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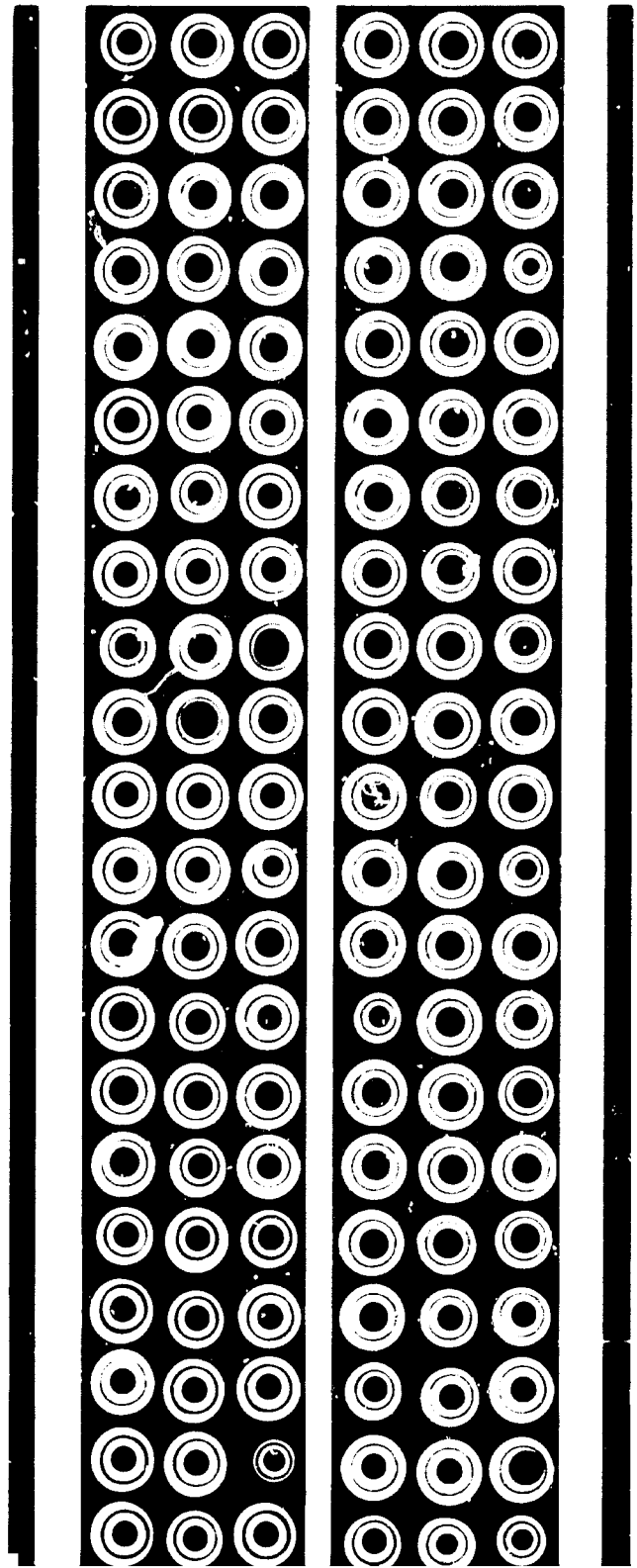

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TVA-01-74

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TVA Development of Pan Granulation Processes For Nitrogen Fertilizers

Background Work by TVA On Granulation of Fertilizers

TVA pioneered in fertilizer granulation technology in the early 1950's, and as a result nearly all of the granular NPK fertilizers in the United States are now produced by the TVA continuous ammoniator-granulator process. This comparatively simple, but quite versatile, rotary drum unit with accessory equipment for drying, cooling, and product sizing is also used in many plants throughout the world. An adaptation of this process, that was developed in pilot-plant work in 1960-61, was rapidly adopted by industry for production of granular diammonium phosphate (DAP) with a grade of 18-46-0. Minor changes in operating technique allow production of monoammonium phosphate (MAP) with a grade of about 10-54-0 or 11-52-0. Ammonium phosphates, produced almost exclusively by the TVA process, became the leading phosphate fertilizer in the United States and the world in the 1970's.

From the start of world production of ammonium nitrate, and later of urea, these straight-nitrogen fertilizers were produced mainly by prilling. Droplets of concentrated solution or melt are sprayed at the top of a tower and solidify to form small spheres as they fall downward through a column of cool air. Prilling is an established and dependable process, but there are some shortcomings.

- It is difficult to produce prills larger than about 8 mesh; they are not well suited for bulk blending, aerial application, or for coating with sulfur.
- Prills are partially hollow, and this decreases their crushing strength. Fines and dust may be formed by the degradation of fragile prills during handling and shipment.
- Very large volumes of air must be moved through prilling towers. Equipment for moving the air is expensive and requires considerable energy. Collection of dust and fumes from such a large volume of air, exiting at the top of a tall tower, is difficult and very expensive.

Because of these problems, researchers at TVA's National Fertilizer Development Center (NFDC) visualized the need for development of a granulation process that would be more flexible, more versatile in the size of granules that could be produced, and that would produce granules with better physical properties than prills.

Selection of Equipment and Process for Granulation of Nitrogen Fertilizers

TVA introduced ammonium nitrate in crystalline form in 1949 in a demonstration program. This product was good except for the comparatively small size and irregular shape of the crystals. A procedure was worked out and equipment was retrofitted in a demonstration plant for granulating the crystals in a rotary drum. They were agglomerated by use of a small amount of steam and by ammoniating a small proportion of sulfuric acid in the drum. The granular product was well rounded and had good crushing strength, but the addition of a granulation step following a somewhat tedious crystallization process would be expensive and cumbersome. The advent of bulk blending in the mid-1950's, and its rapid growth in the United States in the 1960's, created still greater interest in granular nitrogen fertilizers. TVA researchers at NFDC began test work specifically directed toward development of a fully suitable granulation process for nitrogen fertilizers.

Familiarity with the versatile rotary drum-type of

granulator, that works so well for production of granular NPK fertilizers and ammonium phosphates, naturally led to further work with this type of equipment. Granulation by use of a concentrated solution instead of by agglomeration of crystals was quite difficult. The high solubility of ammonium nitrate made granulation very sensitive to moisture and temperature. Recycle was not a very effective control for preventing formation of large pellets and balls.

The pugmill or blunger was known to not be well suited to granulation of nitrogen fertilizers because of the kneading action that induces excessive plasticity. The inclined pan pelletizer had been used for several years for agglomerating fine particles of iron ore concentrate, and fines from production of glass and ceramics. An outstanding feature is the inherent classifying action in the pan; oversize formation is minimized, and solutions can be easily placed to specifically contact the fine material. In commercial operation, the pans had been used mainly to form pellets of 10 to 19 mm (3/8 to 3/4 in) diameter—much larger than are generally suitable for fertilizers. There was some practice in granulation of superphosphates in Germany, Australia, and a few other countries, but this was an agglomeration-type of semigranulation. The uncertainties posed questions to be resolved by a pilot-plant test program that started in the early 1960's.

Adaption of the Inclined Pan for Granulation of Fertilizers (Pilot-Plant Studies)

For the pilot-plant studies, a small batch-type concrete mixer was fitted with a pan that was 15 centimeters deep and 1 meter in diameter. The drive was adapted for variable speed, and provisions were made to allow easy change of the angle of inclination. From the start of test work the inclined pan proved to be readily adaptable to efficient granulation of ammonium nitrate, and ammonium phosphate nitrate grades of 30-10-0 and 25-25-0. Good results were obtained later in granulating 30-0-0-5S ammonium nitrate-sulfate.

The granulation step was simplified, since ammoniation of the acids could be readily and efficiently completed in solution or slurry systems. The neutralized solutions could be readily concentrated to a low water content by heat of reaction and evaporation prior to feeding to the pan. The hot concentrated solutions could be sprayed easily; they flowed readily onto and coated the surface of undersize particles and recycled material uniformly. This technique of operation of the pan produced well-rounded granules, mainly by layering, as the melt or highly concentrated solution solidified. The excellent classifying action of the pan limited oversize production to a very small percentage and allowed operation with a low recycle ratio of about 2:1. In proper operation of a pan granulator, the solutions are sprayed only onto undersized particles, whereas in a drum or pugmill particles of all sizes are wetted. In this type of operation the pan granulator is utilized to take full advantage of its excellent classifying action without attempting any ammoniation in the pan (1).

Pilot-Plant Tests of Granulating High-Nitrogen Grades

The pilot-plant tests were made with the 1-meter-diameter pan at a production rate of 0.5 metric ton per hour.[1] A flowsheet of the pilot-plant system is shown in figure 1. The solutions were prepared by ammoniating acids and concentrating them through use of the heat of reaction and supplemental heat; concentration was done prior to the final neutralization step to decrease loss of ammonia, particularly from solutions containing ammonium phosphates. The final neutralized solution was of about 95%-96% concentration. It was fed through steam-jacketed lines and sprayed through a series of nozzles onto undersize particles and recycle in the upper portion of the pan. The stream of recycled material entered the pan at the top, and the sprays of hot solution were located so that the recycled material cooled the solution enough to cause it to solidify evenly on the undersize particles. When the particles reached the proper size, they were discharged continuously by the natural classifying action of the pan before any appreciable amount of oversize was produced. The action in the pan when operated in this manner is illustrated in figure 2. Data for pilot-plant tests in production of granular ammonium nitrate, ammonium phosphate nitrate, and ammonium nitrate sulfate grades are shown in table 1.

Comparatively low rates of recycle of about 2:1 were satisfactory. The material discharged from the pan was dried, cooled, and sized in conventional equipment. Granulation efficiency was quite high, so the products could be screened closely, such as 6 to 9 mesh. With proper control of operation, very uniformly sized, well-rounded granular products were produced. Drying of products of this type that con-

[1] All tonnage values in this report are metric.

Table 1. Pilot-plant tests of the production of ammonium nitrate-based fertilizers by the pan-granulation process (1 meter diameter pan)

Product	Ammonium nitrate	30-0-0 Ammonium nitrate sulfate	30-10-0 Ammonium phosphate nitrate	25-25-0 Ammonium phosphate nitrate
Operating time, hours	4.0	5.0	6.3	4.0
Production rate, t/h	0.5	0.5	0.5	0.5
Feed rates, lb/h				
Nitric acid (60%-61% HNO_3)	1,354	1,010	1,013	609
Phosphoric acid (electric furnace, 61% P_2O_5)	-	-	-	506
Phosphoric acid (wet process, 54% P_2O_5)	-	-	185	-
Sulfuric acid (93% H_2SO_4)	-	171	-	-
Water	-	76	-	-
Ammonia (gaseous) first stage	235	248	220	200
Ammonia (gaseous) second stage	1	8	-	-
Recycle fines	1,104	1,779	750	1,123
Recycle ratio	1.1	1.8	0.75	1.1
Operating temperatures, °C (°F)				
Solution to pan	168 (334)	189 (369)	170 (335)	174 (343)
Granulator product	98 (208)	98 (207)	109 (226)	106 (222)
Dryer product	120 (249)	123 (251)	138 (278)	109 (227)
Recycle fines	64 (147)	48 (117)	44 (110)	44 (110)
pH				
Neutralizer, first stage	4.0	2.7	4.7	5.0
Neutralizer, second stage	6.1	6.3	5.6	5.4
Product	5.3	5.4	-	-
Chemical analysis, solution to pan, %				
Total N	32.8	31.1	29.5	24.9
NH_3N	16.4	17.6	-	15.8
P_2O_5	-	-	10.4	28.1
$(NH_4)_2SO_4$	-	22.5	-	-
H_2O	6.5	3.5	-	-
Water insoluble	-	-	2.1	-
Screen analysis (Tyler), %				
Granulator product				
+6 mesh	38.1	15.8	12	10
-6 +8 mesh	29.8	36.0	22	41
-8 +10 mesh	19.1	38.7	56	42
-10 mesh	13.0	9.5	10	7
Screened product				
+6 mesh	0.8	0.5	0	0
-6 +8 mesh	53.3	28.5	48	28
-8 +10 mesh	45.4	67.9	48	67
-10 +12 mesh	0.4	2.7	4	4
-12 mesh	0	0.1	0	1
Chemical analysis, %				
Unconditioned product				
Total N	34.6	31.9	30.4	24.9
NH_3N	17.3	18.3	16.6	15.7
P_2O_5	-	-	11.5	28.5
$(NH_4)_2SO_4$	-	22.5	-	-
H_2O	0.6	0.03	0.5	0.2
Water insoluble	-	-	2.2	-
NH_3:H_3PO_4 mole ratio	-	-	1.23	1.12

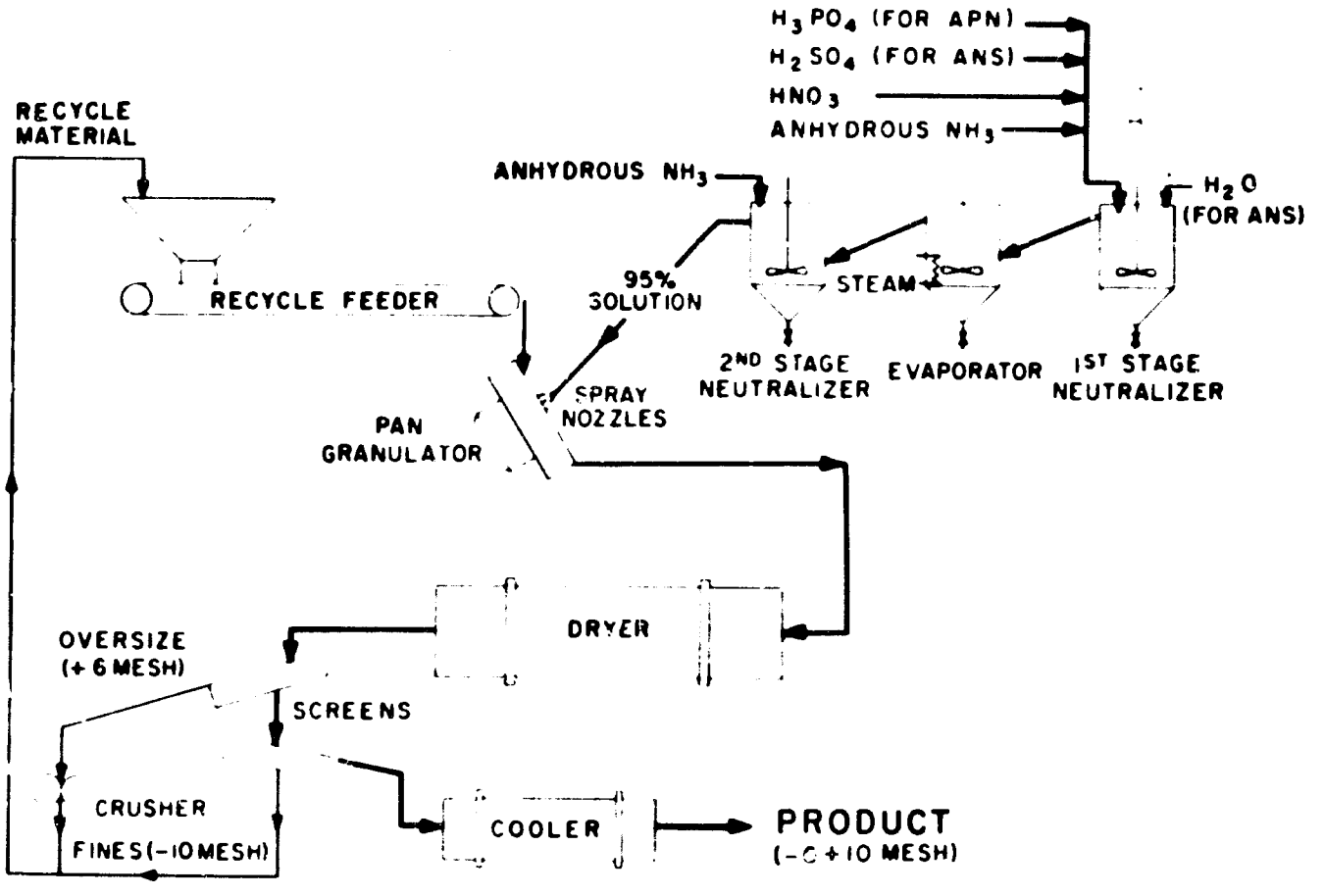

Figure 1. Flow diagram: pan-granulation pilot plant for production of granular ammonium nitrate, ammonium phosphate nitrate, and ammonium nitrate sulfate

tained large amounts of ammonium nitrate required comparatively long retention time, and use of drying gases of moderate temperature. The products had good storage properties when dried to a moisture content of 0.2%-0.4% and conditioned with about 2% of diatomaceous earth or other appropriate conditioner. (In later work using 99+% solution no drying was needed.)

Critical features for best operation of the pan granulator are listed below.

- Angle of inclination.
- Rotational speed.
- Location of solution sprays
- Concentration and temperature of solution.
- Temperature of recycle.
- Temperature of material in the pan.

Best operation was with a pan angle of 60 to 65 degrees and with a speed of about 30 revolutions per minute for the 1-meter pan. The spray header was adjustable in an upper quadrant of the pan to allow optimum positioning of the sprays with respect to movement of undersize material. The best position generally was in the quadrant of 9 to 12 o'clock with counterclockwise rotation of the pan (figure 2). Spray nozzles of the solid cone type were satisfactory. Solution temperature of 165°C (330°F) and bed temperature of 100° to 105°C (210° to 220°F) were best. The importance of sufficiently high temperature of the solution preheat in obtaining well-rounded granules was borne out in later plant-scale experience (2).

Electric-furnace phosphoric acid was used in most of the pilot-plant tests in production of ammonium phosphate nitrate (APN) grades. However, use of wet process acid of 54% P_2O_5 content in production of 30-10-0 and 25-25-0 in a few tests was completely satisfactory and gave somewhat better granulation. The concentration of nitric acid used ranged from 55% to 61%.

Although most of the pilot-plant work was with N and N-P grades, some tests were made of the addition of potassium chloride to produce grades such as 24-6-12, 17-17-17, and 21-0-21. Operation was better

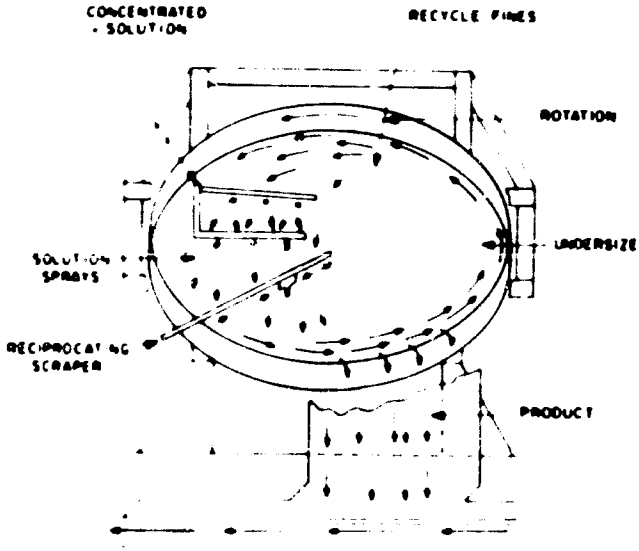

Figure 2. Sketch of plan operation

and the granules were smoother when the potash was fed to the pan with the recycle instead of being dissolved in the solution prior to spraying.

Intermediate-Scale Tests with 6- and 8-Foot Diameter Pans

After successful operation of the pilot-plant system had been demonstrated using the 1.0-meter pan, a larger pan 1.8 meters (6 ft) in diameter, together with solution preparation equipment, was fabricated and installed for tests in a demonstration plant where the regular operation was granulation of ammonium nitrate crystals in a rotary drum. Good granulation of both ammonium nitrate and APN of 30-10-0 grade was obtained at a production rate of about 2.3 tons per hour. Later, the size of the pan was increased to 2.5 meters (8 ft) in diameter to permit production of about 3.5 tons per hour in routine operation on a sidestream in the plant. Operation of the larger pans with lower loss of heat emphasized the importance of temperature of recycle on granulation efficiency and on recycle ratio required for proper control. The recycle ratio was 1.5 or 2:1 when the recycle was at a temperature of about 60°C (140°F); with recycle at 70°C (160°F) the requirement was 2.5:1 (2).

Comparative data for optimum operating conditions with pans of the three sizes used in the development work are tabulated below. These data were for production of ammonium nitrate and 30-10-0 APN.

Diameter of pan Meters	(ft)	Depth of bed cm	(in)	Angle of inclination, degrees
1	(3)	25	(9)	65
1.8	(6)	35	(14)	65
2.5	(8)	40	(16)	65

Pan speed, r/min	Capacity of pan t/h	Surface area/ ton product M^2	(ft^2)
25-30	0.5	1.2	(13)
18-22	2 to 2.5	1	(11)
15-18	3.5 to 4	1.2	(13)

The data shown above were quite consistent and appeared to give a good basis for scale-up to units of commercial size on the basis of pan surface area required. Surface area in the range of 1.0 to 1.3 square meters (11 to 14 sq ft) per hourly ton of product was indicated. Angle of inclination of about 65 degrees was best with each size of pan. The optimum rotational speed was derived from a calculation related to critical speed of a drum granulator. The following relationship was used for this calculation (3):

Speed of rotation (r/min)

$$= K_1 [1-\cos\Theta] \frac{(76.5)}{\sqrt{D\,(ft)}} \quad \text{or} \quad \frac{(42)}{\sqrt{D\,(meters)}}$$

Where Θ = angle of inclination of the pan
D = diameter of pan
K_1 = a constant derived from experimental data, usually 1.0 to 1.6

The depth required for a pan granulator of a given diameter can be estimated by the following relationship:

depth of pan = $K_2 (\sqrt{D})$

Where K_2 = constant obtained from dimensions of experimental pan.

K_2 = 1.3 to 1.6 when D = pan diameter in inches or 0.8 to 1.0 when D = pan diameter in cm.

Experience with Plant-Scale Pan-Granulation System

On the basis of successful pilot-plant tests and intermediate-scale operation, a demonstration-plant

system for production of 20 tons per hour of granular ammonium nitrate and APN products was designed and constructed by TVA.

Operation started in 1966 and continued for about 6 years producing ammonium nitrate, 30-10-0 and 25-25-0 (APN) and 30-0-0-5S ammonium nitrate sulfate (ANS). Two pans of 4.3 meters (14 ft) diameter were selected on the basis of experience with the smaller units that indicated scale-up on a factor of about 1.3 square meters (14 sq ft) of pan area per hourly ton of product. Speed of rotation and angle of inclination of the pans were variable. Each pan was driven with a 50-horsepower motor. The pans were constructed of plain carbon steel. Type 316 stainless steel was used for solution preparation equipment and for the spray system. Production capability of a single pan was found to be higher than the 10-ton-per-hour design rate; a single pan was routinely used for production rates as high as 15 to 18 tons per hour and for some periods at about 20 tons per hour.

Since the rolling action in the 14-ft pans at the maximum designed speed of 14 r/min did not appear to be so good as in the smaller test units, a change was made to increase the speed of rotation to 16 r/min and later to a fixed speed of 19 r/min. A depth of about 40 cm (16 in) for the walls of the pan was best. The 14-ft pans also gave best results when operated at an angle of about 65 degrees. Design of suitable scrapers for keeping the bottom of the pan free of buildup of solids was a problem that was solved in a series of mechanical changes. A reciprocating unit with a single tooth moving across the face of the pan at a speed of about 1.5 meters/min (5 ft/min) kept the bottom clean (figure 2). A fixed scraper prevented excessive buildup on the walls (4).

Since TVA demonstration programs with straight ammonium nitrate had been completed, only a few hundred tons of this fertilizer was produced in the demonstration plant. Operating time was sufficient to indicate that a good product could be produced. A total of a few hundred thousand tons of 30-10-0 and 25-25-0 grades of APN and 30-0-0-5S ANS was produced in part-time operation for about 6 years. These new fertilizers gave very good results in agronomic and industrial evaluation in TVA programs. The granular products were particularly well suited for effective bulk blending. The ANS proved to be a very effective source of secondary nutrient sulfur in the Western United States and Western Canadian Provinces. Industry in those areas began substantial production of these fertilizers after the TVA demonstration products had been effective in early market development.

Changes in the Demonstration-Plant System

Most of the problems in the demonstration plant were related to the two-stage neutralization system for the acids. A major change was installation of a second-stage neutralizer with greater volume and more surface area than the one installed originally. (If a system of this type were to be installed now, the equipment and operation would be simplified by use of 99.5% solution or melt of ammonium nitrate as shown later in figure 4 for granulation of urea.)

With the larger second-stage neutralizer, operation at the preferred low pH in the first stage was practical. This eliminated most of the problems that had been caused by stoppages due to crystal formation in lines, valves, and spray nozzles when operating with higher pH in the first stage. The first-stage neutralizer operated best at a pH of about 1.3 (NH_3 to H_3PO_4 mole ratio of about 1.4) and at a temperature of 150°C (300°F) to maintain maximum fluidity of the slurry in the neutralizer and concentrator. Operation of the first stage at low pH and high temperature gave a solution of higher concentration than was provided in previous operation. Ammonia losses from the concentrator and the amount of steam required for the evaporation step were greatly decreased by the low pH method of operation. Typical demonstration plant data for the preferred low pH method of operation are given in table 2.

Maximum production of 30-0-0-5S for a 24-hour period was 472 tons. The product had very good appearance, the granules had good strength, and storage properties were good when the product was treated with about 2% of diatomaceous earth conditioner (4).

Ammonium Phosphate Nitrate

Both 30-10-0 and 25-25-0 grades of APN granulated satisfactorily when 2 to 3 kg of recycle was used per kg of feed solution to the pan. Granulation was satisfactory with one pan at rates up to 23 tons per hour after the speed of the granulator had been increased to 19 r/min and the slope increased to about 70 degrees. A recycle ratio of about 4 to 1 was required to cover satisfactorily the bottom of the pan after the speed and slope were increased.

Both the low- and high-pH methods of operating the solution system were used. At first, essentially all the ammonia required for neutralization was added to the first-stage neutralizer and only enough ammonia was added in the second stage to make up for

Table 2. Typical operating data for plant-scale production of high-nitrogen fertilizers by the pan-granulation process[a]

	(30-10-0) ammonium phosphate nitrate	(30-0-0-5S) ammonium nitrate sulfate
Operating time, hours	251	268
Production rate, t/h	21.3	18.2
Speed of pan, r/min	19	19
Slope of pan, degrees from horizontal	70	70
Approximate recycle ratio	4.5 to 1	4.5 to 1
Operating temperature, °C (°F)		
First-stage neutralizer	60 (300)	145 (290)
Concentrator	145 (289)	157 (315)
Second-stage neutralizer	170 (338)	165 (325)
Recycle fines	72 (162)	57 (134)
Granulator product	92 (197)	85 (186)
Predryer product	88 (190)	75 (168)
Precooler product	38 (100)	30 (86)
Dryer product	95 (200)	95 (200)
Cooler product	42 (108)	45 (110)
Concentration of solution, %		
First-stage neutralizer	94.7	79.5
Concentrator	94.2	90.4
pH		
First-stage neutralizer	1.6	1.3
Second-stage neutralizer	5.1	6.3
Granulator product	5.0	5.5
Chemical analysis of conditioned product, %		
Nitrogen	30.6	30.9
P_2O_5	10.1	—
Sulfur	—	5.2
Moisture	0.34	0.15
Conditioner	1.9	2.2
Screen analysis of conditioned product, % (Tyler), cumulative, %		
+5 mesh	0.0	0.0
+6 mesh	0.0	0.0
+8 mesh	15.0	20.0
+12 mesh	94.0	97.0
+16 mesh	99.0	99.0
−16 mesh	1.0	1.0

[a]Low pH operation of first-stage neutralizer.

losses in the system. The first stage was operated at a pH of 5.2 and a temperature of about 140°C (280°F). Ammonia losses were very high in the concentration step at this high pH.

The preferred low-pH method of operation of the solution system was practical after the larger second-stage neutralizer was installed. The low pH of 1.5 to 1.6 (NH_3 to H_3PO_4 mole ratio of 0.65) in the first stage was regulated by automatic control of the ammonia flow rate. The return of mother liquor to the first stage was adjusted to hold the temperature of the solution at about 150°C (300°F) in that unit. The concentrator was operated at low pH, and enough ammonia was added in the second-stage neutralizer to bring the pH of the final solution to the range of 5.0 to 5.3. The concentration of the solution fed to the pan remained at about 95% and the temperature was about 170°C (340°F). The entire solution system operated satisfactorily after this change. Ammonia loss from the concentrator was essentially eliminated and total losses were decreased to an acceptable level. Typical operating data are shown in table 2, and a screen analysis profile throughout the process is shown in table 3 (4).

Particle size, appearance, and physical properties of the products were good when treated with about 2% of diatomaceous earth conditioner.

A more effective hood was installed over the pan to collect ammonia fumes and dust. A 'Type-R Rotoclone scrubber was installed for recovery of these materials as a solution which was returned to the solution preparation system. This system was easy to operate and maintain, and gave very good results.

The higher production rates obtained in the demonstration plant indicate a scale-up factor of about 1 square meter (10 sq ft) of pan surface per hourly ton of product. This is considerably more favorable than the design factor of 1.3 square meters (14 sq ft) that was based on pilot-plant experience.

Production of Granular Urea and Urea-Based Fertilizers

As the practice of bulk blending spread in the United States and production of urea grew, there was wide interest in use of this fertilizer of high concentration (46% N) in formulating bulk blends. Conventional prills are smaller than the other granular materials such as ammonium phosphates, triple superphosphate, and potassium chloride that are customarily used in bulk blending. Because of this difference in particle size distribution, uniform blends that contain urea prills are difficult to produce, and they segregate badly in handling, storage, and transport. The good TVA experience in use of a pan granulator for production of ammonium nitrate-based products led naturally to selection of the inclined pan for granulation of urea when studies were started in 1963.

Pilot-Plant Test Work In Production of Granular Urea

The first pilot-plant tests of granulation of urea in the pan involved use of solutions of 96% to 98% concentration. Granulation generally was good, but drying of the product was required to obtain product moisture content low enough for satisfactory storage properties. The need for a comparatively large and costly rotary dryer in the system would detract from

Table 3. Typical screen analysis of materials in solids system (grab samples from 30-10-0 production at low pH)

	Screen analysis, mesh % (Tyler)							
	+6	-6 +8	-8 +10	-10 +12	-12 +14	-14 +16	-16 +20	-20
Recycle fines	2	6	26	27	16	12	5	6
Product from granulator	4	11	32	22	11	11	4	5
Product from predryer	2	10	31	26	10	12	2	7
Product from precooler	0	15	64	17	2	2	0	0
Product from dryer	0	14	66	16	2	2	0	0
Product from cooler	0	14	66	15	3	1	1	0
Conditioned product	0	16	64	15	4	1	0	0

a pan-granulation process, since prilled urea by the time of this work was made with essentially anhydrous solution or melt and drying was not required. In subsequent pilot-plant tests the use of solution of 99.3%-99.8% concentration was tested in the pan. The concentrated solution was prepared by melting prilled urea. Good granulation was obtained and the granules were well rounded when the temperature of material in the pan was controlled in the proper range. No drying was required (5). A photograph of the pilot-plant pan granulator is shown in figure 3.

From the start of work in granulating urea, it was apparent that crystallization is sharper and occurs in a narrower range of temperature than had been experienced with ammonium nitrate. Ammonium nitrate, therefore, stayed plastic longer, coated more evenly on undersize in the pan, and a broader range of temperature of material in the pan was practical. It was necessary to control the temperature of urea melt, recycle, and material in the pan in a considerable narrower range to produce well-rounded and hard granules.

Both "high-temperature" and "low-temperature" methods for granulation of urea were tested in pilot-plant work (5). In "high-temperature" granulation, the temperature and quantity of recycle material were regulated to keep the bed temperature in the granulator at 110° to 120°C (230° to 250°F). This is only a few degrees below the crystallization temperature of the feed solution. When the temperature of the recycle was 50° to 58°C (120° to 135°F), a recycle to feed ratio of 1.0 to 1.5 was required to obtain best results with the "high-temperature" method. In this type of operation, the granules are somewhat plastic and attain a spherical shape by rolling in the pan. The chief disadvantage of high-temperature granulation is that an upset in recycle temperature or rate may allow the entire bed in the granulator to melt and thereby interrupt operation.

In "low-temperature" granulation, the temperature and quantity of recycle material are regulated to provide a granulation bed temperature substantially below the crystallization temperature of the feed solution. This results in thin layers of solution crystallizing on the fines and recycle material during each pass under the sprays. This method of operation is particularly well suited for producing uniformly large products desired for special uses. The bed temperature with "low-temperature" granulation was 100° to 105°C (210° to 220°F). A recycle ratio of 2 or 3 to 1 was required to control the temperature of the granulation bed in this range when the temperature of the recycle material was in the range of 50° to 60°C (120° to 135°F). One major advantage of "low-temperature" granulation is that the system is more stable and small variations in recycle will not greatly affect granulation. Granules of larger size, suitable for either forest fertilization or use as a substrate material for sulfur coating, are easily produced by low-temperature granulation (5).

The granular urea made from 99.5% solution did not require drying. This greatly simplifies the process and decreases the investment, since a large rotary dryer and accessory equipment for moving air, collecting dust, and scrubbing are not required. Even though the moisture content was no higher than about 0.1%, the granules caked in storage unless treated with 2% of activated clay-type conditioner. Later, in operation of a demonstration-scale plant, the addition of about 0.4% of formaldehyde solution to the melt in the pipeline between the evaporator and pan gave good storage properties without need for further conditioning (6).

Pilot-Plant Tests in Production of Urea-Ammonium Sulfate and Urea-Ammonium Phosphate

A few tests were made in the pilot plant in 1966 to explore production of granular urea-ammonium sulfate (UAS). Grades studied were 35-0-0-10S and

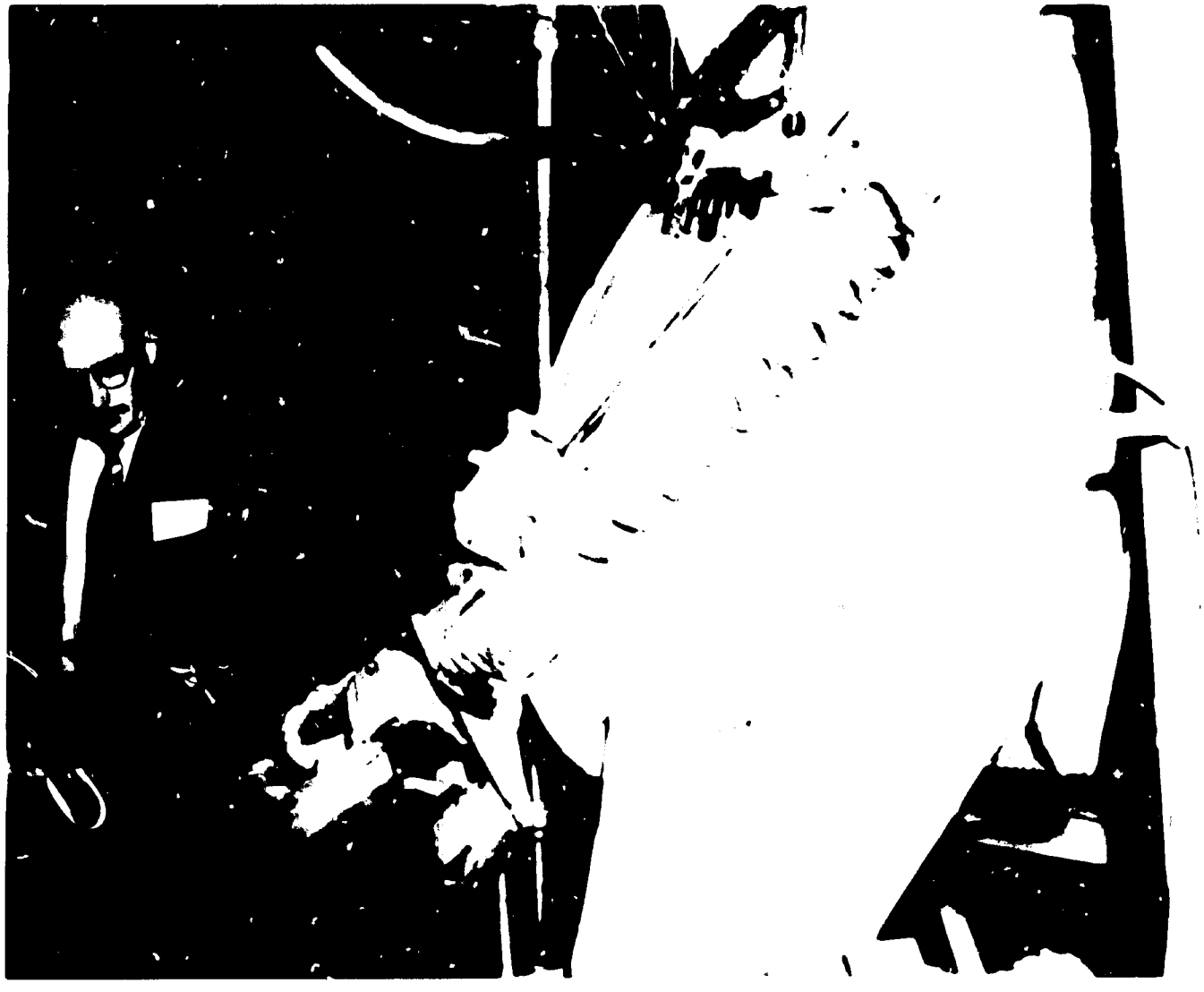

Figure 3. Pilot-plant pan granulator

40-6-0-4S. These products were shown to be very good sources of secondary nutrient sulfur. Two procedures for addition of ammonium sulfate were tested. In one, dry crystalline ammonium sulfate was added to the recycle fines stream; in the other procedure, the solid ammonium sulfate was mixed with the urea solution before granulation. Satisfactory granulation was obtained using either procedure. However, operation was simpler where the ammonium sulfate was added as a dry solid with the recycled fines; this method of operation was used in the TVA demonstration plant.

Urea-ammonium phosphate (UAP) grades 28-28-0, 33-20-0, and 35-17-0 were readily produced in the pilot-plant pan-granulation system in 1965. Concentrated urea solution (96%-98%) and ammonium phosphate slurry from a preneutralizer were sprayed as separate streams onto recycle in the pan granulator. The product was dried, cooled, and screened at minus 6 plus 10 mesh. The comparatively high moisture contents of the combined fluid feeds (20%-26% for the ammonium phosphate slurry and 2%-4% for the urea) required recycle ratios that ranged from 3 to 5. The products had excellent physical properties and stored well when dried to 0.6% moisture and conditioned with 2% of clay or diatomaceous earth. Either wet-process or electric-furnace phosphoric acid was suitable as a source of P_2O_5 for the process. This type of operation, with elimination of the drying step, would be necessary for a viable commercial pan-granulation process for UAP. Production of UAP grades using 99% urea solution was not tested.

In addition, TVA has produced UAP grades of 28-28-0 and 35-17-0 for 6 years in a demonstration

plant (15-17 t/h) by a pipe reactor-pugmill granulation process (7). A pipe reactor-rotary drum granulation process for UAP grades has shown advantages in pilot-plant work (8).

Demonstration Plant Production of Granular Urea

The TVA demonstration-plant system that had been used for production of fertilizers based on ammonium nitrate by pan granulation was adapted for production of granular urea. The system was generally simplified, since use of 99.5% solution eliminated the need for a dryer. The only new major equipment required was an air-swept, falling-film, vacuum evaporator for concentrating 75.0% urea solution from the synthesis unit to about 99.5%. A flow diagram of the process is shown in figure 4. Operation in production of granular urea at about 9 tons per hour started in late 1973; the production rate is limited by the capacity of the urea plant (6).

Good granulation was obtained during initial operation, but some conditions that were favorable to good performance in the pilot plant were difficult to obtain in larger plants. Most of the existing equipment, used earlier in production of ammonium nitrate-based fertilizers, was oversized because it had been designed for production rates of 18 to 22 t/h while the design production rate of the urea plant is only about 9 t/h. The larger equipment caused greater heat losses and made it difficult to maintain operating temperatures in the preferred range, particularly during periods when the ambient temperature was low.

Dust generation in the plant equipment has been a problem since operation began. Dust is generated in the crushers, in the granulator where the concentrated solution sprays strike the scraper and bottom of the pan, and by attrition of the granular product as it passes through the cooler and other equipment. Dust is undesirable because of the increased cost of collecting and reprocessing it in the solution system. Also, it is believed that the presence of dust in the granulator results in granules with lower crushing strength than those that would be produced if the dust was not present. Decreasing the speed of the chain mill used for crushing oversize from 1,200 r/min to 900 r/min significantly decreased the amount of dust formed in that part of the system.

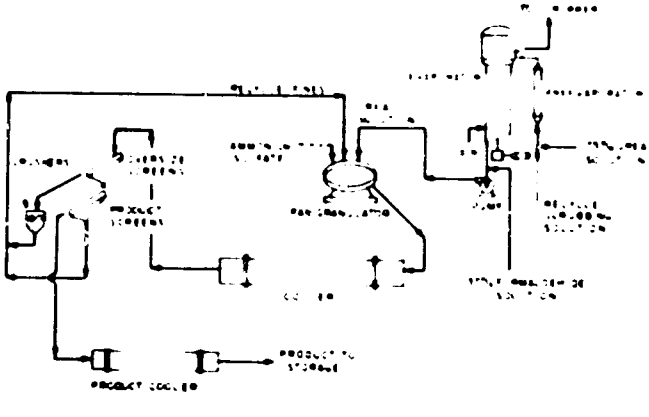

Figure 4. Flowsheet for pan granulation of urea and UAS

Operation was also improved after the following plant changes were made:

- The effective diameter of the pan granulator was decreased to 3.7 meters (12 ft) and a more efficient solution spray distributor with additional nozzles and broader coverage was installed.
- A new spray-type, direct-contact condensing scrubber was installed to recover the process material that was being discharged with the urea evaporator exhaust gases. Emissions from the evaporator then were within required environmental standards.
- A system was installed to feed formaldehyde solution to the urea melt before it is granulated. The formaldehyde is added to increase the hardness (crushing strength) of the product and to provide an incorporated conditioner. Use of formaldehyde (0.4% by wt) increased the product grade to 46% nitrogen as compared with 45% when the higher proportion of oil-wax/kaolin clay combination was used as conditioner. Incorporation of formaldehyde consistently provided granular product with good storage characteristics without the use of any external conditioning agent. The crushing strength of the granules was more than doubled, so less dust was formed by attrition.

A photograph of the 3.7 meter (12 ft) diameter pan granulator for urea is shown in figure 5.

The demonstration plant operates on a routine basis with only a moderate amount of attention when making products for TVA's fertilizer demonstration programs with industry and agriculture. Main uses are in bulk blending and for direct application. Close control of process conditions and more operator attention are required when a particularly smooth and spherical product is required for special uses, such as substrate material for sulfur coating (9, 10, 11). Some additional minor equipment changes are planned which should allow the plant to operate with less at-

Figure 5. Demonstration plant pan granulator

tention. Typical operating data are shown in table 4.

The natural classifying action of the pan granulator results in a product with a large proportion of the granules in the size range that matches major commercial granular products, such as diammonium phosphate, granular potassium chloride, and triple superphosphate, used in bulk blending. The size of granulator product during a period when smooth spherical product was being made is compared with the TVA-size specifications for screened product in the following tabulation.

Table 4. Operating data—pan granulation of urea in TVA demonstration plant

Production rate, t/h	8.5
Pan granulator	
Speed, r/min	18
Slope, degrees	66
Number of sprays	10
Urea	
75% solution	
Temperature, °C	93 (200°F)
Feed rate (100% basis), t/h	8.5
Concentrated solution to granulator	
Temperature, °C	145 (290°F)
Concentration, %	98.5-99.0
Granulation	
Recycle	
Temperature, °C	45 (115°F)
Ratio, lb/lb product	2.1
Granulator product	
Temperature, °C	100 (210°F)
Screen analysis, %	
+6 mesh	7
−6 +10 mesh	59
−10 +16	28
−16 mesh	6
Rotary coolers	
Temperature, °C	
Precooler	55 (130°F)
Product cooler	45 (110°F)
Recycle cooler	47 (115°F)
Product	
Chemical analysis, %	
Total N	45.5
Biuret	1.1
H_2O (Karl Fischer)	0.1
Conditioner	
Oil-wax mixture	0.3
Clay	0.7
Screen analysis, %	
+6 mesh	0
−6 +8 mesh	30
−8 +10 mesh	55
−10 +12 mesh	10
−12 +16 mesh	5
−16 mesh	0

Mesh size	Granulator product	Screened product specification, %
+6	8	0
-6 +8	28	35
-6 +10	67	85
-6 +16	89	100
-16	3	0

It can be seen from these data that about 89% of the granulator product would meet TVA product-size specifications if only the plus 6 and minus 16 mesh fractions were removed by screening. Allowing for some breakage and attrition of the granulator product during handling, it is obvious that a considerable amount of onsize product is used to meet the 2:1 to 3:1 recycle requirements for control of granulation.

Screening the product as minus 6 plus 8 mesh would provide a "natural" source of recycle without returning product to the granulation system. The closely sized product (-6 +8 mesh) would be very good for direct application or as a substrate for sulfur coating, but it would not be very suitable from the standpoint of size distribution for bulk blending with conventional granular materials.

Recent Process Improvements

Process losses from the demonstration plant after improvements were made are negligible. Dust from various sources is collected in wet scrubbers of the low energy-type, and the scrubber solution is returned to the evaporator for reprocessing. A recirculating spray-type scrubber removes ammonia and particulate materials from the gaseous exhaust stream from the evaporator. The plant is in compliance with the State of Alabama's stack gas emission and effluent water standards.

The principal disadvantage of the dust collection and recovery system in the TVA demonstration plant was that the wet scrubbers had to operate with relatively dilute solution (about 50%) to prevent salting whenever a surge of dust entered a scrubber. Return of the dilute solution increased the load on the evaporator and made operation within a tight water balance difficult. A bag filter system was installed in late 1979 to collect a large part of the dust in dry form. Early operation indicates that the solution returned to process will have a concentration of about

75%, instead of 50% which was necessary before the bag filter was installed. Although rigid operating procedures and greater maintenance surveillance are required with bag filters handling hygroscopic dusts, use of such equipment has become much more prevalent in fertilizer granulation plants. In many situations a bag filter unit is about the only practical means for meeting environmental standards.

Quality and Physical Properties of the Granular Urea

Quality of the granular product is good. The granules are spherical and have a smooth, but not polished, surface texture when granulated at a temperature in the range of 105° and 107°C (220° and 225°F). The finished product usually contains from 0.10% to 0.15% moisture and from 1.0% to 1.5% biuret, which is suitable for all fertilizer uses except foliar application. The biuret content is largely a function of the amount of scrubber solution recycled to the process.

The product has good storage and handling characteristics when coated with about 0.3% of an oil-wax mixture and about 0.7% of kaolin clay. The oil-wax mixture functions primarily as a dust suppressant. However, as described earlier, the practice of incorporating 0.4% of formaldehyde provides good storage and handling characteristics without the use of surface conditioning agents. The formaldehyde solution gives best results when fed into the concentrated solution in the pipeline between the evaporator and pan (figure 5). The solution consists of 37% formaldehyde, 12% methanol, and 51% water. The methanol is added to ensure fluidity at ambient conditions. The product with either treatment is essentially dust-free during handling, storage, and shipping (6).

The granules (-6 +8 mesh) containing about 0.4% of formaldehyde have a crushing strength of 1.8 to 2.7 kg (4 to 6 lb) compared with a range of 0.9 to 1.6 kg (2.0 to 3.5 lb) for granules that do not contain formaldehyde. Urea prills of a comparable size have a crushing strength of only 0.5 to 1.0 kg (1 to 2 lb) (10).

Operation of the demonstration plant has established the feasibility of the TVA pan-granulation process for producing granular urea. Operating difficulties and some process control problems were encountered because of the use of mismatched equipment (larger size than needed) that was already installed. It is evident that most of the difficulties could be eliminated in a commercial plant designed specifically for this purpose.

Users of the TVA granular urea have been enthusiastic about its appearance and properties, and it has been particularly suitable for bulk blending. Granular urea with particle-size distribution and surface texture suitable for preparation of sulfur-coated urea (SCU) in TVA's new demonstration unit is being produced (12).

Urea Costs—Granulation Versus Prilling

Air prilling and granulation are the most common means of producing solid fertilizer-grade urea. In selecting the process to be used for new installations, the producer should consider production costs, customer preference for specific product properties, and environmental controls required for each process.

The granulation processes (both pan and spray drum) are more amenable to control of product size and quality than prilling. Control and recovery of fumes and dust also is easier and less expensive. The larger products made by granulation are much better suited for use in bulk blending, forest fertilization, aerial application, and sulfur coating. Both the prills and granular products are suitable for direct application. A decided customer preference for the granular products appears to be developing.

Control of atmospheric emissions is easier for the granulation processes than for the prilling process, largely because of the much smaller volume of air handled in the granulation system. Also, prill towers are tall and scrubbing equipment would need to be installed and operated at the top of the tower or extensive large ductwork would be required to bring the air to a ground-level scrubber. Installation costs for dust collection and recovery, therefore, are much less for granulation than for air prilling.

It is difficult to get a realistic estimate of the comparative costs of producing urea by the various processes. The relative production cost for each process would vary with the size of the installation, geographic location, utility costs, and the regulations for atmospheric emissions.

A TVA estimate published in 1975 (13) indicated production cost for pan granulation to be about $1 per ton more than for prilling in a 400-ton-per-day plant and about $2 per ton higher in a 1,000-ton-per-day plant. However, there is a question whether the exhaust air from prill towers would meet the atmospheric emission standards for many areas. Installation of adequate pollution control equipment for the prill towers as mentioned above would probably increase production costs for prills to at least that for pan-granulated urea.

Because of increasing restrictions on allowable at-

mospheric emissions and consumer demands for the larger and harder granular product, many producers are taking a close look at the pan-granulation process for use in new facilities that are to be built; the improved quality should justify a slightly higher cost for the granular urea product.

Status of Development and Demonstration of TVA's Pan-Granulation Process and Outlook for Commercial Use

TVA's thorough pilot-plant development work on pan-granulation processes for ammonium nitrate and urea, and 6 years of demonstration plant experience with each have proved their viability for commercial use. The demonstration plant experience at production rates of 18 to 22 tons per hour for ammonium nitrate-based products and about 9 tons per hour (capacity of urea solution plant) for granular urea have shown that the basic processes are sound. Equipment and process technique have demonstrated versatility and flexibility. Production capability for the TVA pan granulators was demonstrated to be considerably higher than was projected from pilot-plant experience. Product quality from the standpoints of granule size range that can be produced, shape, crushing strength, and storage properties has been excellent. A simple treatment of the concentrated urea solution with 0.4% of formaldehyde ensured 46% N grade without need for further conditioning, and greatly increased the crushing strength of the granules.

Improvements were made during the course of TVA demonstration plant operation, and problems that remain are related mostly to those inherent to auxiliary equipment from other processes that were too large or poorly suited. The pan-granulation system fully meets rigid (1979) standards of air and water quality as specified by the State of Alabama, and U.S. regulatory agencies. Operating experience has shown that a few basic changes should be made in the system when designing a commercial plant. Collecting a large part of the dust dry by use of a bag filter, as implemented in the TVA demonstration plant in late 1979, results in scrubbing liquor of about 75% concentration. Return of this more concentrated solution to the evaporator will not overload the water balance of the process.

Equipment arrangement would be more compact, and equipment would be matched in capacity in a plant specifically designed and built for this process. This would facilitate materials handling and decrease the formation of dust by attrition; lower heat losses in size-matched equipment would facilitate control of the process.

Engineers at TVA's NFDC have worked with engineering firms in planning designs for medium and large pan-granulation facilities. The N-ReN Corporation of Pryor, Oklahoma, has a pan-granulation unit for ammonium nitrate which began operation in December 1975. This unit, which uses the TVA process, replaced a prill tower that was shut down because of particulate pollution problems. The design rate of this unit is 400 tons per day. The operation at the N-ReN plant demonstrated that a dryer is not needed for the process when the feed to the pan is essentially anhydrous (99.5%) ammonium nitrate melt.

A medium-sized pan-granulation facility of the TVA type for urea is being constructed in Indonesia and is scheduled for operation in 1980. A larger facility in Pakistan, with multiple pans for production of granular urea, is being designed. Interest in pan granulation of urea has intensified. This is expressed in many requests to TVA for information by correspondence and in detailed discussions by representatives of industry and engineering firms with NFDC staff.

Other Pan-Granulation Processes

Norsk-Hydro a.s. of Rjudan, Norway, (14) has reported on a commercial pan-granulation plant for producing ammonium nitrate; some operating features are substantially different than in the TVA process. In this plant, the melt is sprayed into a deep part of the bed in the lower area of the pan granulator. The product is discharged at a temperature only a few degrees below the fusion temperature, and the granular product passes through a polishing drum before it is cooled. The granular products are reported to be very hard and strong. Ammonium nitrate prills are coated with ammonium nitrate melt in a pan granulator to produce large granules for forest fertilization. Granular ammonium nitrate of a conventional size also is produced in this facility. Granular urea is produced by a similar process in which the feed material consists of urea melt and recycled solids from a screening step.

Cominco, Limited, of Trail, British Columbia, Canada, has reported on a process for granulation of urea or ammonium nitrate by spraying the melt on solid particles in an inclined pan. The melt is sprayed onto a fast moving part of the bed in the granulator in a manner similar to that used in the TVA process.

A published reference states that cooled air is used in the product cooler and the system is operated at temperatures considerably lower than those used by TVA (15).

African Explosives and Chemical Industries Limited, Johannesburg, South Africa, (16) has reported on the use of a pan granulator for producing ammonium nitrate. One feature of their process is cooling of the granulator product to below the 32°C (90°F) crystal transition temperature before the final drying step. The granular product is reported to be suitable for use as a blasting agent when mixed with fuel oil.

Attractive Features of Pan Granulation

There are several features of pan granulation that should make this process attractive, particularly to those planning new facilities.

- Granules can be readily produced in a wide range of sizes suitable for bulk blending or for coating with sulfur for controlled release properties. Considerably larger (5 to 8 mm) granules can readily be produced for forest fertilization or other uses with aerial application. The contrast in size and appearance of prills, regular pan-granulated urea, and forestry-size granular urea is shown in figure 6.
- The granules have excellent crushing strength. Granular urea with addition of 0.4% of formaldehyde has a nitrogen content of 46%, and crushing strength is several times that of prills. No further conditioning is needed for good storage and handling properties.
- Equipment has high production capability—at least 18 to 24 tons per hour for a single pan.
- The use of 99.5% solution avoids the need for a dryer. This greatly facilitates the compactness of layout, and less energy is required. There are much lower volumes of gases to handle and treat for dust and fume recovery, and the most expensive system of a conventional granulation process (dryer and accessories) is eliminated.
- The ability to fully meet rigid standards for dust and fume emissions is greatly facilitated in comparison with prilling, where huge volumes of air must be handled and treated. Bag filter systems, that are particularly effective for dust recovery, are practical for the comparatively low gas flows for pan granulation.

There are a few disadvantages to take into account.

- Investment and production costs might be higher by about $1 per ton of product as compared with prilling. With fully adequate control of fumes and dust in a prilling operation, investment and production costs likely would be about equal.
- Control of granulation of urea is somewhat "touchy" if production of particularly well-rounded granules is desired. A system designed specifically for this process would facilitate the needed controls.

TVA Patents on Pan Granulation

The following patent and defensive publication were issued to TVA on pan-granulation processes.

U.S. 3,165,395—issued June 12, 1965

Defensive Publication T 909,016—issued April 17, 1973

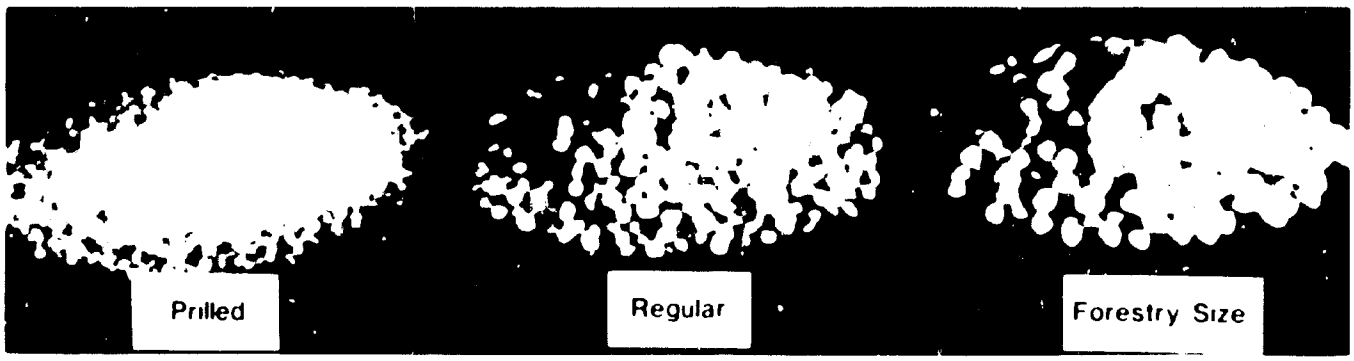

Figure 6. Comparison of particle sizes of urea products

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15. Thompson, J. B., and G. C. Hildred, U.S. Patent 3,408,169 (October 29, 1968), assigned to Cominco, Ltd.
16. British Patent 1,024,558 (March 31, 1966), granted to African Explosives and Chemical Industries, Limited, Johannesburg, South Africa.