0.018 | 50 |  | 0.90 |  |
|  |  |  |  | 8.90 |  |
| Operating costs ${ }^{a}$ |  |  |  | 4.12 |  |
| Total |  |  |  | 13.02 |  |
| Return on capital |  |  |  |  |  |
| investment (ROI) |  |  | 10\% |  | 15\% |
|  |  |  | 1.03 |  | 1.54 |
| Production cost + ROI |  |  | 14.05 |  | $\overline{14.56}$ |
| Production cost + |  |  |  |  |  |
| ROI, \$/unit N |  |  | 0.39 |  | 0.41 |

${ }^{a}$ Including interest, plant overhead, general administrative expense, and in-plant handling.
urea has already been established in some areas. On this basis, a moderately large plant, 500 tons per day of sulfurcoated urea, was used as the model for cost estimating for this process.

The estimate has been limited to production cost plus return on investment. The costs of sales and distribution vary widely in the industry and have been omitted here. The estimate is also on a "coating-only" basis; that is, the cost of the substrate also has been omitted. The results of the estimation of the production cost plus return on investment, coating-only basis, for a 500 -ton-per-day plant as part of a fertilizer complex is shown in Table VI.

The total capital investment of about $\$ 1.7$ million includes $\$ 1.0$ million for installed plant equipment, plant building, incremental storage (direct and indirect costs) plus $\$ 0.7$ million for working capital. The coating raw material
costs (\$8.90) include sulfur at $\$ 28$ per ton and wax at $\$ 100$ per ton. The operating costs (\$4.12) include labor and supervision, maintenance, supplies, analyses, utilities, depreciation, taxes and insurance, interest, plant overhead, and general administrative expense. The total production cost plus return on investment ( $15 \%$ ) of about $\$ 14$ per ton product ( $\$ 0.41$ /unit N ) would be added to the transfer (captive) cost of the substrate, plus freight and handling, plus general sales and sales administrative costs to obtain the bulk wholesale delivered price of the product.

It should be noted that the sulfur coating is slowly oxidized to $\mathrm{SO}_{4}=$ in the soil and is therefore acid forming. It is estimated that on acid soils that require liming an additional 300 lb of limestone per acre might be required to offset the physiological acidity of the sulfur when applying 200 lb per acre of nitrogen as sulfur-coated urea.

## AGRONOMICS

The agronomic response of various crops to sulfur-coated urea (SCU) has been reported by TVA agronomists in several publications (Mays and Terman, 1969a,b; Allen et al., 1968). Results or conclusions obtained are summarized below. A single application of SCU on coastal bermuda grass in the spring gave us good or better results than four (split) applications of ammonium nitrate. Excessive (luxury) consumption and excessive growth of grass forage shortly after a single application was avoided with sulfur-coated urea. Sulfurcoated urea produced better distribution of protein throughout the forage growth period. The sulfur residues oxidize slowly to plant available sulfate. Heavy surface application of SCU to grasses did not result in burning or N (decomposition) losses as with uncoated urea. Sulfur-coated urea resisted leaching and denitrification in cases of heavy rainfall and resulted in higher yields under these conditions.
When this work was begun, emphasis was given to production of products with very low initial dissolution rates. However, recent data from several experiments indicate that initial rates of 15 to $40 \%$ would be desirable for many crop
situations. Such coatings allow the early response that is needed.

It is believed that sulfur-coated urea could be used to advantage for turf, pasture, and hay production, where high leaching and decomposition losses are prevalent or for longterm crops (pineapple, sugar cane, and timber). About $50,000 \mathrm{lb}$ of sulfur-coated materials have been distributed in 36 states and 22 foreign countries for testing.

## CONCLUSIONS

The continuous sulfur-coating process for the production of controlled-release fertilizer has been shown to be technically feasible on a pilot-plant scale and is believed to be adaptable to plant-scale operation.

Agronomic tests indicated that sulfur-coated urea is an effective and useful controlled-release fertilizer. These results, together with estimates of production costs, indicate that such products could be economical by reducing overall applied fertilizer costs when applied once a season under one or more of the following situations: long-term crop requiring multiple applications of conventional fertilizers, weather or soil conditions are conducive to high leaching and decomposition losses, and luxury consumption by a crop that causes inefficient early use of nitrogen and deficiency of nitrogen later in the growing season.

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