LEAD SUBSTITUTION AND ELIMINATION STUDY, PART II

Timothy P. Martinez, and Michael E. Cournoyer, Ph.D.*
Los Alamos National Laboratory,
Los Alamos, NM 87545
(505) 665-7616

ABSTRACT

Within the Nuclear Materials Technology Division of Los Alamos National Laboratory, lead is used as shielding for a variety of operations, including actinide chemistry, weapons production, radiochemistry, and analytical chemistry. In this study, waste minimization issues associated with replacing lead shielding with non-hazardous materials are addressed. These include institutional program available to support this effort, the hazards and accompanying controls grouped with lead shielding, operations that use lead bricks and how this effects the selection of the substitute. Life cycle management issues are also examined. As a final step, an approach to get buy-in from both technical and budget minded employees is presented.

INTRODUCTION

The control of hazardous materials is an ongoing process that starts during the design phase of a process or facility and continues through the performance of daily job tasks. At Los Alamos National Laboratory (LANL), hazards involving materials are controlled in a variety of ways. All of these methods fall into one of five categories (1). These categories, ranked in the order of preference, are elimination, substitution, engineering controls, administrative controls, and Personal Protective Equipment (PPE). Elimination is the complete removal of a hazardous material. It is often done in the planning stages of an operation or facility. Substitution is the replacement of a highly hazardous material with a less hazardous one.

Relative to the organization of material presented here, the objective is to provide a sense of perspective concerning the utilization of institutional programs, specifically pollution prevention and chemical tracking, for addressing hazardous material protection issues relative to elimination and substitution requirements. To this end, using lead shielding as an example, major factors are identified. These include programs at LANL that are available to effectively meet these requirements, hazards associated with lead shielding, operational conditions that influence the choice of shielding replacement, commercially available substitutes, actually versus theoretical cost of elimination, and life-cycle issues. Each factor is assessed in general terms relative to criteria used to select a lead substitute. The intent is for the advantages of replacing lead with non-hazardous materials to become apparent to employees involved with working in a nuclear facility. While personnel involved with waste minimization and industrial hygiene will support this effort, arguments will be presented that will motivate the chemical researcher and line manager as well.

INSTITUTIONAL PROGRAMS AND TOOLS

As part of LANL's Environmental Science and Waste Technology Division Division, the Environmental Stewardship Office (E-ESO) manages numerous lab projects including Pollution Prevention (P^2). A main goal of this program office is to add environmental considerations to operational decisions along with such traditional factors as performance, price, health, and safety. Operational divisions like Nuclear Materials Technology (NMT) Division are required to incorporate these P^2 goals and have performance measures to reduce hazardous and mixed waste generated from operations as much as is technically and economically feasible. Eliminating or substituting a less hazardous material in operation is covered under the Hazard Evaluation and Elimination Program element of the division's Hazardous Materials Protection Program (HMPP). Since the magnitude of a risk involves both the probability and severity of the associated harm, it can be reasonably assumed that eliminating a hazardous material can reduce the probability of the harm. Funding for these types of efforts are obtained from the waste minimization program office discussed above. At NMT Division facilities, the primary use of metallic lead is the shielding of against primary gamma rays and fast neutrons. Finding ways to eliminate lead or substitute lead with a non-hazardous material not only meet the objectives of these programs, but also is a proactive step in protecting personnel, facilities, the environment, and the public from hazardous materials.

Another requirement of the HMPP is maintaining an inventory of all hazardous materials. Using the LANL Automated Chemical Information System (ACIS) operated by the Industrial Hygiene and Safety Group (ESH-5), a list of all locations, amount, and operations involving hazardous materials may be obtained. By using this system to track chemicals that require shielding, all lead shielding operations are effectively targeted. ACIS also maintains a database of hazards associated with each material. Information from this database was also used to assess the hazards discussed in the next section.

LEAD SHIELDING HAZARDS (2)

The three principle hazards associated with lead bricks are the following:

- **Health** Lead is a cumulative and persistent toxic substance that poses a serious health risk.
- **Ergonomics Factors** The excessive weight of solid lead bricks has the potential to mash fingers and toes and damage equipment.
- **Environmental** Under the Resource Conservation and Recovery Act (RCRA), lead shielding is a D008 listed waste and is regulated by law and has strict storage and disposal requirements once the lead brick becomes waste.

NMT DIVISION OPERATIONS WITH LEAD SHIELDING

Primary Gamma Rays

Due to its high density, lead, especially in brick form, has been used for shielding against primary gamma rays. Many NMT Division operations generate large amounts of gamma radiation with the main sources of gamma rays coming from various isotopes of uranium and plutonium. With freshly purified plutonium, most of the radiation comes from *soft* (17-keV) x-rays. More penetrating (60-keV) gammas are emitted by ^{241}Am, which *grows in* as ^{241}Pu decays. All grades of plutonium contain ^{241}Pu. In plutonium, that is more than 10 years old (since purification), these gammas are usually the source of most of the external radiation. Although all the major uranium isotopes decay by the emission of alpha particles, they are sometimes accompanied by gamma ray emissions. Most of the high-energy gamma rays, which cause a deep dose (an external radiation dose that penetrates to the internal organs), arise from the daughters that *grow in* as the uranium ages. Important daughters are thorium, protactinium, radium, and radon. Gamma radiation dose rates from a large sheet of most types of uranium are generally less than 5 mrem/hour. Uranium-233 (with its uranium-232 contaminant) is an exception. The daughters of ^{232}U and ^{233}U emit high-energy beta particles and gamma rays resulting in a dose rate of several rem/hour. As the ^{232}U decays, the concentration of daughters increases, causing the dose rate to increase by about a factor of 10. Other miscellaneous projects such as Hanford Waste and PORTS (Portsmouth Gaseous Diffusion Plant) generate gamma rays resulting from the following sources: Technetium-99, Neptunium-237 (Pr-233), Cesium-137, Cesium-137, Cobalt-60, and (alpha, n).

Fast Neutrons

Fast neutrons are most effectively shielded by hydrogen. Fast neutrons are slowed to thermal energies by collisions with hydrogen atoms. NMT operations that use lead brick for this application include the Pit Manufacturing Project. Neutron shield for glove boxes is limited to about 6 inches (15 cm) due to the physical limitation of most workers (3). Lead slow fast neutrons by inelastic scattering at the higher energies. Comparing lead against other fast neutron shielding materials is beyond the scope of this report and will not be discussed.

SHIELDING MATERIAL CRITERIA

Primary Interaction Mechanism (4)

There are three primary types of interaction mechanism that contribute to the effectiveness of materials used as shielding: Photoelectron Effect (PE), Compton Effect (CE), and Pair Production (PP). The PE interaction is observed with low-energy x-rays and gamma rays less than 750 keV. The effect is based on the atomic number (Z) of the shielding material (Z**4), since the higher the atomic number the greater the electron density. The CE interaction is observed with x-rays and gamma rays with energies between 750 keV and 5 MeV. This effect is dependent on density alone. The PP interaction is observed with high-energy x-rays and gamma rays greater than 5 MeV. The effect is also based on the atomic number of the shielding material (Z^2+Z), but not as much as the PE interaction. Since no operations in NMT Division work with sources that exhibit this effect, no substitution material will be discussed, although bismuth would be the material of choice. As reported previously, Uranium-233, Plutonium-239 (fresh), Amercium-241, Technetium-99, Neptunium-237 (Pr-233), and Cesium-137 primary interaction mechanism is the Photoelectron Effect. Cesium-137, Cobalt-60, and (alpha, n) primary interaction mechanism is the Compton Effect (5).

The shielding properties of lead (Z = 82, Density = 11.30 gm/cm^3) versus bismuth (Z = 83, Density = 9.80 gm/cm^3) and tungsten (Z = 74, Density = 19.11 gm/cm^3) are compared within the PE range in Figure 1 (6). Based on mass attenuation coefficient data, the density factor contributes more to the shielding effectiveness than the atomic number of the material in the low-energy gamma ray range. For gamma rays with energies over 2 MeV, tungsten is almost twice as effective as lead or bismuth.

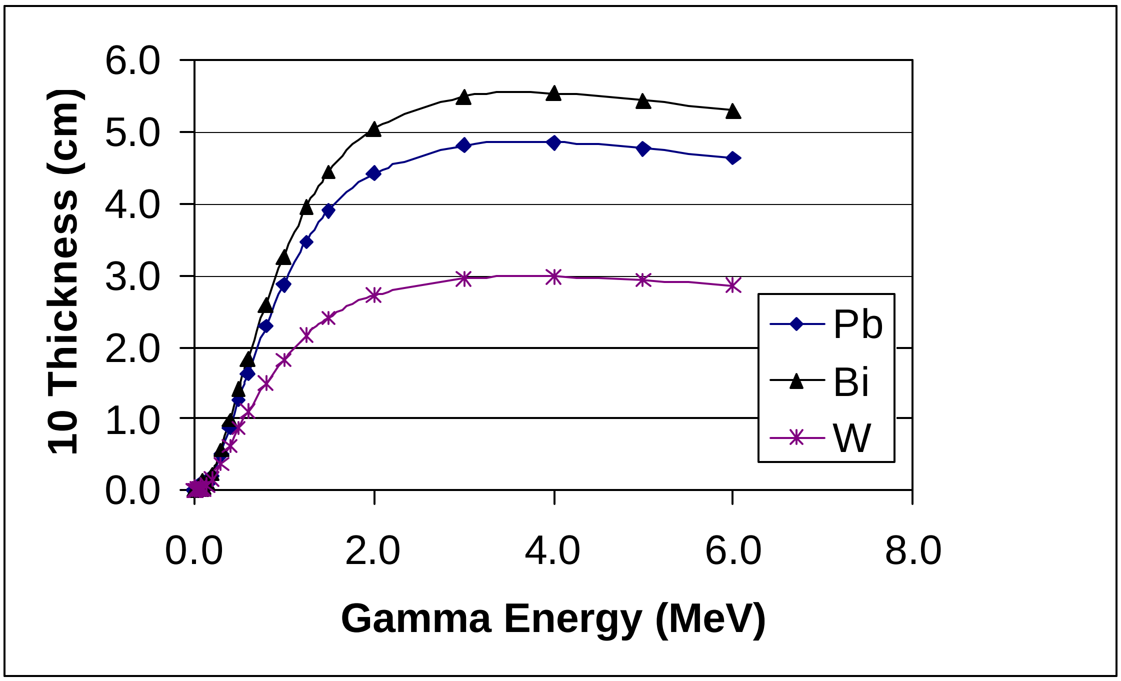

Figure 1. Comparison of Gamma Shielding Materials.

Non-hazardous Material Substitutes

The previous discussion on interaction mechanism is important in selecting the appropriate substitute. For shielding against lower-energy sources that undergo the PE, bismuth, a non-hazardous replacement for lead with a higher atomic number, provide sufficient protection without hampering the productivity of the worker. For medium range radiation sources that undergo the CE, tungsten with a density, almost twice that of lead is the non-hazardous material of choice.

A case has been made for replacing lead with alternative materials on shielding efficiency. Bismuth and tungsten are less hazardous than lead for the following reasons:

- No exposure level has been established for bismuth. The 5 mg/m^3 value for tungsten is equivalent to nuisance dust associated with rust. In other words, the simple movement of bricks exposes one to lead, while operations such as grinding are needed for generating hazardous levels of tungsten.
- Bismuth and tungsten do not pose an environmental hazard, while lead is listed as a RCRA waste (as discussed earlier).

Coating Lead Shielding (Substitution)

Coating lead shielding with a metal or polymer coating converts the hazardous constituent, lead, into a less toxic form. *Canned lead* shielding provided by Westinghouse or General Electric as part of a tooling package is used widely. Canned lead is lead to which Stainless Steel cladding has been applied and in which the lead is then contained. A temporary, strippable coating made from polyurethane that can be left in place for later removal is also commercially available. Other organic polymer coatings include lead sheeting coated with vinyl. Metal coating would be preferred over organic polymers because they are relatively immune to radiation damage. It would be difficult to find a polymer that can hold up for 20 years against the abrasion and radiation conditions to which lead bricks might be subjected. Nevertheless, it is safe to say that strippable and durable polymer coatings are applicable for operations where the shielding will not be exposed to high-level radiation. There is an issue regarding the removal of a coating without transferring radioactive particulates. Metal-coated surfaces are easier to clean than those coated with lead because metals used as coatings can produce hard, smooth surfaces while lead itself is soft, easy to scratch, and hard to clean.

Composite Shielding

Shielding materials composed of homogeneous mixture of lead, tungsten, or bismuth and an inert polymer, polyethylene, are commercially available. Composite bricks made from lead and polyethylene have been designed that provide adequate effectiveness against gamma radiation and mitigate the toxic and ergonomic hazards associated with lead bricks. For example, commercially available product made from lead and bismuth called *LIGHT-LEAD* and *Bix-light*, respectively, have densities one-quarter that of lead. *Ecomass* is a tungsten material with the same density of lead. Following the same method used in Figure 1, the shielding properties of these composite materials are compared against pure lead in Figure 2. Thus, for the lead and bismuth composites the relative shielding effectiveness is reduced to one-quarter that of lead. Note: when bismuth and lead materials have the same density, bismuth is slightly more effective than lead at shielding. This type of composite shielding can reduce the number of injuries associated with improper lifting and dropping a lead brick on ones hand or foot. These injuries are currently controlled with foot-protection and SOPs that cover hand injuries. For most radiochemistry operations, this type of shielding provides ample protection. The tungsten composite is interchangeable with a lead brick as far as shielding effectiveness and ergonomic hazards are concerned.

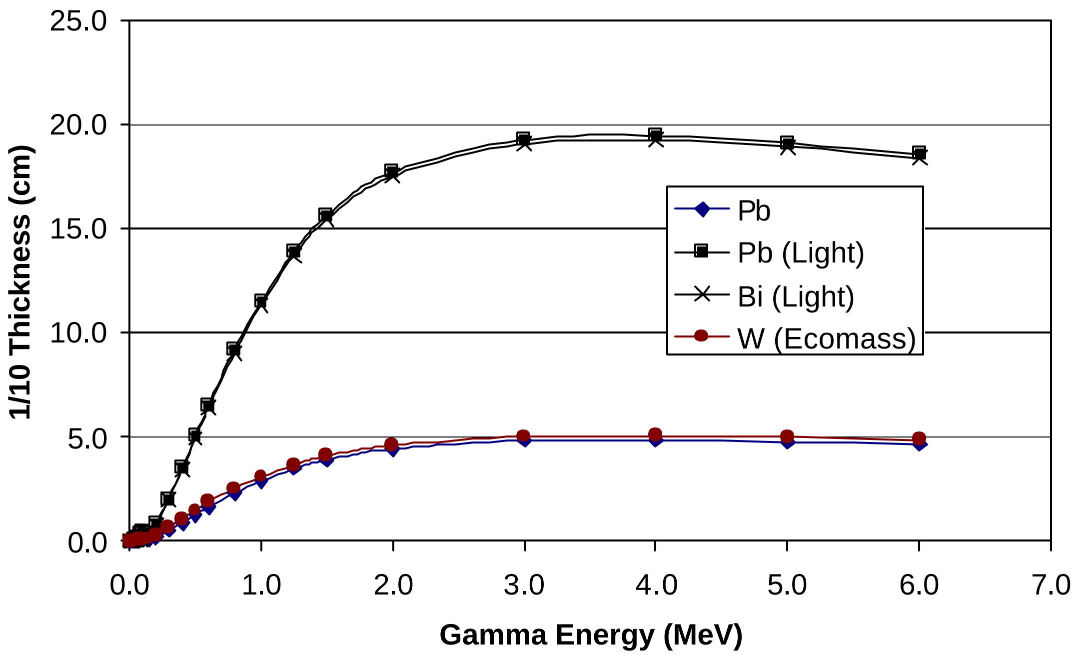

Figure 2. Comparison of Lightweight Gamma Shielding Materials.

Direct Cost Comparison

In a previous paper, direct cost comparisons for lead replacement materials were reported based on prices for the commodity metals.[e] The cost for substituting bismuth and tungsten were 14 and 30 times the price of lead, respectively. These values should be considered theoretical because the cost of obtaining lead and lead substitutes from commercial sources is considerably higher as shown in Table I (7).

Table I. Life Cycle Costs for Shielding Materials.

Brick Material	Weight (kg)	Z#	Cost per Brick* Theo.	Actual	Disposal	Total**
Lead	12	82	$6	$28	$139	$167
Light Lead	4	82	$12	$58	$43	$101
PVC Coated Lead	12	82	$12	$207	$139	$346
Light Tungsten	12	74	$104	$312	$0	$312
Light Bismuth	4	83	$25	$65	$0	$65

*Brick dimensions = 5 cm x 10 cm x 20 cm

**Total = Actual (Theo. If Actual not available) + Disposal

The brick dimensions were assumed to be 5 cm x 10 cm x 20 cm. Concerning the theoretical costs of the coated lead bricks, it was assumed that this process double the price of the original brick. Light bricks were estimated based on the amount of metal.

No additional disposal costs are associated with shielding made from tungsten or bismuth. On the other hand, lead is an RCRA-listed metal and the average additional cost of disposal at LANL is estimated at $139 a brick made from 100% lead. The actual cost varies with the circumstances; however, the average cost for onsite waste handling and offsite disposal is $11.75/kg. Comparing the cost of the light bismuth and lead brick directly, the former is less expensive without sacrificing shielding effectiveness. When the disposal cost is added to the original cost of the lead brick (~$167), the light tungsten brick is about twice the cost, although the shielding effectiveness is similar. Therefore, short-term use of lead bricks is more financially attractive than a tungsten substitute. Interestingly, the PVC coated lead brick is even more expensive than the light tungsten brick. Overhead-costs of management, intangible costs, and industrial hygiene support only add to the cost of uncoated lead bricks. As discussed in an earlier paper, these support costs for lead bricks could exceeds that of disposal within 10 years. This along with intangible costs of occurrence reports and audit findings would make replacing lead with tungsten for medium-energy x-rays and gamma rays sources prudent decision in the long-term.

LIFE CYCLE MANAGEMENT

If the lead shielding is being replaced by a non-hazardous substitute, the excess lead must be recycled or reused, otherwise it will be disposed of as a hazardous waste, mixed low-level waste (MLLW) or mixed transuranic (TRU) waste. This brings up two important issues that need to be addressed:

- The current moratorium on recycling metal from areas posted for radiological hazards (8).
- Waste with no disposal path.

Currently, there is suspension of the unrestricted release of scrap metal from nuclear facilities destined for recycling. The suspension is part of a new DOE policy aimed at ensuring that contaminated materials are not recycled into consumer products and at improving DOE management of scrap materials at its nuclear weapons production sites. To conform to DOE moratorium, LANL immediately suspended the recycling of all scrap metal originating from any area posted for radiological hazards. This suspension includes lead bricks; however, all scrap metal already in recycling vendor's bins is not affected. This moratorium does not affect the recycling of radioactively contaminated or activated metals that are sent offsite to a DOE recycling facility for intentional conversion into products used in radiological facilities or activities.

Non-defense mixed TRU waste, like contaminated lead bricks, is considered waste streams with no identified treatment or disposal path. Lessons learned from recent off-normal events, such as glove box glove failure have caused contamination outside the glove box (9). These operating experiences clearly show how lead brick, which are normally a hazardous waste, can quickly be transformed into a mixed-waste problem, including mixed-waste without a disposal path. These and other issues that have been raised are now addressed in the following section.

DISCUSSION

Environmental Considerations

Ideally, environmentally preferable products are one of the following: bio-based, bio-degradable, energy efficient, water or energy conserving, reusable, recycled- containing post-consumer material or recovered material, recyclable- consisting of components that can be completely recycled, free of pollutants, packaged with minimal (recyclable) materials. Bismuth and tungsten shielding materials meet some of these criteria by being free of pollutants and energy conserving (reduction of industrial hygiene support). As discussed below, the surplus lead is also recycled. Operations that are currently going through the approval process are being reviewed for lead shielding considerations. Gloveboxes that in the past have been lead-lined are being reevaluated. By removing lead as a shielding material in the planning stages of an operation, all the cost associated with an adequate Lead Management Program (training, medical surveillance, PPE, industrial hygiene support, work control issues) are avoided.

Mitigation of Hazards

Once lead shielding is adequately coated, the toxic effects and incompatibility hazards of lead are nullified. Chemical workers need not be on expensive medical surveillance programs, and industrial hygiene support which includes characterizing workplace air concentrations, worker identification, assistance to operating groups to control lead exposures, and exposure assessments. Initial and annual lead awareness training is still required. The integrity of the coating should be inspected annually, especially the non-metal coating. Hazards associated with falling objects are addressed by reducing the weight of a lead brick by one fourth. Shielding materials, made from lead and bismuth mixed with polyethylene, have been designed that provide adequate effectiveness against gamma radiation and mitigate the ergonomic hazards associated with lead bricks. This type of composite shielding can reduce the number of injuries associated with improper lifting and dropping a lead brick on ones hand or foot. These injuries are currently controlled with foot-protection and SOPs that cover proper lab techniques that minimize hand injuries. Ergonomic improvements are sacrificed at expense of shielding effectiveness, as shown in Figure 2. The effectiveness is reduced linearly with the lightness of the brick. For most radiochemistry and similar operations, this type of shielding provides ample protection. For high radiation operations, other ergonomic factors become prominent (15 cm thickness limitation) and risk of exposure is higher than a minor injury. This leaves the hazards that adversely impact the environment. No matter how many times you recycle the lead bricks they must be disposed of as a RCRA waste at some point.

Prevention of Hazards

There are no toxic or environmental hazards associated with shielding made from tungsten or bismuth. Training, designated areas, work control documents, decontamination procedures, and industrial hygiene support are not needed. Once a project has concluded, these shielding materials can be disposed as non-hazardous waste. Also, a sometimes-overlooked consideration, no additional hazards are introduced by replacing lead with tungsten or bismuth.

Drivers for Lead Replacement Materials

Asking a researcher to substitute lead with something more expensive for health and environmental reasons is a difficult task. Especially if he or she has been working with lead for several years. Argument are more convincing if the following technical merits are pointed out the following:

- Tungsten is more effective shielding material than lead against medium-energy sources that exhibit the Compton Effect because tungsten is denser
- Bismuth is more effective shielding material than lead against lower-energy sources that undergo the PE because bismuth has a higher atomic number

On similar note, buy-in from the finance-side of the organization is easier to obtain, once disposal and industrial hygiene costs are factored in. Line managers will find the reduction in training time for their workers and elimination of liability associated with any hazardous material more appealing than that they are meeting the performance requirements of the institution.

Waste Without A Disposal Path

Using a LANL-developed technology, lead is decontaminated to free release ($2.2/kg), and it could also be sent to a smelter once the DOE moratorium unrestricted release of scrap metal from nuclear facilities is lifted (10). Another option is possible when the operations of the Waste Isolation Pilot Plant (WIPP) are expanded to include non-defense generated mixed TRU wastes. Legislation has been proposed for such a move.

DOE Moratorium

Regardless of the moratorium on recycling of radioactively contaminated metals being lifted, sending lead bricks originating from area posted for radiological hazards to Oak Ridge National Center of Excellence for Metal Recycle would solve this problem. The National Center of Excellence for Metal Recycle is an Oak Ridge, Tennessee-based, company has successfully demonstrated a process for reusing contaminated lead as a shielding material for radioactive waste containers. This process offers the DOE a cost-effective strategy for reusing a material that would otherwise require costly disposal as a mixed waste. GTS-Duratek Inc. has already recycled nearly 21 tons of potentially contaminated lead into shielding for seven large steel containers destined for use in the safe storage of radioactive waste. The lead originated from the DOE's Hanford Site in Washington State. The new containers are destined for ultimate use at DOE's Pacific Northwest National Laboratory (PNNL), also located in Hanford. A similar operation can be developed for LANL TRU waste drums destined for WIPP.

CONCLUSIONS

When practicable, complete elimination of lead in the workplace, especially at nuclear facilities, is desired. Coating lead bricks mitigate the toxic hazards and cost associated with control the hazard. Relief from ergonomic hazards is accomplished by using less-dense bricks with a corresponding reduction in shielding efficiency. Bismuth and tungsten are more effective shielding materials and eliminate the toxic and environmental hazards associated with lead without adding hazards. Disposal cost associated with lead and long-term overhead costs justify more expensive commercially available non-hazardous substitutes. Explaining the benefits of the shielding efficiency of the lead substitutes to researcher is more effective than making a case for the health and environmental features. By using Oak Ridge National Center of Excellence for Metal Recycle to recycle the lead bricks for later use in WIPP storage drums and a LANL decontamination technology, Life Cycle issues are by-passed. Finally, through implementation of the Hazard Evaluation and Elimination Program element of the HMPP, NMT Division contributes to the LANL's performance measure by effectively collaborating with two LANL support organizations, ESH-5 and E-ESO.

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