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REASSESSMENT OF SAFEGUARDS PARAMETERS*

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ABSTRACT

The International Atomic Energy Agency is reassessing the timeliness and goal quantity parameters that are used in defining safeguards approaches. This study reviews technology developments since the parameters were established in the 1970s and concludes that there is no reason to relax goal quantity or conversion time for reactor-grade plutonium relative to weapons-grade plutonium. For low-enriched uranium, especially in countries with advanced enrichment capability, there may be an incentive to shorten the detection time.

INTRODUCTION

To strengthen safeguards, the International Atomic Energy Agency (IAEA) is reassessing several of the basic safeguards parameters that are used in defining and structuring approaches and inspection criteria. The objective of this study is to provide information on the technical basis for the safeguards parameters, updating information available at the time of their initial consideration. The implications of modifying the parameters will be analyzed in terms of the impact that such changes might have on the effectiveness of safeguards. This study will take into account new developments in safeguards concepts and approaches and developments in the nuclear fuel cycle and nuclear technology.

The Statute of the IAEA,[1] Article XII, discussed the application of safeguards to special fissionable material. The latter is defined in Article XX as

> ...plutonium-239; uranium-233; uranium enriched in the isotopes 235 or 233; any material containing one or more of the foregoing; and such other fissionable material as the Board of Governors shall from time to time determine... .

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Safeguards are applied under Information Circular 66 (INFCIRC 66)[2] for nations not signatory to the NPT and under INFCIRC 153 (corrected)[3] for NPT signatories. Paragraph 28 of INFCIRC 153 (corrected) states

> The Agreement should provide that the objective of safeguards is the timely detection of diversion of significant quantities of nuclear material from peaceful nuclear activities to the manufacture of nuclear weapons or of other nuclear explosive devices or for purposes unknown, and deterrence of such diversion by risk of early detection."

Neither INFCIRC 66 nor INFCIRC 153 defines "timely detection" or "significant quantity."

In 1968 a group of consultant experts reported to the UN General Assembly on these matters.[4] The 22nd session of the General Assembly adopted the suggestions of the report. Following the NPT in 1968, the IAEA convened a series of expert groups under the NPT. By the late 1970s, the Standing Advisory Group on Safeguards Implementation (SAGSI)[5] had developed advice on significant quantities and conversion times, which the IAEA subsequently accepted as technical parameters to be considered in the planning, implementation, and evaluation of safeguards (Refs. 6 and 7). The values for significant quantity were derived from the 1968 report to the UN General Assembly and are 25 kg of contained ^{235}U for high-enriched uranium and 8 kg for plutonium. The values for conversion time were developed based on technical data from the Secretariat and are of the order of days (7–10) for plutonium and high-enriched uranium metal; of the order of weeks (1–2) for pure compounds; and of the order of months (1–3) for irradiated fuel. The conversion time for low-enriched uranium is one year. In the original United Nations report, significant quantity had been considered in terms of the amount of nuclear material estimated to be required for a nuclear weapon yielding a 20 kiloton explosion. Conversion time is defined as the minimum time required to convert forms of nuclear material to the metallic components of a nuclear device. The IAEA considerations in

implementing goal quantities and conversion times into inspections at the national and facility level were discussed in Refs. 6 and 7.

More than two decades has elapsed since the IAEA adopted the significant quantities and conversion times. Now, in light of experience with safeguards implementation and technical developments in nuclear technology, questions have been raised about the continued adequacy or acceptability of these technical parameters. This present study addresses whether conversion time and significant quantities should be changed in light of new processing technologies to allow for different isotopic compositions for weapons-grade and reactor-grade plutonium.

II. SIGNIFICANT QUANTITY

The materials that are identified as nuclear weapons material are ^{233}U, ^{235}U, and plutonium, and are subject to safeguards. Some pertinent data on these materials are summarized in Table I.

TABLE I. Critical Data on Selected Nuclear Materials

Element	Density	Critical (kg)	Q[a] (n/kg/s)	H[b] (W/kg)
^{233}U	18.64	16.1	0.8	0.3
^{235}U	18.80	47.9	0.3	nil
^{238}Pu	19.41	10.2	2.6×10^6	556.7
^{239}Pu	19.50	10.4	22	1.9
^{240}Pu	19.58	36.9	1.0×10^6	6.9
^{241}Pu	19.66	12.8	49	13
^{242}Pu	19.74	79.6	1.7×10^6	0.1
Pu	19.54	14.3	4.0×10^5	13

[a]Q = spontaneous fission neutron yield.
[b]H = heat generation in watts per kilogram.

To manufacture a nuclear device, the designer must have a proper amount of nuclear material (for example, 8 kg of plutonium); a high explosive that can rapidly compress the nuclear material; and a neutron source that releases its neutrons at the required time after initiation of the high explosive. Hypothetically, a mass of 4 kg of plutonium or ^{233}U is sufficient for one nuclear explosive device. If the plutonium goes critical before the neutron source releases its neutrons, for example, from spontaneous fission of the even numbered plutonium isotopes, preignition occurs and the probability of attaining the maximum yield from the mass of plutonium decreases. Nevertheless, some yield, the "fizzle" yield, will be achieved. This is in the 2-kt range for both weapons and reactor-grade plutonium. The probability of attaining the "fizzle" yield is 100%. For weapons-grade plutonium, the probability of attaining the maximum yield is reasonably high; for reactor-grade plutonium, the probability is still substantial.

Two major factors must be considered in designing/constructing nuclear weapons. One is the high, spontaneous fission neutron yield from the even numbered isotopes of plutonium that make them less desirable for nuclear weapons, but still usable. The second is the high heat generation from the short half-life alpha emitter ^{238}Pu that makes it undesirable for nuclear weapons. The high heat generation can cause problems through coupling of the metal to other components such as the high explosive or electronic parts or can cause phase transitions in the plutonium. In general, heat generation greater than 100 watts from a weapon is considered too high for manufacture of nuclear weapons. The amount of ^{238}Pu that can be tolerated in a device can be calculated from Table I. For a weapon containing 8 kg of plutonium, the maximum ^{238}Pu that can be tolerated would be approximately 2% of the total plutonium. For this reason, ^{238}Pu in amounts greater than 80% is exempt from international safeguards.

The heat generated by the plutonium can seriously affect the metallurgical properties of plutonium metal. Plutonium has six allotropic forms, each in a different crystallographic space group and with different densities. Some plutonium metal properties are summarized in Table II. The alpha phase is stable at room temperature and undergoes a 12% volume increase in transformation to the beta phase. The coefficient of thermal expansion along each of the three crystallographic axes is different so that heating produces severe distortion and cracking of the metal. The delta phase is cubic, and thermal expansion is uniform along the three axes. The delta phase can be stabilized at room temperature by alloying, for example, with aluminum or gallium. Although it has an approximately 20% lower density than the alpha phase, the stabilized delta phase is preferred for weapons regardless of the plutonium sources because of its superior metallurgical properties.

The critical masses shown in Table I are for a bare sphere; the critical mass can be significantly reduced by suitable reflectors. The reflected critical mass for pure ^{235}U is approximately 15 kg, rising to 50 kg for 50% enriched and 250 kg for 20% enriched uranium (Ref. 8, p. 17). For ^{235}U of 94% enrichment, the critical mass

TABLE II. Plutonium Allotropes

Range of Allotrope	Stability (°C)	Space Group	Density (G/cm^3)
Alpha	<122	Single monoclinic	19.84
Beta	122–206	Body centered monoclinic	17.70
Gamma	206–319	Face centered orthorhombic	17.14
Delta	319–451	Face centered	15.92
Delta prime	451–476	Body centered tetragonal	16.07
Epsilon	476–640	Body centered cubic	16.51

is reduced from 52 to 18 kg when a 17.8-cm-thick uranium reflector surrounds the spherical core (Ref. 8, pp. 19-20).

III. CONVERSION TIME

A. Enriched Uranium

As noted previously, the conversion time for compounds of high-enriched uranium (HEU) is "of the order of weeks;" the conversion time for low-enriched uranium (LEU) (<20% ^{235}U) is "of the order of 1 year." The conversion time for HEU is based on the fact that only conversion from oxide or fluoride to metal is required, and this is a relatively simple process. Conversion of LEU to weapons-grade material requires further enrichment prior to conversion to metal and requires a significantly longer time. The conversion time for LEU was established based on existing enrichment technologies in the early 1970s, which were electromagnetic separation and gaseous diffusion with the gaseous diffusion pathway receiving the most emphasis because it was the predominant technology of the time.

1. Electromagnetic separation. The first weapons-grade uranium was produced in kilogram amounts at Oak Ridge using calutrons (Ref. 9, pp. 176-179). Each calutron had a magnet diameter between 4 and 5 m and was capable of producing approximately 100 mg of ^{235}U per day. Over 1000 calutrons were used to make enough HEU for 1 or 2 weapons over a 3-yr period. The cost was over 1 billion dollars and was very energy intensive (approximately 3800 kWh/SWU). The enrichment factor, defined as isotopic ratio at two successive stages in the process, was approximately 20 in the early Y-12 calutrons. Improvements in calutron design since 1945 have improved the enrichment factor to greater than 30, but energy requirements still are large. Collection efficiency (the amount of product that actually arrives at the collecting slit) is generally <15% of throughput.

2. Gaseous Diffusion.[9,10] The gaseous diffusion process is based on the principle that the velocity of a gas is dependent on its molecular mass, with lighter molecules moving slightly faster. The ratio of the velocity of two gases depends on the square root of the ratio of the masses, so that for $^{235}UF_6$ and $^{238}UF_6$ the ratio is

$$V235/V238 = \sqrt{352/349} = 1.0043 \ .$$

This is the ideal diffusion separation factor.

The diffusion barrier must be able to differentiate between the two molecules. Barrier design and materials and their mode of manufacture generally are classified, but sintered nickel powders, and other materials have been used (Ref. 10, p. 124). The amount of barrier material is large, typically 1 m^2/kg SWU. A single large stage with a capacity of 12 000 SWU/yr requires approximately 12 200 m^2 of barrier. An ideal diffusion cascade to produce 3% enriched uranium would require approximately 1300 stages.

The time for a diffusion plant to reach equilibrium is relatively long, 11-21 days. To produce weapons grade ^{235}U would require over 3500 stages and an equilibrium time of 1 yr.

The diffusion process also is energy intensive, requiring 2300-3000 kWh/SWU.

3. Centrifuge Enrichment.[9,10] Centrifuge enrichment is based on the difference in centrifugal force on molecules of different weight in a rotating cylinder, with heavier molecules moving to the outside of the cylinder. Centrifuge enrichment was pursued in the US and in Germany during World War II, but development was slower than gaseous diffusion due to several severe technical problems (Ref. 8, pp. 130-132):

a. Design of the rotating chamber had to allow for internal reflux so that all of the gas does not concentrate at the outer wall with the high rotational speed.

b. The high rotational speed caused critical vibration problems by setting up critical rotational frequencies that could destroy the cylinder. These frequencies had to be damped and new bearings designed to prevent wobble.

c. The high rotational speed leads to severe mechanical stress in the outer cylinder wall. This stress exceeded the tensile strength of materials that were available in the 1940s and 1950s. High strength maraging steel and graphite-fiber composites were not available until the late 1950s or 1960s. Although maraging steel and high strength graphite are under strict export control requirements, Pakistan and Iraq were able to obtain the materials clandestinely.

d. Sophisticated computer programs were required to solve hydrodynamic countercurrent flow equations to optimize centrifuge design to increase separative power.

Using optimal design, hypothetical separation factors (the degree of separation in a single element or stage of a separation process) of 1 to 2 or greater can be realized. Eleven stages are required to produce 3% enriched uranium. For a plant of 1000 t SWU/yr approximately 66 000 hypothetical centrifuges having a separation factor of 1.51 are required.[9]

The energy requirements are at least a factor of ten lower than for diffusion, typically 100-300 kWh/kg SWU.

The time to reach equilibrium is much shorter than for diffusion, approximately 2.5 min/cascade.

4. Laser Enrichment. Laser enrichment processes are based on the slight shift in absorption frequency for the different isotopes of uranium and can provide high enrichment factors. If the frequency of incident light can be made sufficiently narrow to excite the ^{235}U but not the ^{238}U atom or molecule, then separations could be affected. Both atomic (AVLIS) and molecular (MLIS) processes have been studied. The molecular process involves the use of UF_6 gas, whereas the atomic process uses vaporized U metal. The vaporized metal has more complex materials handling problems. The development of suitable lasers appears to favor the atomic process and may have been the basis for the US decision to concentrate its R&D efforts on the AVLIS process.

In either process, laser energy costs are expended primarily on the ^{235}U, with little going into ^{238}U. Thus conceivably energy costs are low compared to diffusion and aerodynamic processes, in the range of 10-50 KWh/SWU. Only a few stages would be required to produce HEU.

A demonstration AVLIS process has been developed at Livermore; commercialization is not planned at this time. Other countries, notably Japan, France, and China, are developing AVLIS technology if not facilities.

5. Aerodynamic Processes. In aerodynamic processes, uranium isotope separation is based on the centrifugal forces in a fast curved flow of UF_6 and a light auxiliary gas, generally H_2 or He. Gas accelerations exceed those in a centrifuge by several orders of magnitude without moving parts. Two aerodynamic processes have been developed, the Becker nozzle in Karlsruhe and the UCOR process in South Africa.

For the Becker process (Ref. 10, pp. 245-266), a jet of gas consisting of 96% H_2 and 4% UF_6 is expanded through a narrow slit. The gas moves at high speed and centrifugal forces cause heavier molecules to move to the outside. The slits are small, nominally 0.2 mm, with the lighter ^{235}U fraction skimmed to the inner portion of the nozzle.

The nozzle process is energy intensive because of the pumping and pressurizing of large quantities of gas. Costs of 3000-3500 kWh/SWU have been quoted. The advantage of the process lies in the higher separation factor of 1.015 compared to 1.004 for diffusion. This results in smaller scale and lower holdup with consequent faster attainment of equilibrium.

The South African UCOR process (Ref. 10, pp. 143-146) has been described as "a combination of a stationary-wall centrifuge with a highly asymmetric cut and a cascade system (the helicon system), which is eminently suited to asymmetric separation." A module with a capacity of 10 t SWU/yr would be approximately 3.6 m in diameter and 10 m in length. Energy consumption of the UCOR process is comparable to the jet nozzle. The equilibrium time to achieve 3% enrichment is approximately 16 hours. The small inventory makes the process well suited to produce HEU.

The South African pilot enrichment plant at Velendaba began operation in 1971 and produced its first HEU in 1977. From 1978 to 1989 South Africa produced six nuclear weapons with enough HEU for a seventh. Each weapon was estimated to contain 50-60 kg of HEU and have a yield of 10-18 kt.[11]

6. Chemical Enrichment. Chemical enrichment methods are based on the small difference in oxidation/reduction potential of ^{235}U and ^{238}U. The U(III), U(IV), and U(VI) ions have different chemical properties. Two chemical enrichment methods have

been developed to the pilot plant stage: the ion exchange method in Japan and the solvent extraction method in France.

The Japanese ion exchange process (Ref. 9, pp. 150-153) uses a proprietary ion exchange resin and chemical reduction/oxidation between U(IV) and U(VI) to separate ^{235}U and ^{238}U. The process has a reported single-stage separation factor of 1.0013 (about one-third of that of the gaseous diffusion process), but the stage holdup time is approximately a factor of 10 lower than for diffusion, and energy consumption also is lower, approximately 400-700 kWh/SWU. The technique is touted as being proliferation resistant because of potential criticality problems at high enrichments. These could possibly be overcome by use of borated glass or annular columns. Kilogram amounts of 3% enriched uranium have been produced.

The French solvent extraction process was developed to pilot plant scale in the mid 1980s. The process is based on U(III)/U(IV) oxidation/reduction and solvent extraction with tributyl phosphate in an organic aromatic diluent.The project was abandoned because the French did not believe that it could compete economically with centrifuge enrichment or the AVLIS process. The equipment was sold and reportedly eventually purchased by Iraq and installed at Tuwaitha for further research. The single-stage separation factor is 1.0025-1.003. In spite of the low separation factor, the process has some appeal because of the low specific energy consumption of less than 600 kWh/SWU.

Parameters of importance to production of HEU by various enrichment processes are summarized in Table III. In general, facilities based on separation processes having single-stage separation factors <1.01 will be large, complex, and expensive to build and operate. Facilities based on aerodynamic processes (including centrifuge) have relatively high single-stage separation factors and will be smaller to build and less expensive to operate but will require sophisticated technology. Because of ease of batch recycle, the time to obtain weapons-grade uranium will be relatively short.

B. Plutonium Reprocessing

Plutonium was first recovered from spent fuel for weapons purposes but is now of interest for use in power reactors. A number of processes have been developed to recover plutonium from spent fuel, both for weapons and reactor use purposes.

1. Bismuth Phosphate.[13] The first process developed to recover plutonium from spent fuel was the bismuth phosphate process. It was a batch process requiring several precipitation/dissolution steps to attain the required decontamination factor. The process is comparatively easy to scale up to plant operations and requires only simple equipment, and remote maintenance and operation are feasible. Disadvantages are (1) it does not recover uranium, (2) large amounts of waste are generated, (3) many batch operations are required, and (4) plutonium recovery is only about 95%. These disadvantages would be of minimal concern to a potential proliferant.

TABLE III. Parameters of Importance to Proliferation Through Various Uranium Enrichment Processes

Process	Single-Stage Separation Factor	Specific Energy Consumption (kWh/SWU)	Relative Size of Facility	Ease of Batch Recycle
Calutron		3000–4000	Large	Medium
Gaseous diffusion	1.0040–1.0045	2300–3000	Large	Complex
Centrifuge	1–2	100–300	Small	Easy
Laser				
Molecular		10–50	Small	Easy
Atomic		10–50	Medium	Complex
Aerodynamic				
Nozzle	1.015	3000–3500	Medium	Medium
Helikon	1.025–1.03	3000–3500	Medium	Easy
Chemical				
Solvent extraction	1.0025–1.0030	<600	Large	Complex
Ion exchange	1.0013	400–700	Large	Complex

2. Solvent Extraction.[13] Several different solvent extraction processes have been developed to extract plutonium and uranium from fission products. Early work was directed to recover plutonium for weapons use, hence the fuel was generally uranium metal or a uranium-aluminum alloy.

The early Redox process used hexone as the organic phase; aluminum nitrate was used as a salting agent to minimize extraction of fission products.

The Purex process uses 30% tri-n-butyl phosphate (TBP) as the extractant, generally dissolved in kerosene. The salting agent is HNO_3, hence wastes are reduced relative to the Redox process.

Thenoyl trifluoroactene in hexone also has been studied but again requires aluminum nitrate as a salting agent.

The Purex process has been much studied and developed through the past 40 years and is the basis for all commercial reprocessing plants today. The process generally has three solvent extraction cycles. The first cycle separates uranium and plutonium from the bulk of the fission products; the second cycle partitions uranium and plutonium; and the third cycle provides additional fission product and uranium decontamination and some concentration of plutonium.

Major improvements in the process have been in improving plutonium and other actinide recoveries and in introducing newer solvent extraction contactors. Pulsed columns and mixer settlers can be replaced by centrifugal contactors, particularly in the first decontamination cycle where radiolytic decomposition of the solvent could result from long contact times with resultant loss of plutonium with fission products.[14] The centrifugal contactors have shorter contact time and if used in all three cycles could significantly shorten throughput time for plutonium reprocessing.

Between 1976 and 1979, Pacific Northwest Laboratories operated a small reprocessing plant for waste vitrification demonstration purposes.[15] The facility had a capacity of 50 kg/day of LWR fuel or 40-50 g/day of plutonium. Pulsed columns were used to decontaminate uranium and plutonium from fission products and ion exchange to separate uranium from plutonium. The primary plutonium column had a capacity of 450 g of plutonium per batch, approximately the amount contained in 50 kg of fuel (one day of throughput). Thus, a significant quantity could be processed in 18 days.

3. AIROX Process.[16-18] The AIROX process is a dry process for extending the life of spent fuel and, strictly speaking, is not reprocessing because there is no attempt to separate uranium and plutonium from each other or from all of the fission products. In the AIROX process, the spent fuel is chopped and oxidized to convert from UO_2 to U_3O_8. In the process approximately 50% of the fission products are removed, mostly in the form of Kr, Xe, Cs, I, Ru, Tc, Cd, and In, with some Mo and Nb. The U_3O_8 is then reduced to UO_2, and the UO_2 and PuO_2 along with the remaining fission products are sintered into pellets for reuse as fuel.

4. Molten Salt Processes. Molten salt processes have been investigated for recovery of plutonium from either metallic fuels (uranium-plutonium-zirconium alloys) or LWR oxide fuels.

a. LWR Oxide Fuels.[19] The method proposed for molten salt reprocessing of plutonium from spent LWR fuel involves electrolytic generation of metallic lithium or calcium in a molten salt medium. The metal reduces the actinides to the metallic state. The uranium is precipitated from the transuranic actinides from a liquid metallic solution of, for example, An-Mg alloy. The technology has been demonstrated on bench scale but has not scaled up engineering development.

b. Metallic Fuels.[20] The Integral Fast Reactor (IFR) fuel cycle is based on a metallic fuel alloy with a nominal composition of U-20%Pu-10%Zr. A pyrochemical processing scheme has been developed for recovery of plutonium and uranium from the spent fuel. The chopped fuel is placed in a perforated steel basket, which becomes the anode in a molten salt mixture, typically a LiCl-KCl eutectic. The actinides and fission products are dissolved in the molten salt. The actinide chlorides are transported through the molten salt; uranium is collected on a solid cathode; and transuranics with some uranium are recovered in a molten cadmium cathode. With a chemical reduction step, the method can be adapted to reprocessing of spent LWR fuels.

c. Magnetic Recovery of Actinides. Because all actinide compounds are paramagnetic, magnetic separation of actinide-containing mixtures is possible. Magnetic separation of recycle plutonium process residues has been demonstrated on an open-gradient magnetic separator.[21] The method has not been demonstrated on an engineering basis.

5. Preparation of Plutonium Metal. A number of routes have been developed for preparation of plutonium metal from the oxide.

a. Halide Reduction. Halide reduction has been used since the first metal was prepared. Either calcium or lithium can be used to reduce PuF_4, PuF_3, $PuCl_3$, $PuBr_3$, or PuI_3.[22] Calcium is preferred because it is easier to handle than lithium. PuF_4 is preferred because it is not hygroscopic and therefore requires less care. The reaction can be carried out in a refractory-lined bomb using iodine as an initiator to provide yields greater than 99%.

b. Direction Oxide Reduction.[23] PuO_2 can be reduced directly to the metal using either lithium or calcium metal. The PuO_2 is mixed with calcium metal and $CaCl_2$ in a magnesia crucible and heated to 800°C and stirring with a tantalum stirrer. The reaction is carried out under argon gas. The CaC_{l2} dissolves the CaO, yielding plutonium metal of the same purity as the oxide feed.

c. Electrorefining. Electrorefining is used to convert impure plutonium metal to high purity plutonium metal. The technology is similar to that described for recovery of plutonium from spent fuel but much simpler because the major impurities have been removed.

IV. CONCLUSIONS AND RECOMMENDATIONS

Significant technical improvements since 1970 have been achieved both in isotope separation of uranium and in reprocessing of spent reactor fuel to recover plutonium. Enrichment technology today is capable of producing weapons quantities of ^{235}U in matters of months using, for example, the nozzle process as demonstrated by South Africa or the centrifuge process as applied by Pakistan. As noted by Cohen and Benedict,[24] separation of ^{235}U from natural uranium by both methods is technically less difficult than recovery of plutonium from irradiated fuel. Detailed information on equipment designs for both processes are available in the open literature. If LEU is available as a starting material, weapons-grade uranium is even easier to obtain (approximately 80% of the separative work required to obtain 93% enriched ^{235}U has been performed in producing 3% enriched material). With available enrichment facilities, the time to obtain weapons-grade uranium from natural uranium or LEU is less than one year.

The conversion times for plutonium appear to be reasonable for present day plutonium handling technology. This applies to plutonium in spent fuel (1 to 3 months) with the present day practices of the IAEA of monthly inventories for reprocessing facilities.

It has been argued that "no nation has ever considered making weapons out of plutonium from US-type reactors" (the US announced in 1977 that it had exploded a weapon from reactor-grade plutonium), and "[reactor-grade plutonium] can be made to explode, but it is of no interest as a bomb."[25] However, from nuclear weapons design considerations, there is no incentive to treat reactor-grade plutonium differently than weapons-grade plutonium, either in terms of goal quantity or conversion time.

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