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THE REMOVAL OF URANIUM AND THORIUM FROM FUELED-GRAPHITE MATERIALS BY CHLORINATION

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METALS AND CERAMICS DIVISION

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J. L. Cook and R. L. Hamner

APRIL 1964

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THE REMOVAL OF URANIUM AND THORIUM FROM FUELED-GRAPHITE MATERIALS BY CHLORINATION

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ABSTRACT

A nondestructive process was developed for the removal of exposed uranium and thorium from fueled-graphite spheres for gas-cooled pebble-bed reactors. Volatile chlorides were formed and removed by reacting exposed uranium and thorium at high temperature with a dynamic atmosphere of gaseous chlorine diluted with argon. Chlorination experiments were conducted at 800 to 1000°C with uncoated spherical UC_2 and $(U,Th)C_2$ particles, pyrolytic-carbon coated fuel particles, and graphite matrices containing coated fuel particles. The results were evaluated by radiography, chemical analyses, x-ray diffraction analyses, and alpha assay.

It was concluded that exposed uranium alone was more readily removed by chlorine treatment than exposed uranium-thorium combinations, but that both could be removed by treating for 16 hr at 1000°C in a gaseous flow of 80 cm^3/min Cl_2 and 125 cm^3/min Ar.

INTRODUCTION

The nuclear fuel elements conceived for gas-cooled, fueled-graphite systems such as the PBRE[1] and the AVR[2] employ spheroidal fuel particles approximately 200 μ in diameter coated with approximately 100 μ of pyrolytic carbon. The fuel particles may be UC_2, $(U,Th)C_2$, UO_2, or $(U,Th)O_2$; for each of these types, the particles are distributed uniformly in spherical graphite matrices.

The principal functions of the pyrolytic-carbon coatings during reactor operation are to retain fission products and prevent fuel migration. It is highly desirable, therefore, that no fuel particles with

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[1]Oak Ridge National Laboratory, Conceptual Design of the Pebble Bed Reactor Experiment, ORNL-TM-201 (May 1962).

[2]General Atomics, Graphite-Matrix Fuels Development for the AVR, GA-4695 (October 1963).

ruptured coatings be inserted in a reactor. However, it has been demonstrated that damage of a small percentage of the particle coatings can be expected during the coating process, during handling, or during fuel-sphere fabrication operations, leading to internal and external exposure of fuel in the finished fuel element. In view of these considerations, development was undertaken of a method, feasible in principle for production operations, for removing exposed fuel from coated particles and from fueled-graphite components containing coated particles.

PRIOR WORK

Chlorination processes for the removal of uranium and thorium from ores, graphite, and metal matrices have been studied extensively.[3–7] In these studies, recovery processes were developed in which finely ground uranium- or thorium-bearing materials were exposed for relatively short periods to gaseous Cl_2, CCl_4, or both at temperatures from 500 to 1000°C, and the metal elements desired were volatilized as chlorides. Most of these investigations were limited in scope, and a quantitative evaluation of the effectiveness of the reported chlorination processes for uranium and thorium recovery (or removal) could not be made. In work directed at recovery of fuel from spent fuel elements, however, at least 95% of the uranium was removed by chlorination.[8,9] This was considered sufficient basis for investigating such a technique for the purposes of the present study.

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[3] M. J. Bradley and L. M. Ferris, Recovery of Uranium and Thorium from Graphite Fuels, ORNL-2761 (March 1960).

[4] L. W. Baleziak and K. C. Newman, Recovery of Tuballoy from Carbon, CD-GS-40 (June 1945).

[5] Eldorado Mining and Refining, Ltd., Research and Development Report for October, 1959, R-59-10, NP-8069 (Oct. 1959).

[6] Eldorado Mining and Refining, Ltd., Research and Development Report for December, 1959, R-59-12, NP-8306 (Dec. 1959).

[7] Argonne National Laboratory, Reactor Development Program Progress Report, November, 1962, ANL-6658 (Dec. 1962).

[8] T. A. Gens, ORNL-3376 (May 1963)(classified).

[9] R. E. Blanco et al., Processing of Graphite Reactor Fuels Containing Coated Particles and Ceramics, ORNL-TM-667 (Sept. 1963).

CHEMICAL AND PHYSICAL CONSIDERATIONS

The fuels in the coated particles can be either UO_2, $(U,Th)O_2$, UC_2, or $(U,Th)C_2$. In reviewing the chemical reactions that might occur when these compounds are removed from ruptured particles or from contaminated graphite matrices by gaseous chlorine, only the oxides were considered since they are less reactive than the carbides. The formation and behavior of uranium and thorium chlorides are discussed below.

Uranium Chlorides

Four binary uranium-chlorine compounds are known: UCl_3, UCl_4, UCl_5, and UCl_6. Katz and co-workers[10,11] have reviewed the chemical and physical behavior of these compounds. Selected physical and thermodynamic properties are listed in Table 1.

Table 1. Properties of Uranium-Chlorine Compounds

Compound	Melting Point (°C)	Density (g/cm^3)	ΔH of Sublimation (kcal/mole)	$\Delta F°$ of Formation at 298°K (kcal/mole)
UCl_3	835	5.51[a]	55.0	−196.9
UCl_4	590	4.87[a]	47.7	−230.0
UCl_5	327[b]	3.81[c]		−237.4
UCl_6	179[d]	3.56[a]	11.12	−241.5

[a]From x-ray diffraction data.

[b]Estimated quantity.

[c]Direct measurement by immersion in benzene.

[d]With decomposition.

At sufficiently high temperatures and in the presence of chlorine, the lower uranium chlorides are converted to UCl_6. However, the higher chlorides revert to UCl_4 and chlorine at higher temperatures or lower chlorine concentrations.

[10]J. J. Katz and E. Rabinowitch, The Chemistry of Uranium, pp 450–507, McGraw Hill, New York, 1951.

[11]J. J. Katz and G. T. Seaborg, The Chemistry of the Actinide Elements, Wiley, New York, 1957.

Uranium dioxide reacts with CCl_4 or with carbon and gaseous chlorine to form UCl_4 (which in the presence of excess chlorine at sufficiently low temperatures may in turn be converted to UCl_6):

$$UO_2(s) + C(s) + 2Cl_2(g) \rightarrow UCl_4(s) + CO_2(g)$$

$$UO_2(s) + CCl_4(g) \xrightarrow{450°C} UCl_4(s) + CO_2(g) \quad .$$

In the presence of carbon, the equilibrium

$$CO_2(g) + C(s) \rightleftharpoons 2CO(g)$$

would determine the relative amounts of CO and CO_2. Some investigators believe that CCl_4 is actually the agent for forming the volatile UCl_4 in all cases. In fact, chlorine reacted with UO_2 alone yields uranyl chloride (UO_2Cl_2), which is nonvolatile.

The vapor pressure of UCl_4 at 500°C is 0.64 torr and its normal boiling point is 792°C. The hexachloride is much more volatile, its vapor pressure exceeding 1 mm well below its melting point.

Thorium Chlorides

The only thorium chloride with significant stability is $ThCl_4$.[11] Like UO_2, ThO_2 is not effectively chlorinated with gaseous chlorine alone. Dean and Chandler[12] showed that very little $ThCl_4$ is formed when ThO_2 and gaseous Cl_2 are heated at temperatures up to 1050°C; these investigators found that the extent and rate of conversion of ThO_2 to $ThCl_4$ with gaseous chlorine depend on the efficiency of contact between carbon, thorium oxide, and chlorine and on the reactivity of the ThO_2 and carbon.

The chlorination of ThO_2 is thought to take place in two steps:

$$ThO_2(s) + C(s) + Cl_2(g) \rightarrow ThOCl_2(s) + CO(g)$$

$$ThOCl_2(s) + C(s) + Cl_2(g) \rightarrow ThCl_4(s) + CO(g).$$

The reaction is rapid above 650°C.

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[12] O. C. Dean and J. M. Chandler, Nucl. Sci. Eng. 2, 57–72 (1957).

Thorium tetrachloride melts at 770°C and boils under atmospheric pressure at 921°C. It can sublime in vacuum at 750°C. Thus, effective simultaneous removal of both uranium and thorium from carbon matrices requires temperatures of 800°C and above.

DESCRIPTION OF PROCESS

An apparatus for the chlorination of coated particles and fueled-graphite specimens containing coated fuel particles at temperatures up to 1000°C is shown schematically in Fig. 1. The specimen to be chlorinated is placed on a porous carbon (Carbocell) boat and inserted in a clear fused-silica combustion tube, fitted with water-cooled fused-silica end plugs. The downstream end plug has provision for installing a "cold finger" for the condensation of volatile uranium and thorium chlorides ahead of the NaOH scrubber. All piping carrying chlorine is made of nickel, Monel, glass, or Teflon.

The system is purged with argon, which passes through two drying columns, one each of calcium sulfate and activated alumina, before entering the furnace chamber. With power on the furnace, the argon purge continues at approximately 125 cm^3/min. The chlorine gas, which is dried separately by two drying columns, one each of calcium sulfate and activated alumina, flows with the argon into the furnace at approximately 80 cm^3/min after the furnace reaches a temperature of approximately 400°C. On the outgas end the chlorine and argon are bubbled through a saturated NaOH solution, which absorbs the chlorine, and the argon is exhausted to the atmosphere. Because of the hazards of chlorine gas, the entire system is contained in a fume hood.

The specimens being treated are heated in the flowing argon-chlorine mixture at a prescribed temperature for a given period of time. Upon completion of the chlorination, the power is turned off and the specimens are allowed to cool at least to 200°C in flowing argon before removal. After the chlorine flow is stopped, bypass valves are opened, allowing the argon to purge the chlorine from the entire system during the cooldown. This purging of the chlorine from the piping decreases corrosion to lines, flowmeters, and valves.

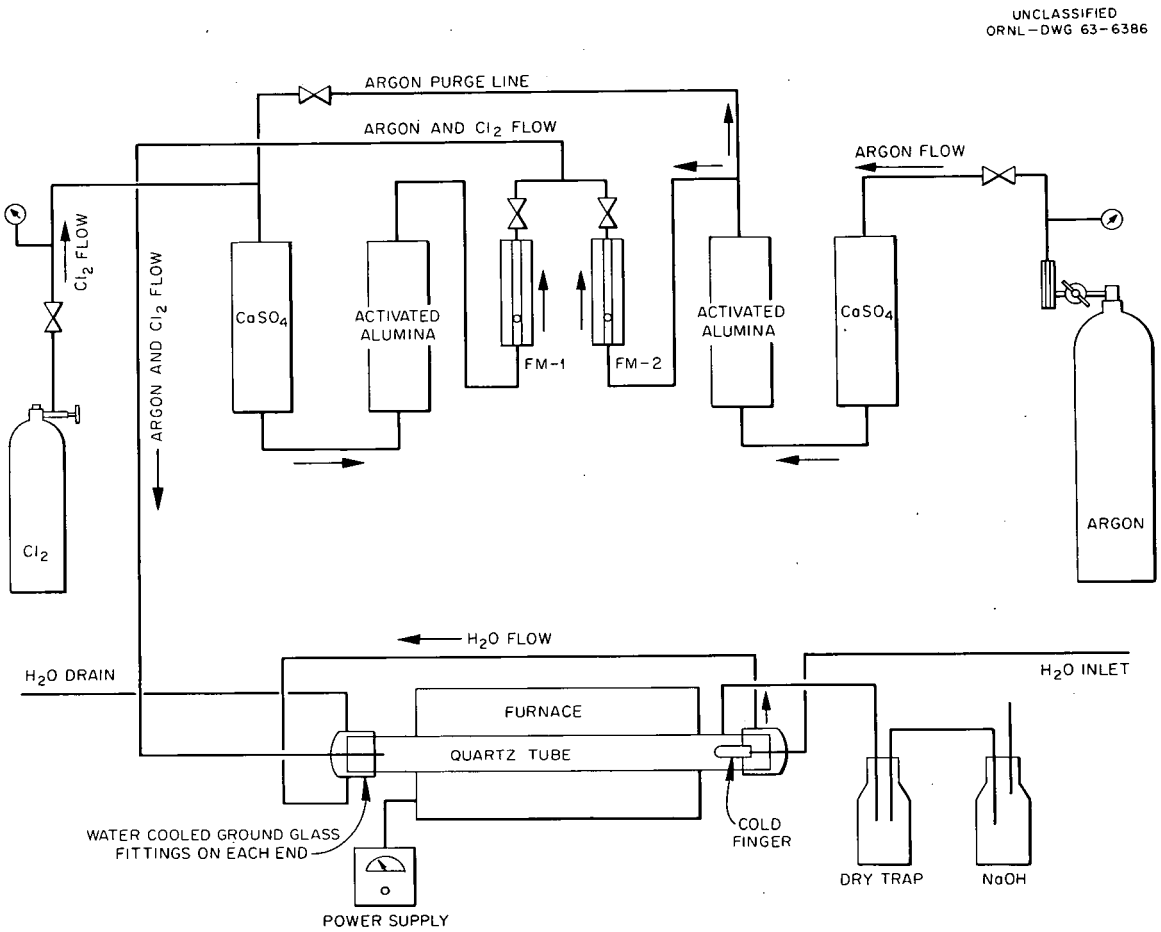

Fig. 1. Schematic Drawing of Chlorination Apparatus.

Carbon tetrachloride was employed as an auxiliary chlorinating agent in preliminary experiments because of its reported efficient chlorinating action.[13] However, since the CCl_4 dissociated at high temperatures (800–1000°C), leaving an undesirable tarlike carbon deposit on the specimens and in the system, its use was discontinued.

EXPERIMENTAL RESULTS

Chlorination was studied using three types of specimens: (1) uncoated UC_2 and $(U,Th)C_2$ particles, (2) UO_2, UC_2, and $(U,Th)C_2$ particles coated with pyrolytic carbon, and (3) fueled-graphite specimens containing coated fuel particles. Since the objectives of these experiments were somewhat different, though related, the results will be presented separately.

Chlorination of Uncoated Fuel Particles

Two lots of spherical uncoated fuel particles were used in the first chlorination studies to determine the effectiveness of the chlorine in the simplest case. A lot of "monocrystalline" UC_2 containing 91 wt % U and a lot of $(U,Th)C_2$ containing 90 wt % heavy metal with a thorium-to-uranium ratio of 0.6 were chlorinated for 5 1/2 hr at 1000°C in a 1:1 mixture of argon and chlorine containing no CCl_4. The results were evaluated by radiography, metallography, x-ray diffraction, chemical analysis, and leaching of the chlorine-treated particles in 8 M HNO_3 for 8 hr at 95°C.

$(U,Th)C_2$ Particles

The residue of the $(Th,U)C_2$ particles retained the shape of the original spherical particles after both chlorination and acid leaching, as shown by microradiographs (Figs. 2 and 3). Comparison of Figs. 2 and 3 reveals that the high-density material associated with the fibrous-appearing structure in the as-chlorinated sample was almost completely removed by the subsequent acid leach. By comparing Fig. 3 with a

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[13] T. A. Gens, Chemical Technology Division, ORNL, Private Communication to J. L. Cook, Dec. 1962.

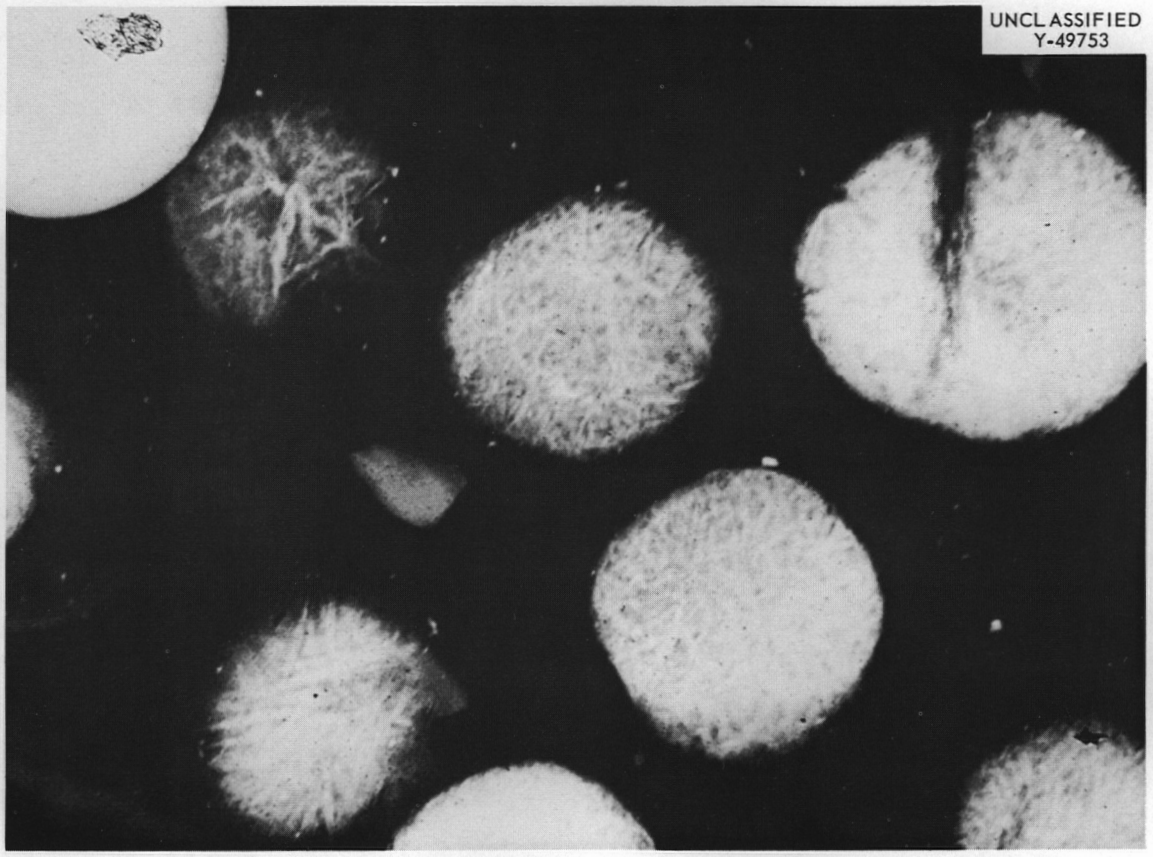

Fig. 2. Microradiograph of $(U,Th)C_2$ Particles after Chlorination. 200×.

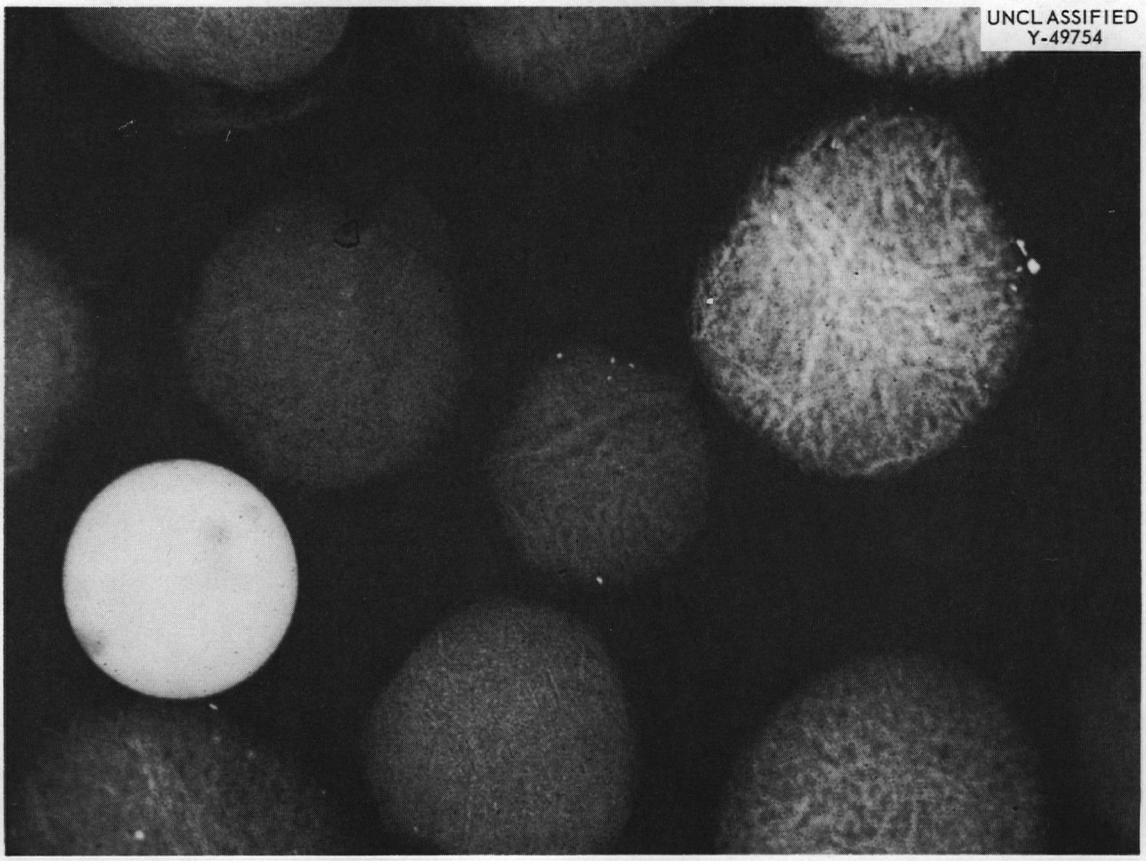

Fig. 3. Microradiograph of Residue from the Chlorination and Acid Leaching of $(U,Th)C_2$ Particles. 200×.

radiograph (Fig. 4) of similar size coke particles that contained less than 100 ppm high-density material, it was estimated that 85 to 95% of the original fuel had been removed. X-ray analysis of the residue revealed faint and very faint $ThCl_4$ diffraction lines after chlorination and acid leaching, respectively. Chemical analysis of the leach solution showed the leachable uranium and thorium remaining after chlorination to be 0.48 and 0.42 wt %, respectively, of the total quantity of fuel. If one assumes that all the fuel remaining after chlorination was leachable, the treatment was 99.5% effective. Since, however, this assumption is not quite valid for $(U,Th)C_2$ materials, the residues after chlorination and after leaching were sampled and analyzed for the quantity of fuel present. Approximately 2 wt % of fuel remained after chlorination; this was reduced by 50% by the nitric acid leach. It is interesting to note that the fuel remaining after chlorination was composed of 46% U and 54% Th, while 87% of the fuel remaining after the acid leach was thorium. The important result is that approximately 98% of the original fuel was removed by the chlorine treatment. Typical metallographic sections, Figs. 5 and 6, show only minute quantities of fuel at magnification to 1000×.

UC_2 Particles

The radiograph of the chlorinated UC_2 particles (Fig. 7) showed only a very small portion of the particles to contain high-density material, and the appearance of the residue was that of empty shells. Analysis of the residue after chlorination revealed that only 0.5% of the original uranium present remained, showing that the treatment was about 99.5% effective in removing exposed uranium. X-ray and metallographic examination detected only carbon in the residue.

Chlorination of Coated Fuel Particles

The chlorine treatment of coated particles was investigated initially as an aid to studies of fuel migration into the coating after high-temperature heat treatment. Radiographic techniques employed to follow fuel migration in coated UC_2 particles could not distinguish between surface contamination and fuel that had migrated into the coating during

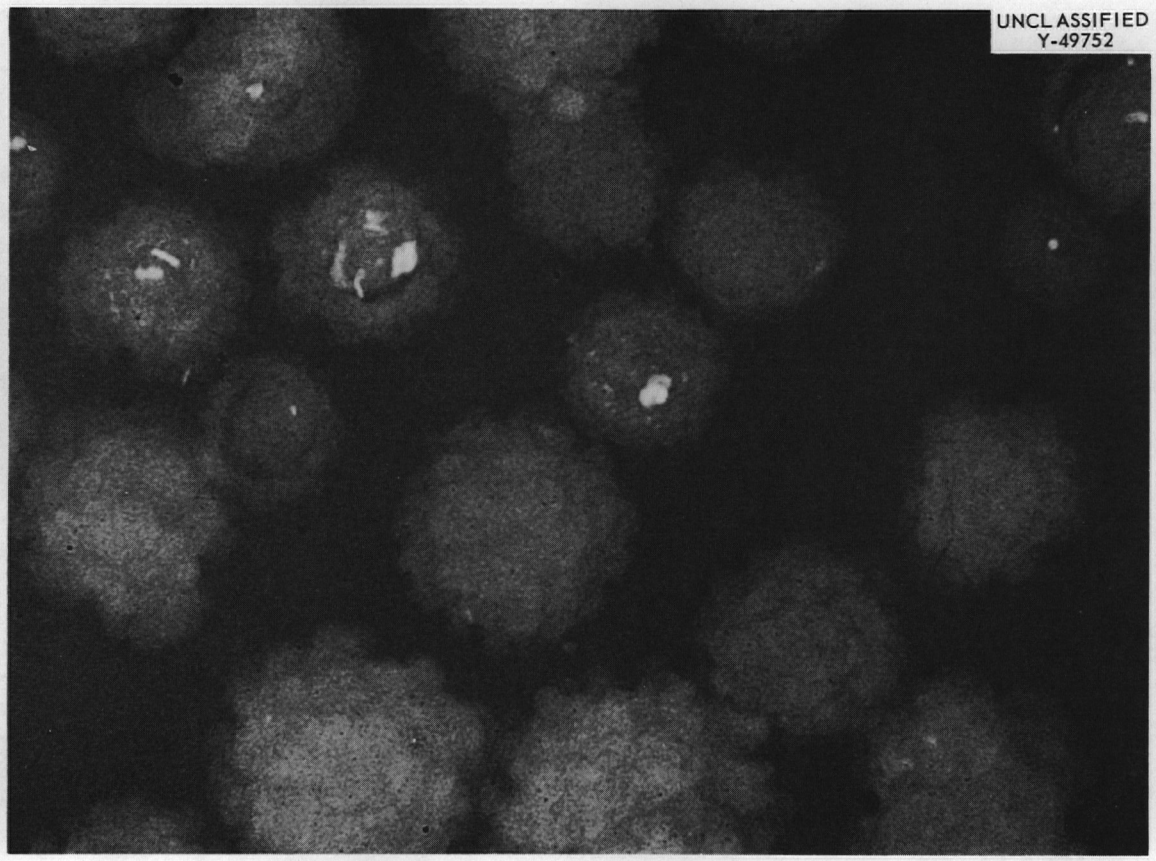

Fig. 4. Microradiograph of Coke Particles. 200X.

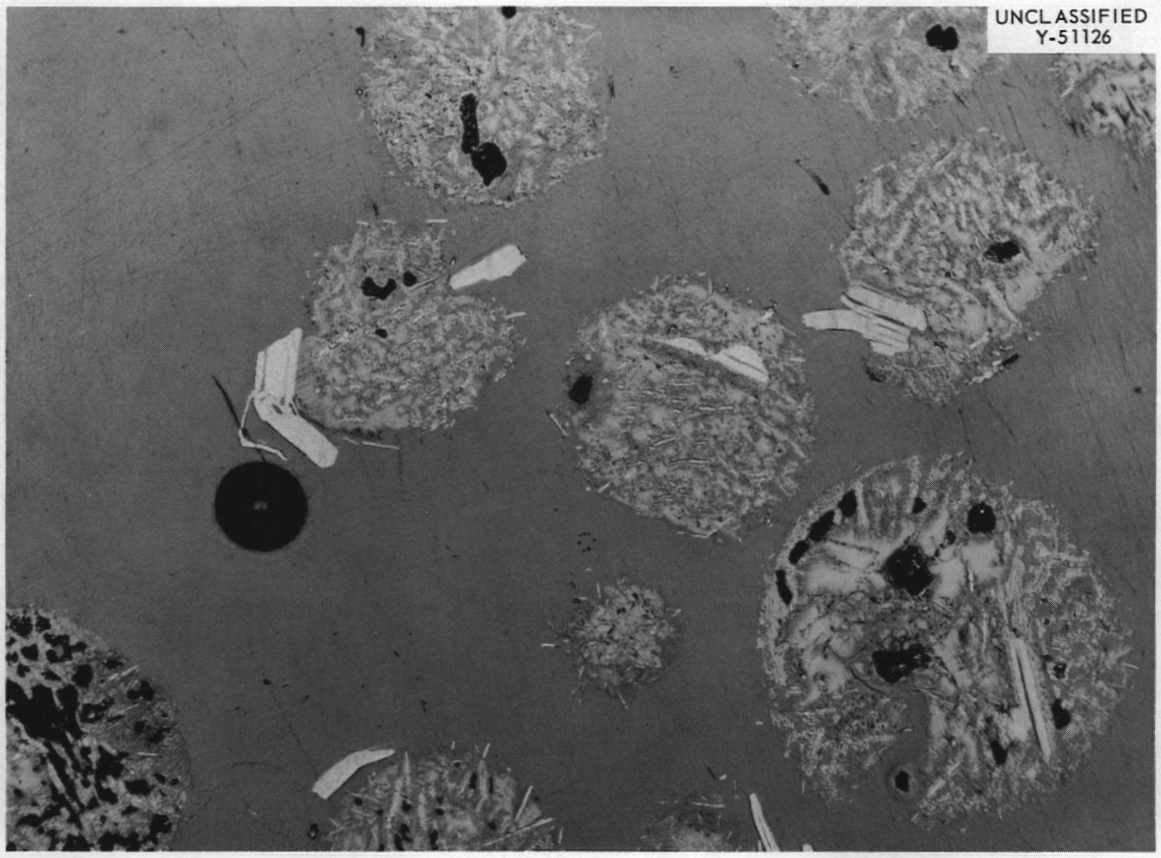

Fig. 5. Photomicrograph of Residue from $(U,Th)C_2$ Particles after Chlorine Treatment. 200×.

Fig. 6. Photomicrograph of Residue from $(U,Th)C_2$ Particles after Chlorine Treatment. 1000×.

Fig. 7. Radiograph of UC_2 Particles after Chlorination. 75×.

heat treatment at 1700°C for 307 hr (Fig. 8). The question was resolved by heating the particles to 800°C for 3 hr in chlorine to remove the surface contamination and then radiographing again (Fig. 9). Comparison of Figs. 8 and 9 clearly indicates that a large quantity of the fuel had been located on the surface of the particles.

Also, Fig. 9 shows a particle with a cracked coating. Almost all of the fuel was removed through the crack, leaving a hollow shell; this suggested that the removal of fuel from particles with cracked coatings might prove useful for eliminating damaged particles from as-prepared unsupported coated particles. If the fuel could be removed from such particles, the pyrolytic-carbon shells might be separated from the sound coated particles by a flotation technique.

Two lots of coated UO_2 particles (Nos. 104 and 105) were chlorinated for 4 hr at 800°C. It had been determined by leaching of unchlorinated samples that Lot 104 had 15.5 wt % and Lot 105 had 0.6 wt % of fuel exposed due to cracked coatings. That microradiography supports these results may be seen in Figs. 10 and 11. Alpha assays made on monolayers of the particles before and after chlorination showed that the alpha activity for Lots 104 and 105 had been reduced to 53 and 38%, respectively, of the original activity. Radiographs of particles from these lots after chlorination were examined and showed that some fuel in cracked particles still remained. This work was not pursued, but the results indicated that chlorine treatment under altered conditions could remove all of the exposed fuel.

Chlorination of Fueled-Graphite Specimens

Surface fuel exposure in fueled-graphite spheres without unfueled shells, as determined by alpha assay, was found in some cases to be approximately 0.04% (observed α activity/calculated activity of the total amount of uranium in the spheres), even after leaching in 16 M HNO_3 and rinsing with water. Internal exposed fuel contents, as determined by anodic leaching, ranged up to 16%. The highest contents were found in spheres with unfueled shells, in which the concentration of coated fuel particles (in the fueled portion) was approximately 29 vol % compared to 8.5 vol % in spheres without unfueled shells.

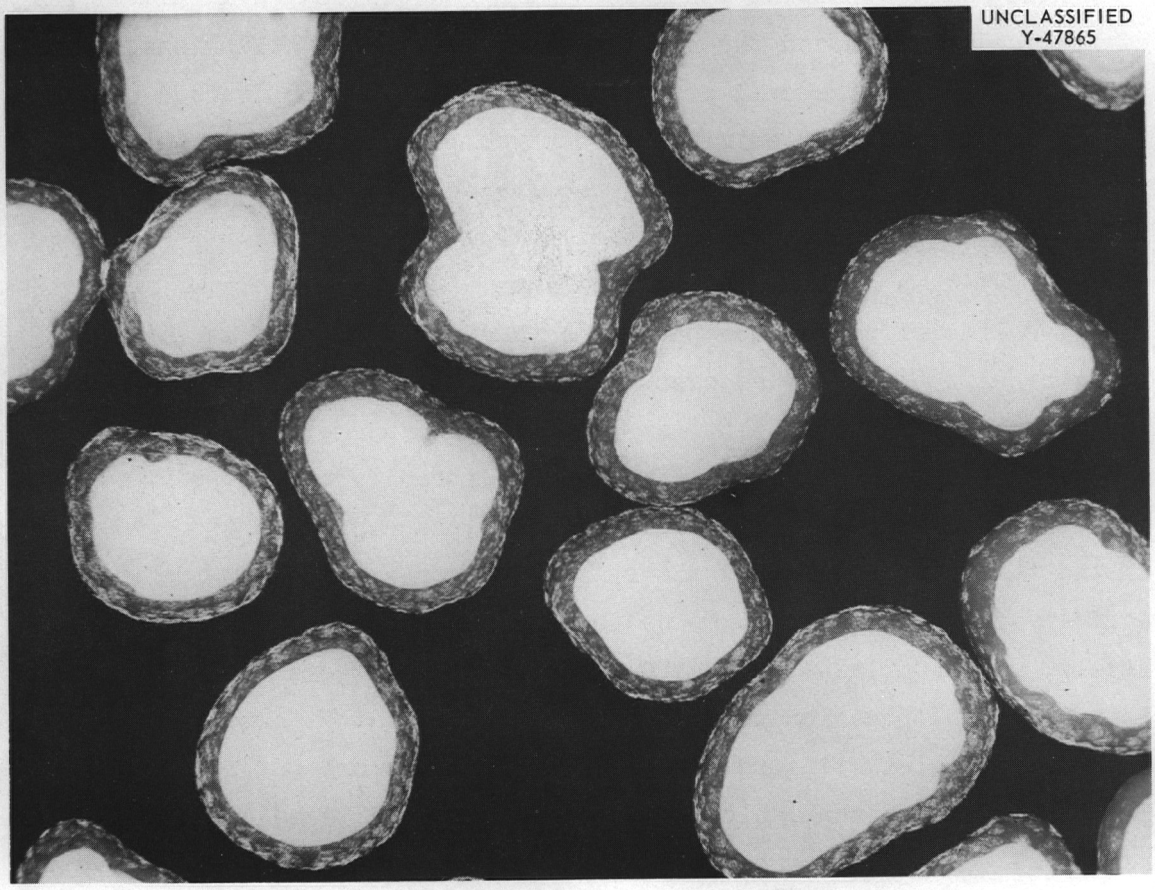

Fig. 8. Radiograph of Lot NCC 203-17 after Heat Treatment at 1700°C for 307 hr with no Chlorine Treatment. 75X.

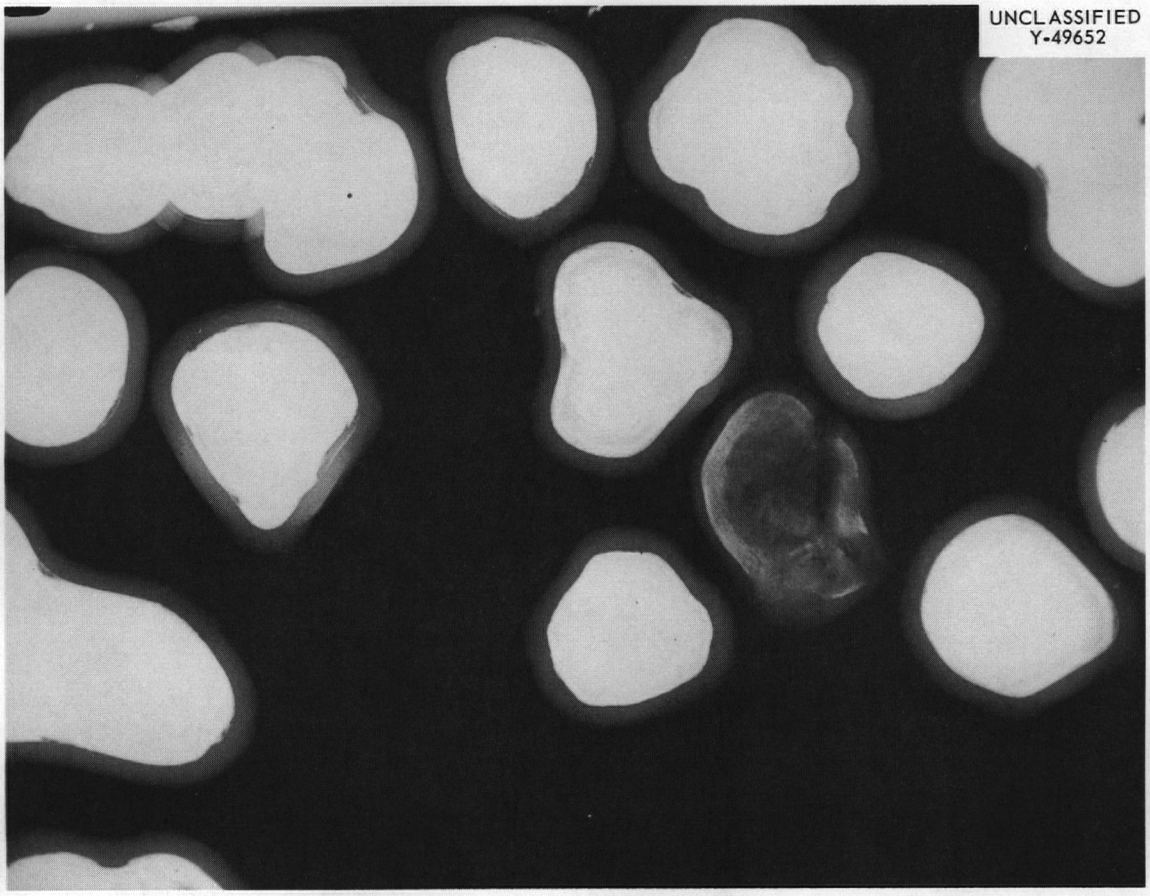

Fig. 9. Radiograph of Lot NCC 203-17 after Heat Treatment at 1700°C for 307 hr Followed by Chlorine Treatment. 75X.

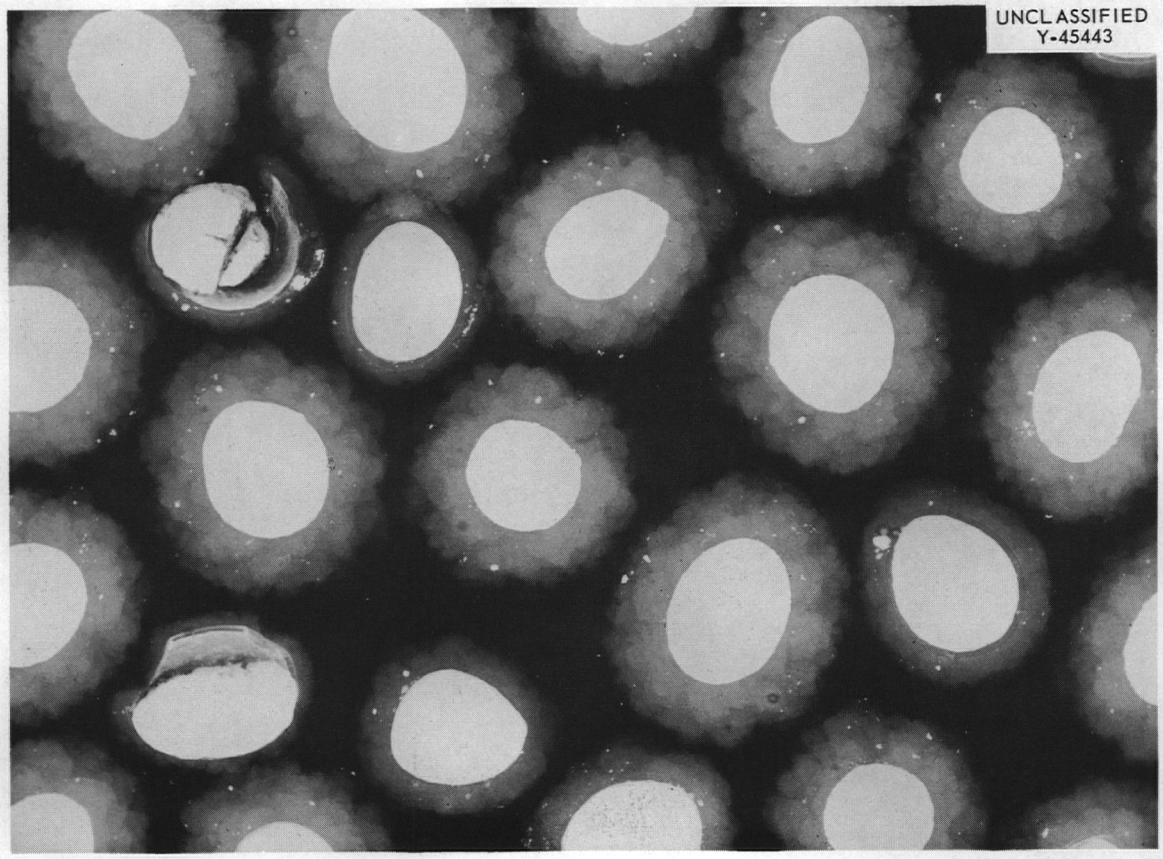

Fig. 10. Radiograph of Lot 104 before Chlorine Treatment. 75X.

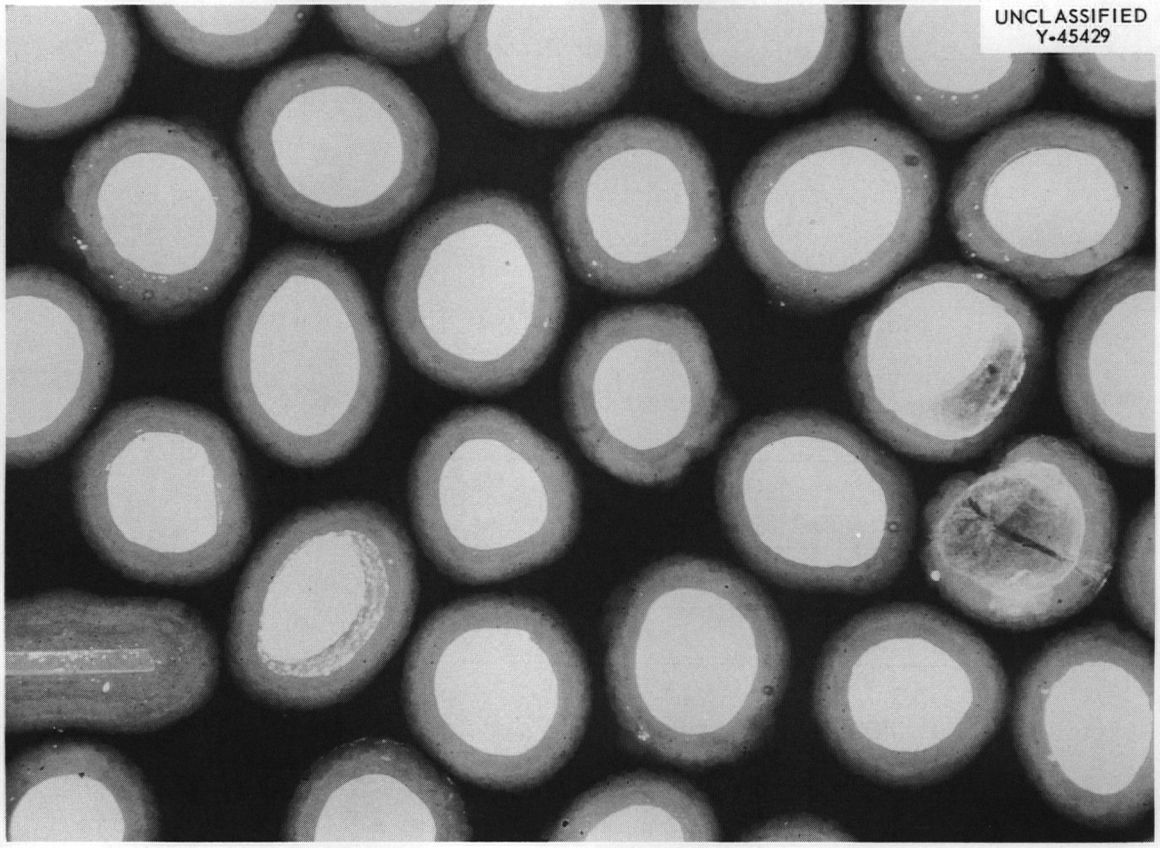

Fig. 11. Radiograph of Lot 105 before Chlorine Treatment. 75×.

A gaseous chlorination process appeared more attractive than leaching with liquid acid for the removal of the exposed fuel, because the gaseous products may be transported continuously away from the graphite matrices. Accordingly, establishing conditions for removal of uranium and thorium in graphite matrices by chlorination was undertaken, with the results evaluated by alpha assay.

In preliminary experiments, specimens 1/4 in. thick and 1 in. in diameter were cut from 1 1/2 in. graphite spheres containing pyrolytic-carbon coated $(U,Th)C_2$ particles with a thorium-to-uranium ratio of 0.6. Both sides of the specimen were counted before and after chlorine treatments at 800°C as a function of chlorination time. The results are shown in Table 2.

Table 2. Results of Chlorine Treatments of Fueled-Graphite Samples at 800°C

Surface Contamination (α counts/min/side)

Specimen No.	Before Chlorination	After 3-hr Chlorination	After 8-hr Chlorination
GA-S1-10	> 1000	300	75
3M-S1-7	> 1000	900	110

Experiments at 800°C were not carried further since it seemed obvious that the exposed fuel could, in time, be removed by chlorine treatment and that such a process would probably be more efficient at higher temperatures.

Further experiments were then conducted at 1000°C (limitation of equipment) under the same conditions of gas flow (80 cm^3 Cl_2/min, 125 cm^3 Ar/min). In the interest of developing a cleanup process with overall efficiency suitable for production, these experiments included a mechanical removal of exposed fuel from the surface before chlorination. The specimens used in these experiments were of two types. Molded fueled spheres, containing coated $(U,Th)C_2$ particles and having no unfueled shells, were obtained from General Atomic (GA) and Speer Carbon Company (SCC). Right solid cylinders, containing coated UC_2 particles, were fabricated in the Ceramics Laboratory. All specimens

contained surface-exposed pyrolytic-carbon coated fuel particles. The GA spheres (GA S3-1, -2, -7, and -15) were treated in the as-received condition (i.e., the specimen had been machined at the equator and leached in fuming HNO_3 for 2 min at the vendor's plant). The cylindrical specimens were damaged at both ends before treatment by rubbing on abrasive paper. The SCC sphere (SCC S1-5) was handsawed in half to expose a maximum flat area for treating and alpha counting. Damage to surfaces was characterized by visual observation and counting of coated fuel particles using a binocular microscope with a gridded eyepiece. The characterization of typical specimens is shown in Table 3.

Table 3. Characterization of Fueled-Graphite Specimens for Cleanup Studies

	Fueled-Graphite Cylinders (Damaged Surfaces)[a]	SCC Hemisphere (Cut Surface)	GA Spheres
Nominal diameter (in.)	0.9	1.5	1.5
Matrix density (g/cm^3)	1.45–1.50	1.78	1.91
Type of fuel particle	UC_2 (normal U)	$(U,Th)C_2$ (fully enriched)	$(U,Th)C_2$ (normal U)
Uranium content (g/specimen)	2.0	1.96	1.96
Thorium content (g/specimen)		1.2	1.2
Nominal coated particle diameter (μ)	375	375	
Nominal fuel core diameter (μ)	175	175	
Damaged area exposed for treatment and counted ($in.^2$)	1.37	1.62	
Number of fuel particles at damaged surface[a,b]	1417	1392	
Percent of damaged area occupied by fuel particles[b]	18.0	15.0	
Particles pulled out by damage (%)	13.9	41.59	
Particles with coating intact (%)	27.5	19.76	
Particles with partially damaged coating (%)	0.0	18.39	
Particles with fuel particle removed (%)	19.48	7.04	
Particles with fuel particle exposed (%)	39.1	13.2	

[a]See text.

[b]Includes all particles originally present in the plane of damage.

Mechanical Cleaning

The alpha activity level of the GA spheres and the cylindrical specimens, in general, was decreased below 50% of the original by ultrasonic cleaning in CCl_4 for two 15-min periods. No count was made on the SCC hemispheres before ultrasonic cleaning because of the highly contaminated, dusty cut surfaces that contained fully enriched uranium.

Chemical Cleaning GA Spheres

After chlorination at 1000°C for periods up to 22 hr, the activity level had not changed appreciably from the mechanically cleaned state. Although 0.3 to 0.4% of the fuel (as determined by anodic leaching) was available to the chlorine, the low magnitude of the initial surface contamination (23 to 72 counts/min) made measurement of decontamination by counting difficult and unreliable.

When unchlorinated GA spheres were leached with 4 M HNO_3 at 105°C for 22 hr, the surface activity increased from 30% of the activity before ultrasonic cleaning to 70% and remained at this level after ultrasonically "rinsing" in hot water.

Fueled-Graphite Cylinders

The initial activity level of the damaged and ultrasonically cleaned surfaces was approximately three times greater than that of the GA spheres. Chlorination treatment for 2 hr after mechanical cleaning reduced the alpha activity to background. The activity level remained at background through additional treatment up to 22 hr, even though anodic leaching of a specimen after 2 hr chlorination showed approximately 0.01% of the original uranium was still available for transport to the surface.

SCC Hemispheres

The counting data obtained in conjunction with cleaning by chlorination of the damaged "cut" surface of a hemispherical SCC specimen are probably the most reliable from these experiments, because of the very high initial alpha activity level (4.25×10^3 counts/min). The results are:

Chlorination time (hr)	4	8	12	16
Activity level (percent of initial)	4.5	1.53	0.35	background

DISCUSSION AND CONCLUSIONS

The chlorination of uncoated UC_2 and $(U,Th)C_2$ particles produced some very interesting and unexpected results. The carbon residue retained the shape of the original particle and was sufficiently strong to be handled and to withstand nitric acid leaching. The reaction apparently was not rapid or violent, since the carbon residue retained its integrity and a period of 5 1/2 hr at 1000°C was required for almost complete removal of the uranium and thorium. The rate of fuel removal of course was influenced by the high density and the low surface area per volume ratio of the particles.

Under the same conditions of temperature and gas flow, the removal of uranium by chlorination is more efficient than the removal of uranium combined with thorium. As determined by chemical analysis, chlorination of uncoated UC_2 particles removed 99.5% of the uranium, while chlorination of uncoated $(U,Th)C_2$ particles with a thorium-to-uranium ratio of 0.6 removed 99.1% of the uranium and 98% of the combined heavy metals. While chlorination removed surface-exposed fuel, as determined by alpha assay, from graphite matrices containing damaged coated UC_2 particles in only 2 hr at 1000°C, 16 hr was required for matrices containing damaged $(U,Th)C_2$ particles. It was expected that uranium would be removed more rapidly than thorium because UCl_4 is more volatile than $ThCl_4$. There is the additional possibility that uranium is transported as the very volatile UCl_6, although this higher chloride is probably not stable at this temperature. In the case in which the uranium was not removed from damaged coated UO_2 particles in 4 hr at 800°C, the slow removal was undoubtedly influenced by the low temperature (less than 1000°C) and by the low reactivity of the very dense UO_2 particles.

Based on the limited work described here, we conclude that a practical production process can be established for the removal of surface-exposed fuels at 1000°C. Although conditions have not been optimized, it is anticipated that higher temperatures would speed up the process. Temperatures may be limited, however, because of the reduction in the strength of the graphite matrices when halogen treated at extremely high temperatures. The study of the effects of chlorine

concentration, gas flow rates, and vacuum techniques would probably reveal further improvements. The use of CCl_4 as an auxiliary chlorinating agent would probably increase the rate of removal of exposed fuel, but it is not recommended for high-temperature treatments because of the tarlike deposits that are left in the system. However, incorporation of ultrasonic cleaning in a production process is recommended as a step to remove as much loose contamination as possible before chlorine treatment.

Chlorination has been valuable for cleaning coated fuel particles to be evaluated by radiography. Incorporation of chlorination into the pyrolytic-carbon coating process to produce surface-clean particles has been suggested to eliminate the need for additional equipment and perhaps making the operation more efficient. The incorporation of such a cleaning step into the coating procedure could serve to clean up the surface from exposed fuel and to provide sufficient interruption in the coating process to create a physical discontinuity between two given layers of the coating. Such a physical discontinuity has been suggested as being necessary to the successful performance of pyrolytic-carbon fuel particles.[14]

Fuel recovery was not investigated, but condensation of the uranium and thorium chlorides could readily be incorporated into the process for a highly efficient recovery operation.

Further study of chlorination is not being pursued at present. The process, however, is being used for surface cleanup of fueled-graphite spheres to be irradiation tested and for general cleaning of coated particles as a part of the evaluation. Conditions have been standardized as 16-hr treatment at 1000°C in a gaseous flow of 80 cm^3/min Cl_2 and 125 cm^3/min Ar.

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[14]R. A. U. Huddle, Deputy Director in Charge of Materials, Dragon Project, Private Communication to W. O. Harms, Metals and Ceramics Division, Aug. 20–21, 1963.

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