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10

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# TARS, SPENT CATALYSTS, AND COMPLEXES AS PETROCHEMICAL WASTE SOURCES *

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This is one of several papers from an Industrial Wastes Symposium titled, "The Petrochemical Industry." The first three papers were published in THIS JOURNAL (31, 3, 274; 31, 5, 574; and 31, 6, 712). Other papers in the series appear on pages 836 and 846 of this issue.

Tars, spent catalysts, and catalyst complexes are usually disposed of by burning, burying, or by direct discharge to streams. Disposal of these troublesome wastes is not always simple since some of the plants now in operation are highly complex and the result of much expensive research.

Petrochemical processes usually involve the reaction of certain components catalytically to produce a crude intermediate product which is subsequently distilled to produce the desired end-product. These processes, which use petroleum-derived raw materials, produce an almost endless number of chemical products. All these products and processes are included within the meaning of the difficult-to-define term of "petrochemical" or "petrochemical process." In addition, there are many chemical processes using coal-tar-derived intermediates which are similar to the so-called "petrochemical processes."

## Scope of Problem

A hypothetical process may be considered in which an organic liquid *A* and a gas *B* are reacted catalytically to produce crude *AB*. The crude material is subsequently distilled to give the refined product *AB*. There are several places in this process where troublesome disposal problems could arise. The still tars from the distillation step would most certainly constitute such a problem. The spent catalyst, in the form of an organic catalyst complex, a spent-catalyst tar, or spent dry-bed material, would pose another disposal problem, the extent of which would depend on many factors. Before an adequate disposal process could be designed, intelligent answers would have to be found to questions such as:

1. In what form is the spent material when removed from the process?
2. How much of the spent material is produced per unit of time or product?
3. How toxic is the spent catalyst or its combustion products?
4. Can the catalyst be regenerated or can some recoverable product be produced from the spent material?

In considering the disposal of the distillation tars from the simple hypothetical process described, many other factors would have to be taken into account. For example, the consistency (viscosity, specific gravity, etc.) of the tar would have to be known before the necessary equipment to move it from the process site to the disposal site could be designed. The composition of the tar should be understood so that the necessary disposal equipment could

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WHP 42

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be operated without creating air pollution or handling problems. The amount of tar produced per unit of time would have to be determined before designing the size of the disposal equipment.

## Incineration

### *Open-Pit Burning*

Incineration is the method most frequently used for the disposal of troublesome materials and it is probably the oldest waste disposal process known to industry. Some of the present-day uses of this process are no different than they were hundreds of years ago. This does not mean, however, that such uses are wrong or necessarily troublesome. Open-pit burning, which is the simplest and least expensive method of incineration, is as widely practiced today as it was two generations ago. To burn organic materials properly in this fashion requires good planning and proper control, however. Open-pit burning should be done under strict supervision and adequate fire-extinguishing equipment should always be available. The smoke plume from this operation should be kept under observation and the fire extinguished when the wind shifts sufficiently to bring the plume over populated areas. Unsupervised open-pit burning has frequently caused serious air pollution problems to those plants foolish enough to follow this practice.

A modified method of open-pit burning has been developed in the petrochemical industry in recent years. This method employs open ground-level burners mounted in areas where fire is no problem and where the organic tars to be burned can be stored. These burners employ proper oxygen balance and the organic tars can be burned with a minimum of visible evidence. These installations, of course, require pumps, piping, ignition sources, and, very often, a secondary fuel supply but they provide an economical method of incinerating many troublesome wastes.

### *Metallic Materials*

Many catalyst tars, complexes, and distillation residues contain metallic materials. These metals or their oxides will be discharged into the air effluent from the incinerator installation if provision is not made to trap them. When the concentration is very low, the discharge of this material probably does not constitute a hazard because of the dilution effect. However, in many incinerator installations where metallic materials are burned, the metallic residues should be collected and removed from the exit gases before discharge. In this way metallic materials, such as nickel, copper, and platinum, can be recovered for resale and substances which constitute health or corrosion hazards, such as chlorine, nitrogen, and sulfur, may be removed. It is very often necessary to scrub the effluent gases to remove materials of this type.

### *Sulfur and Flyash*

Sulfur-containing tars are probably the most troublesome to incinerate from an air-pollution standpoint. If the exit gas concentration of $SO_2$ is high enough, air pollution is distressingly evident in the surrounding area. The discharge of the exit gases from incinerating chlorinated tars is also very troublesome from the corrosion standpoint. These gases have a tendency, when discharged into humid atmospheres, to hydrate quite rapidly and settle out as a hydrochloric acid mist. The discharge of small quantities of oxides of nitrogen or sulfur does not ordinarily constitute an air pollution problem if these materials are dilute enough in the exit gases. The maximum downwind concentrations of these compounds can be calculated by using mathematical formulas such as the Sutton or the Bosanquet-Pearson equations (1) (2) (3) and, where troublesome materials are to be discharged, such calculations should always be made.

Another problem that frequently arises in the incineration of petro-

WHP 42



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chemical tars and complexes comes from the flyash produced by incinerating tars that contain inert materials. Tarry chemical residues frequently contain filter aids and other inert materials added during the process operation. These materials cannot be readily separated from the tars before disposal and, when the tars are incinerated, these inert substances are invariably discharged in the exit gases as solid particulate matter. If the amount of this particulate matter is great then it must be removed from the exit gases. Calculations of the possible ground concentrations aid in deciding what collection equipment is necessary. Very often a settling chamber, placed downstream from the combustion chamber, will serve to remove this material.

*Design Requirements*

Balanced incinerators of many different types have been designed and constructed to perform a great variety of burning jobs (4) (5). In general, they must be designed to fit the variables encountered in the specific waste-disposal project. Most chemical incinerators are designed for multi-purpose burning and frequently have provision for trash burning included as part of their design. If a balanced incinerator is needed, it should be designed, if possible, to perform all waste-burning tasks necessary in the plant. It is very often feasible to recover waste heat value from incineration of flammable materials through waste heat boilers, pre-heating equipment, etc. If the materials to be burned have acceptable characteristics, it is also possible to use these materials directly as fuels in fuel-burning equipment.

Great care must be taken in the design of balanced incinerators to be used for burning chemical tars. Such variables as the reaction time, stack height, construction material, burner sizes, recycled air, and many others must be carefully considered. For example, the type of burner selected is of great importance because these burners must accommodate the materials to be burned unless they are to be mechanically placed in the fire chamber for burning. If the proper burner is available, it is very often possible to feed highly viscous tars by steam or air atomization through burners after this material has been pre-heated to the point where consistency suitable for pumping is obtained.

The secondary air and fuel supply to balanced incinerators is important because the incinerator must have sufficient air to oxidize completely all materials that are to be burned. A secondary fuel supply is also necessary to maintain combustion temperatures and to level out uneven waste loads and prevent heat-shocking of the incinerator refractory.

In the construction of incineration equipment, the type of interior refractory used is of importance. It must be capable of withstanding the anticipated heat and corrosive attack.

The height of the stack used for combustion gases from an incinerator should also be designed with great care. The downwind concentration of any material that leaves the incinerator stack is influenced, not only by the exit gas temperature, but also by the height of the stack through which the gases exit. Very often an increase in height of a planned incinerator stack can eliminate the possibility of future air pollution problems.

## Burial of Wastes

Incineration is not the only method of disposing of tars and catalyst complexes. It is perfectly acceptable practice to bury these materials in many instances. Burial is usually less expensive than incineration at the outset but becomes more expensive as more property is required. Several methods of burying waste products are being employed in the chemical industry at the present time. It is frequently possible to use the method of sanitary land

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fill with the tarry or catalyst materials being mixed with earth and deposited as fill in an excavated dumpsite. This procedure may be feasible if inexpensive waste land is available. Whether or not the land so used can be subsequently used for construction sites, etc., depends on the ultimate fate of the tarry materials deposited. The possibility of the tarry materials leaching into ground water reservoirs must, of course, be explored.

A more common method of disposing of tars by burial involves placing them in waste drums and burying the drums. This procedure also presents the problem of possible ground water contamination after the drums have deteriorated and also the possibility of fire or explosion in the dumpsite.

Another common method of disposal consists of burying or dumping this type of material in public dumps which is usually just another method of incineration since most public dumps ignite flammable material to reduce volume.

In some areas it is possible to dispose of tarry materials in deep pits. This has been done in strip mine pits where the tarry materials can be placed in drums and buried under 40 to 60 ft of strip mine overburden. But this also constitutes a potential threat of contamination of the ground water supplies. Therefore, thorough knowledge of the subsurface formations is desirable before using this method.

### Direct Discharge to Streams

In discharging tarry materials or catalyst complex materials by sewers to streams, great care must be taken to prevent contamination of the receiving stream. This creates a specific problem when consideration is given to including tars in wastes to streams. It is possible to dilute some of these materials, which are either water soluble or partially water soluble, to a point where they constitute no hazard in the receiving stream. However, when large quantities of non-miscible organic materials are to be disposed of, direct discharge to streams is definitely not the process of choice since to do so simply transfers the problem from the process site to the receiving stream.

In recent years the problem of disposing of tarry materials has become quite critical due to the expansion of the petrochemical and petroleum industry. Many new uses of old processes have been developed in the effort to cope with this problem.

Deep sea disposal of heavy organic tars is one of these procedures. At the present time there are companies using this procedure along the Atlantic Coast, the Gulf Coast, and the Pacific Coast. While highly toxic materials, including radioactive substances, are usually placed in containers and disposed of from ships far out at sea, it would be very expensive to dispose of large quantities of tarry materials in this manner. The Shell Chemical Company has pioneered in the establishment of a modified deep sea disposal system for organic tars. The wastes are hauled in a specially constructed barge some distance from shore and discharged directly into the Gulf of Mexico (6) (7).

Establishment of a deep sea disposal system requires that a thorough study of the marine conditions in the area of disposal be made. The effect of the material on the indigenous marine life must also be determined. Authorities in the area must have assurance that the material will neither greatly affect the marine fishing industry nor return to shore to deposit along public waterways, particularly recreational beaches. Very extensive data from recognized biological and oceanographic consultants are required by all states before approval is given to plans for disposal of wastes by this method. The National Lead Company, for example, now discharges sulfuric acid and ferrous sulfide off the coast of New Jersey. Extensive work was done by the Woods-Hole Oceanographic Institute for the

WHP 42

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National Lead Company before this disposal method was permitted. Very detailed studies were also made for the Shell Chemical Company by the Texas A & M College before their disposal procedure was permitted by the State of Texas. Since the Texas operation is beyond the international boundary, it might be supposed that the State of Texas would have no authority in this case but this is not true. The barge that disposes of the material both originates and t[illegible]ates its trips in Texas ports and, [illegible] the material might adversely affe[illegible] [illegible]as marine fishing interests or the [illegible] of recreational beaches within the boundary of the state, the state can and does exercise rights of control.

## Reclamation Processes

There are many other ways of disposing of tarry materials in addition to those described above, particularly in the case of materials which contain recoverable products. Many companies engage chemical recovery firms to recover usable products from their catalyst complexes and tars. These firms can handle the tars from many different operations and usually incinerate the flammable materials that are not recoverable. In many cases, tars contain particular materials that make them salable as scrap material. For example, at one time Monsanto Chemical Company sold tar containing recoverable quantities of copper to the American Smelting and Refining Company. This company recovered the copper from these tarry materials at a smelter. Tars containing nickel and many other usable metals can also be sold to chemical scrap dealers for recovery of these valuable metals. These particular methods of disposal usually do not, in themselves, produce a profit for the company producing the tars. Often, however, they eliminate the necessity for the purchase of expensive incineration equipment and the possible creation of air pollution problems.

## Conclusions

The problem of disposing of tars, spent catalysts, and complexes is a broad one. While many methods of disposal are currently used in the petrochemical industry, no panacea for disposal of these troublesome substances appears to be at hand. As is so often the case in the industrial wastes field, the disposal of tars from the petrochemical industry requires painstaking study of the composition of the particular material to be disposed of and thorough engineering work on the design of disposal equipment which will be capable of accomplishing the job successfully.

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WHP 42

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# TRANSCRIPT OF PROCEEDINGS

IN THE SUPERIOR COURT

OF THE STATE OF DELAWARE

IN AND FOR NEW CASTLE COUNTY

| | | |
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| MONSANTO COMPANY, | | |
| Plaintiff, | : | Civil Action Number |
| v. | : | 88C-JA-118-1-CV |
| AETNA CASUALTY & SURETY COMPANY, et al., | : | NON-ARBITRATION CASE |
| Defendants. | : | |

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Washington, D. C.

Thursday, February 4, 1993

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