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Continuous Process for the Thermal
Decomposition of Uranyl Nitrate
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December 7, 1950

Mr. C. L. Karl
Area Manager
U. S. Atomic Energy Commission
St. Louis Area
P. O. Box 36, Bremen Station
St. Louis 7, Missouri

Dear Mr. Karl:

The attached report by Mr. William Shelley describes the experiments which were conducted in the Plant 6 Pot Room at the request of the AEC for the Hanford Engineering Works in an effort to gain data regarding a continuous process for the denitration of UNH. The experiments conducted are described in detail and the thermal data are included in this report. The physical and chemical studies of the products produced have not yet been completed and will be included in detail in a subsequent report. As indicated in the conclusion in the attached report the results of this work are very promising with respect to the possibility of a continuous process for this decomposition. The complete detailed report of the remainder of this study will be forthcoming in the near future and we will await with interest your reaction to the desirability of further work in this field.

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Very truly yours,

MALLINCKRODT CHEMICAL WORKS

W. H. Keller

ldm
Enclosure

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December 7, 1950

Work Done By:
William Shelley
Ward Powers
Bruce Olinsted
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Ballinckrodt Chemical Works
St. Louis, Missouri

William Shelley

Written by

Preliminary Studies of a Continuous Process for
The Thermal Decomposition of Uranyl Nitrate Hexahydrate

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(Please circulate Central Files' copy to Messrs. Thayer, Powers, Olsted, Leager, and Ritchie.)

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ABSTRACT

At the request of the A.E.C., experiments were conducted on the continuous addition of liquid UNH to a bed of powdered UO_2 in a gas heated pot in the Pot Room of Plant 6 to gather thermal data regarding the continuous decomposition of UNH and to make observations of the physical behavior of such a system. The thermal data indicate that the size and the energy requirements of the equipment required per pound of UO_2 produced would be considerably smaller than for the present batch-wise process. The products of the reactions through all the successive stages were more dense than the products of the present process. The metal yields in the reduction stage were 98.5%, 97.6% and 97.8%. The yield of 98.5% in the one reactor indicates that this process will produce UF_4 capable of producing as high yields as any yet made. The lower yields in the other two cases fall within the present control limits for yield. The determination of the physical and chemical properties has not yet been completed and will be reportedly shortly in a subsequent paper.

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Preliminary Studies of a Continuous Process For
The Thermal Decomposition of Uranyl Nitrate Hexahydrate

Introduction

At the request of the A.E.C. and Hanford Engineering Works, equipment was fabricated and erected in the Pot Room of Plant 6 so as to manufacture UO_3 continuously by adding liquid hexahydrate to a pot partially charged with powdered UO_3 . The pot used was one of those installed as normal process equipment for the production of uranium trioxide. A steam heated tank was constructed from a fifty-five gallon stainless steel drum for the addition of liquid hex to the pots. The Research Department at Hanford had successfully employed this procedure for the manufacture of UO_3 on a laboratory scale and were interested in determining the rate of heat transfer to a pot of the type normally used for production and in observing the possibility of preventing lumping and caking in such a process.

Experimental

The pot was loaded with a partial charge of orange oxide powder. Heating commenced and the liquid hex added at a rate thought to be equal to the rate of conversion to UO_3 . A sufficient amount of hex was added so as to give an end product approximately equal to that amount normally produced in this pot. Four experimental runs were made with this equipment, plus one run to determine the rate of temperature increase. Of the four manufacturing runs, one produced a very non-homogeneous mixture of black powder, orange powder, and rather large cakes which appeared to be solidified hex coated with black oxide. Each of the other three runs produced approximately normal granulated material though in some cases large cakes were present. The chemical and screen analyses of these products are included below in Table No. I.

Table I

Screen Analysis

<u>Run No.</u>	<u>I</u>	<u>III</u>	<u>IV</u>	<u>Typical Unmilled Product</u>	
+20	46.80%	23.70%	6.70%	+20	16.0%
-20+35	7.80	6.02	4.53	-20+35	16.9
-35+38	7.40	6.25	9.52	-35+50	12.0
-38+60	10.10	14.18	23.00	-50+70	9.0
-60+80	4.66	9.80	12.10	-70+100	6.5
-80+100	2.34	4.75	8.25	-100+200	11.2
-100+140	3.33	6.15	10.30	-200+325	5.5
-140+200	1.79	2.98	5.35	-325	21.3
-200+325	1.05	2.16	5.09		
-325	14.73	24.01	15.16		

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Powder Density gm/cc

<u>Run No.</u>	<u>I</u>	<u>III</u>	<u>IV</u>	<u>Typical Unsifted Product</u>
Pour	3.82	4.15	3.98	3.65
Tap	4.11	4.57	4.55	4.40

Chemical

Total HNO_3	.75%	.25%	.13%
H_2O	.24	.04	.04
NA Insoluble	.0014	.0037	.0036
Fe	.0012	.0009	.0012
Mo	<.0001	<.0001	<.0001
Cr	2 ppm	3 ppm	4 ppm
Ni	2 ppm	1 ppm	1 ppm

For a period of thirty minutes during run No. 3 the rate of hex addition and the temperature of the off gases from the pot remained constant. The data collected during this period was used to calculate the required heat data. During this period the over-all heat transfer coefficient from the atmosphere of the furnace to the powder in the pot is calculated to be about 25 B.T.U. per sq. ft. per degree Fahrenheit per hour. The results during the heating cycle described previously indicate a heat transfer coefficient of about four units.

The uranium trioxide produced in Run No. 3 was converted to uranium dioxide, which assayed 97.4% UO_2 . This material was then converted to UF_4 which contained 2.9% water solubles and .07% ammonium oxalate insolubles. The green salt was reduced to metal in three reactors producing an average yield of 98%. These reactions were done in the normal production equipment.

Conclusions

From these preliminary studies it is thought that a continuous process for the manufacture of UO_3 from hexahydrate is feasible. Data now being accumulated upon the conversion of this UO_3 to metal will give more complete information.

Several different arrangements of process equipment are possible for this process. The process development department at Hanford is considering among others, the use of pots similar to those installed in Plant 6 with a bottom discharge for continuous removal of the powder. Such equipment presupposes that the entire contents of the pot consists of powder and the reaction is proceeding instantaneously during addition of the hex. In the first run attempted during this series, a sample was taken immediately after the addition of the last of the liquid and this sample contained .75% free nitrate and .25% water indicating that the material was not completely converted. Further heating of the powder produced in the Hanford equipment should complete the reaction.

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The design of Plant 6 required that the motors of the stirrers installed in the pots be of sufficient size to insure stirring at the stage of the reaction when the material was essentially wet powder. Before and after this stage, in the liquid or solid stage, much less powerful equipment would be required. To avoid the capital expenditure of heavy agitator motors and rather large power consumption, a continuous reaction should avoid this pasty stage. It is thought that this could be accomplished by mixing the liquid with a small amount of powder at a temperature of 250° in a blender or screw or dough mixer. Such a mixer would require a very efficient mixing for a short period. From this mixer the wet powder could be blended with a constantly recycling stream of UO_3 at approximately the reaction temperature and carried through a reaction chamber maintained at 600° F. It is thought that such a system would produce UO_3 at almost an instantaneous rate upon reaching the reaction zone. The gases produced during the reaction must be filtered to remove the entrained orange oxide dust so as to prevent contamination of the nitric acid produced when these gases are scrubbed. If this system were incorporated with the closed circuit grinding and classifying system a very closely sized product could be manufactured.

Other process equipment could undoubtedly accomplish this conversion. It is thought that any particular sequence would require less power and provide a more healthful atmosphere than the present method of UO_3 manufacturing.

WJShelley
ldm