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(54) AMMONIUM NITRATE NEUTRALIZER

(71) We, MISSISSIPPI CHEMICAL CORPORATION, a corporation organized and existing under the laws of the State of Mississippi, U.S.A., of Yazoo City, Mississippi, U.S.A., do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to a commercially feasible process and apparatus for neutralizing nitric acid with ammonia whereby the "smog" formation usually associated with such neutralization, is effectively avoided.

Ammonium nitrate is usually prepared by neutralizing nitric acid with ammonia. In the usual commercial process, this is accomplished by continuously feeding an aqueous solution of nitric acid and a stream of ammonia through opposing spargers into a neutralization vessel containing a solution of the reactants and products. As the neutralization proceeds, product concentration is increased by boiling off the water by the exothermal heat of reaction, which is then vented into the atmosphere. One difficulty with this commercial technique, however, is that not only does the exothermal heat boil off water, but it also causes the formation of a quantity of an unrecoverable vapor-like microparticulate ammonium nitrate fume. The fume, with unreacted ammonia and nitric acid, is eventually vented into the atmosphere in the form of an objectionable "smog". This smog can cause environmental damage by poisoning the surrounding air and possibly destroying nearby animal and plant life, and by creating an undesirable visual effect, making it objectionable to locate a neutralization plant near inhabited areas.

The reason for the "smog" formation seems to be that the exothermal heat generated from the nitric acid neutralization reaction, vaporizes a quantity of the acid, which then combines with the ammonia vapor to form ammonium nitrate microparticles, which are generally unrecoverable. This theory seems to be supported by the observation that the

extent of ammonium nitrate smog is approximately proportional to the quantity of nitric acid vaporized.

In view of this theory, it was first contemplated to eliminate the smog formation by cooling the neutralization reactor contents, so as to reduce the quantity of nitric acid vaporized. While that expedient was successful in reducing smog formation, it has not proven to be entirely satisfactory from an economic point of view, since it meant the loss of the exothermal heat needed to evaporate the water contained in the weak reactant and product solution.

The use of more dilute nitric acid, likewise reduced smog formation, but it meant that a substantially greater quantity of water had to be evaporated in order to effectively concentrate the product ammonium nitrate to a usable strength.

Unable to solve the problem economically by eliminating the source of that problem, the art then relied upon various scrubbers, filters and condensers, designed to reduce the quantity of ammonium nitrate and the quantity of unreacted ammonia and nitric acid escaping into the atmosphere. Not only were the costs of such auxiliary equipment high and the costs of maintaining the equipment high, but those prior art techniques were unable to eliminate a sufficient amount of the ammonium nitrate microparticles to reduce the smog to acceptably low levels, unless total gas condensation was effected, which would result in undesirable quantities of waste material being discharged into the waste water stream.

Numerous attempts were made to redesign the basic nitric acid neutralizer equipment, so that volatilization of nitric acid could be reduced without deleteriously affecting the product concentration. None of these prior art techniques, however, were entirely successful in providing a commercially acceptable design which would sufficiently reduce the level of smog formation.

Attempts at such equipment redesign date

as far back as 1939 with the *Fausser* neutralizer, U.S. Patent Application Serial Number 306,071, published under the Alien Property Control Act. In that design, the reactor comprised a central reaction zone located concentrically within a vessel and in fluid flow communicated therewith. The nitric acid was introduced directly into the central zone through a sparger located approximately adjacent to the entrance of the central zone. Ammonia was introduced into the vessel at some distance from the inlet to the central zone, and neutralization was effected as the ammonia entered the central zone from the outer vessel. Although that design reduced the extent of acid volatilization, it was insufficient to provide an acceptably low smog level. Moreover, that system required a vertical shaft mixer to circulate the aqueous solution between the central zone and the outer vessel, which was inefficient and increased the product cost.

Later systems were developed, such as those disclosed in Japanese Patent No. 15,292/68 and Norwegian Patent No. 15,958, which also used a *Fausser* type central zone, but they, likewise, fed the nitric acid in high concentrations directly into the core where neutralization occurred. All of these prior art systems were inadequate, since a high concentration of nitric acid was reacted with ammonia and hence the heat of reaction was sufficiently high to cause high degrees of acid volatilization.

A need existed, therefore, for a means of neutralizing nitric acid with ammonia in an economically efficient manner, with a minimum formation of the deleterious ammonium nitrate smog.

According to one aspect of the invention there is provided a thermal siphon-pressure pump neutralizer for neutralizing nitric acid with ammonia, which comprises:

a reaction vessel suitable for containing an aqueous reaction medium and having a vapour outlet and a product outlet, said product outlet defining the level of said aqueous reaction medium in said vessel when the apparatus is in normal use,

an elongated fluid impervious hollow cylindrical member positioned within said vessel and defining a second reaction zone within said member and a first reaction zone within said vessel outside said member, said member having an inlet positioned below the level of said product outlet and an outlet positioned above the level of said product outlet,

ammonia inlet means for introducing ammonia and/or ammonium hydroxide into said vessel in proximity to said member inlet and for directing said ammonia and/or ammonium hydroxide towards said inlet, and

nitric acid inlet means for introducing nitric acid into said first reaction zone, said means being positioned between said product

outlet of said vessel and said inlet of said cylindrical member. 65

According to another aspect of the invention there is provided a method of neutralizing nitric acid with ammonia in a thermal siphon-pressure pump neutralizer as defined herein, which comprises: 70

maintaining a body of aqueous liquid within said reaction vessel to a level defined by said product outlet of said vessel,

introducing nitric acid into said first reaction zone via said nitric acid inlet means, introducing ammonia and/or ammonium hydroxide into said vessel via said ammonia inlet means, and 75

removing excess ammonium nitrate-containing liquid from said product outlet of said vessel, 80

and wherein the method further comprises introducing sufficient amounts of said nitric acid and said ammonia and/or ammonium hydroxide to maintain a thermal siphon-pressure pump effect within said apparatus and introducing said nitric acid in concentrations and amounts insufficiently high to raise the concentration of nitric acid entering said second reaction zone to 15% or more. 85 90

It is an advantage of this invention, at least in the preferred embodiments, that it can provide an economically feasible nitric acid-ammonia neutralization method which minimizes the extent of smog formation to acceptably low levels. 95

Another advantage of this invention, at least in the preferred embodiments, is that it can provide an apparatus for accomplishing said method, which is simple in design, and which does not require auxiliary scrubbing equipment to reduce the smog formation to acceptably low levels. 100

In the invention nitric acid is diluted in a first reaction zone, which is contiguous with, and in fluid communication with, a second reaction zone; ammonia is fed into proximity to the inlet of the second reaction zone, and the predominant portion of the neutralization is conducted within the second reaction zone. Circulation between the first and second zones is effected by the thermal siphon and pressure lift caused by the exothermal heat released during neutralization and by the density difference between the product solution as compared with the reactant solution. By proper alignment of the ammonia inlet with the inlet to the second reaction zone, a venturi-type constriction can be created to enhance mixing and to enhance the rate of recirculation. In one embodiment, means are provided within the second reaction zone and/or within the ammonia inlet means, to enable recovery of at least a major portion of any ammonia gas entrapped within the steam generated by the exothermal heat of neutralization. In another embodiment a water or 105 110 115 120 125

weak ammonium nitrate inlet is provided for enabling a quantity of the water vaporized from the system to be continuously replenished and advantageously means are employed within the second reaction zone for enhancing contact between the liquid medium and ammonia gas entrapped within the steam generated by said heat of neutralization of said ammonia-nitric acid reaction.

Figure 1 is a schematic view of the neutralizer of the present invention;

Figure 2 is a schematic view of the neutralizer of the present invention with its auxiliary inlet and outlet systems;

Figure 3 is a bottom section of the neutralizer of the present invention showing the use of a funnel insert emplaced within the second reaction zone to enable recovery of ammonia gas entrapped within the generated steam. This figure also shows the use of an optional propeller vane system emplaced within the second reaction zone to effect swirling of the rising liquid to enable increased gas-liquid contact; and

Figure 4 is a bottom section of the neutralizer of the present invention, showing a conical skirt emplaced around the ammonia inlet conduit to reduce eddy current formation and to smooth the flow of liquid into the second reaction zone.

In the present invention, nitric acid in a sufficiently low concentration is reacted with ammonia such that vaporization of the nitric acid is inhibited. Since the nitric acid is maintained essentially in the liquid state, the ammonium nitrate being formed will likewise be in a liquid state, and not in the undesirable vaporous or microparticulate state.

The lower the acid concentration, the lower will be the quantity of acid vaporized. Some "smog" reduction occurs when the nitric acid concentration at the time of the reaction is as low as 15%, but really substantial smog reduction begins to occur when the nitric acid concentration is less than 2%. Exceptionally superior results occur at acid concentrations of between 0.5 and 1.0%, and most preferably, for best results, the acid concentration should be 0.5% or less.

Since the concentration of product ammonium nitrate in prior art systems was generally dependent upon acid concentration, it is quite surprising that the present system can achieve an acceptably high product concentration without sacrifice to plant economy. Moreover, the present system can reduce the formation of ammonium nitrate smog down to levels approximating only 0.1% by weight or less of the stack gas effluent, or as little as a few parts per million of air.

Referring to Figure 1, which is a schematic view of the reactor system 1 of the present invention, nitric acid enters inlet line 3 into a first reaction zone 5 of a reactor vessel 2 through sparger 7. A second reaction zone

11 is defined by an elongated fluid impervious cylindrical member 10 and is provided with an inlet 9 and an outlet 12. The cylindrical member 10 is positioned within the reaction vessel 2 such that when the vessel 2 contains an aqueous medium, the inlet 9 of the member 10 will be beneath the level of the medium in the first reaction zone and the outlet 12 of said cylindrical member will be above the level of the medium.

The nitric acid sparger 7 shown in Figure 1 consists of four perpendicularly crossing pipes, each containing a plurality of small diameter nitric acid ports. The precise configuration of the nitric acid inlet is not critical, however, and it may take a variety of forms suitable for feeding nitric acid at a controlled rate, such as a simple pipe inlet, or a ringed sparger.

The acid entering the first reaction zone 5 may be highly dilute or may be in highly concentrated form. For best efficiency, the acid should be approximately in commercial concentrations of between 55 to 60%, which permits the production of ammonium nitrate in concentrations of 77 to 84%, which is a commercially usable form. Lower or higher acid concentrations, however, may be used. The only limitation is that the nitric acid inlet must be situated at a sufficient distance from the inlet 9 to the second reaction zone 11 such that when the nitric acid enters the second reaction zone 11, it will have the desired concentration, i.e. less than 15%.

The concentration of the nitric acid entering the second zone 11 is a function of the recirculation rate between the second zone 11 and the first zone 5, the distance between the nitric acid inlet sparger 7 and the second reaction zone inlet 9, and the concentration of the inlet nitric acid entering the system through sparger 7. Some additional control of the second zone inlet concentration can be provided by incorporating baffles between the sparger 7 and the inlet 9, or by otherwise varying the degree of turbulence in the first reaction zone.

Ammonia is introduced into the system 1 through an ammonia inlet sparger 15. Either ammonia vapor or ammonium hydroxide liquid or vapor may be used as the ammonia source. The incoming gas or vapor may contain amounts of carbonate, carbon dioxide or inert gas, depending upon the source of supply or depending upon the concentration desired. One good supply source for ammonia is urea plant off-gas.

The ammonia enters into proximity to the second reaction zone inlet 9. It is preferable to feed the ammonia into the first reaction zone just adjacent to the inlet 9 and in the direction of that inlet. By proper spacing between the ammonia inlet 15 and the second reaction zone inlet 9, a constriction X can be created which will provide a venturi-like

effect for the solution entering the second reaction zone 11, which serves to increase the rate of circulation and the degree of turbulence, and hence enhances the mixing of the ammonia and nitric acid reactants. The actual spacing X will depend upon the size of the particular unit and the ratio between the diameter of the walls of the cylindrical member 10 to the diameter of the walls of the ammonia inlet 17. The greater the value X, the smaller will be the venturi effect and the smaller will be the pressure drop. Of course, if the diameter ratio approaches 1:1, and the distance X is quite small, the rate of recirculation can be made to approach zero.

The respective diameters of the inner diameter of the reaction vessel 2, defining the first reaction zone 5, and the outer diameter of the walls of the cylindrical member 10, will also have an effect on the degree of pressure drop in the system. The ratio of vessel 2 diameter to cylindrical member 10 diameter should be small enough to create sufficient turbulence in the recirculating solution, but large enough that the system pressure drop is not excessive. Suitable ratios have been found between the ranges of 1.5:1 to 4:1.

Although the second reaction zone 11 is shown in the Figures as being defined by a single cylindrical member 10, a bundle consisting of a plurality of cylindrical members, i.e., 3 or 4 such members, may be used to get different pressure effects. The various members 10 may be separated by a finite distance within vessel 2 or may be touching. When more than one member 10 is used, a single ammonia inlet 17 may be used, or multiple inlets may be used, one for each member, or one for several members.

The opening of the ammonia inlet 15 may be simply an open pipe, but preferably constructed so that the ammonia is evenly dispersed into the media. Good results have been obtained with a Number 4 mesh screen 19 having a 16.6 per cent open area.

It is possible to introduce the ammonia directly into the second reaction zone 11, but this would decrease the extent of mixing and turbulence and would decrease the rate of recirculation and could result in increased stack losses. Accordingly, introduction of the ammonia directly into the second zone 11 is acceptable, but is somewhat less efficient.

Since the ammonia, in the preferred design, is introduced just outside of the second reaction zone 11, neutralization will actually begin in the first zone 5, in advance of inlet 9. The predominant amount of neutralization, however, will occur within the second reaction zone 11.

The exothermal heat released by the neutralization reaction will cause the water to boil, but because of the low acid concentra-

tion, vaporization of the nitric acid will be restrained.

Recirculation between the first and second reaction zones 5 and 11, is effected by the turbulent rise of gases through the aqueous medium within the second reaction zone 11, which acts as an air-lift, or pressure pump. Recirculation is also effected by the reduced density of the solution in the second reaction zone 11 as compared with that in the first reaction zone 5, which imparts a driving pressure head which will cause the solution to surge upwardly through outlet 12, which is disposed vertically with respect to the reaction vessel 2, so that it fountains above the level of the medium in the first and second reaction zones.

A curvilinear deflector 21 may be positioned above the outlet 12 so as to deflect the solution downwardly, back into the first reaction zone 5. Gases, possibly containing entrained liquid, will circumvent the deflector and will proceed out of the system through the vapor outlet 23. The deflector 21 should be so positioned that it does not adversely affect the circulation of the solution. If it is located too high above the second reaction zone outlet 12, it will lose its effectiveness of diverting the liquid back to the first reaction zone.

When properly positioned, liquid will run off the deflector in a continuous "sheet" or film, so that the gases circumventing the deflector will be forced to pass through the liquid curtain which will then serve to remove most of the entrained liquids.

The use of a deflector, however, is optional, and the same effect could be obtained by extending the column between the liquid medium and the vapor outlet high enough so that the entrained liquid will be permitted to agglomerate and fall back to the first reaction system.

The gases from the reaction may be vented directly into the atmosphere, since the quantity of ammonium nitrate, ammonia, and nitric acid entrained or vaporized, is quite low, and within ecologically acceptable limits.

If desired, however, the gases may be passed through a mesh mist remover 25 situated within the vapour outlet so as to recover still further any entrained liquid particles. Any liquid entrapped in the mesh is permitted to drop back to the first reaction zone 5.

It may be desirable to increase the diameter of the reaction vessel above the deflector, as compared with the diameter of the vessel below the deflector, so as to reduce the space velocity of the vapors sufficiently that a large amount of the liquid entrained in the vapors will be able to drop out of the vapor and back into the first reaction zone.

Any of a variety of conventional separators or scrubbers may be added to the system to

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5 further remove vapor phase or liquid phase
nitric acid or ammonia. It is extremely un-
economical and difficult to remove any vapor
state or microparticulate ammonium nitrate
in the exhaust fume, and, accordingly, the
neutralization technique itself must be capable
of preventing the formation of such vapor
state or microparticulate material within
acceptable limits.

0 When the unit is operating with reasonable
efficiency, the ammonium nitrate losses in the
exhaust gas can be as low as 0.1% or lower.

15 An overflow ammonium nitrate outlet 27
is provided at, or near, the surface level of
the solution in the first reaction zone 5,
for removing the product from the system.
The particular location for the product outlet
27 is not critical, but it should be above the
level of the nitric acid inlet 3.

20 The nitric acid entering the sparger 7
is diluted by being admixed with the am-
monium nitrate solution recirculated from
the second reaction zone 11. As the nitric
acid enters the second reaction zone 11, it
25 is neutralized and removed by the ammonia.
The acid concentration in the aqueous media,
therefore, is considerably reduced as it leaves
the second zone, and is returned to the first
zone. The greater the rate of recirculation,
30 the lower will be the acid concentration of
the acid entering the second zone. The con-
centration should, of course, be less than 15%.
Of course, the rate of recirculation is depen-
dent upon many variables, including concen-
35 tration of the inlet nitric acid, separation
distance between the nitric acid and the am-
monia spargers and size of equipment.

Good results have been obtained where the
first reaction zone 5 contained 125 pounds
40 of recycle per pound of commercially pro-
duced nitric acid feed, which corresponded
to 0.05% nitric acid in the second zone out-
let.

45 Under automatic pH control, it is not
feasible to operate the neutralizer under over-
all neutral conditions; there will always be
some free acid or ammonia in the product.
If the concentration of the ammonia or nitric
acid in the product is allowed to deviate too
50 far from neutral, however, stack losses can
become severe and seem to increase ex-
ponentially in acid concentration. It has also
been found that more ammonium nitrate stack
losses occur when the reaction is run overall
55 slightly acidic than when it is run overall
slightly basic.

60 If the reaction is conducted on the acidic
side, the vapor pressure of nitric acid over
the boiling nitric acid-water ammonium nitrate
solution will primarily be a function of the
nitric acid-to-water ratio in the solution. As
the ammonium nitrate concentration is in-
creased, while maintaining a constant acid con-
centration and a constant rate of recirculation,
65 nitric acid vaporization will be increased due

to the increased nitric acid/water ratio. The
practical result is that as the concentration
of the product is increased, the degree of
nitric acid vaporized will be increased. To
off-set this increased nitric acid loss, the rate
of recirculation must be increased to reduce
the nitric acid/water ratio and thereby to
reduce the acid vapor pressure, but there is a
practical limit as to the extent of recircu-
lation possible.

Of course, when the system is run with
excess ammonia, the neutralization reaction
will begin as soon as the acid enters the
first reaction zone, but the degree of neutral-
ization usually is insignificant as compared
with the degree of neutralization within the
second reaction zone. This preneutralization,
of course, is beneficial in that it further
directly reduces the acid concentration and
causes increased turbulence and recirculation,
which still further reduces acid concentration.

In designing the equipment, the second re-
action zone should be properly sized such that
the residence time of the reaction components
in that zone will be sufficient to permit the
reaction to go to substantial completion be-
fore the solution surges out toward the de-
flector.

The surrounding solution level should be
adjusted to a proper height to provide an
adequate rate of recirculation. In general, the
lower the liquid level, the slower will be
the recirculation rate, since the smaller will
be the pressure head. The longer the cylin-
drical member 10, defined in the second re-
action zone 11, the greater will be the energy
losses within the zone, and the slower will
be the rate of recirculation.

Although the neutralizer of this invention
has been referred to as a thermal siphon
neutralizer, the pumping action which provides
for the recirculation between the first and
second reaction zones, is actually a combina-
tion of two mechanisms: (1) the thermal
siphon and (2) the air-lift pump. As am-
monia enters the neutralizer, liquid flow is
induced in the manner of an air-lift pump.
The reaction between ammonia and the acid
generates steam in the second reaction zone
which sustains the air-lift pump. The tem-
perature and density differences of the
material inside and outside the second re-
action zone induces flow in the manner of
a thermal siphon reboiler.

It has been found that some of the inlet
ammonia gas will be entrapped by the gener-
ated steam, and unless otherwise separated,
will be carried into the exhaust gas stream.
The quantity of ammonia entering the ex-
haust stream is usually small and not neces-
sarily objectionable. However, it has been
found that a major portion of the ammonia
in the exhaust gas can be eliminated by fixedly
emplacing a funnel 40 between the walls 10
of the second reaction zone 11, as shown in

Figure 3. The funnel 40 is preferably of a generally frusto-conical configuration, having a cylindrical portion 44 at the apex of a frusto-conical portion 42. The degree of slope of the conical portion 42 is not particularly critical, although it has been found that some degree of slope is necessary to reduce pressure drops due to entrance losses. Moreover, the sloping section will enhance the rate of circulation within the second reaction zone 11, so that the circulation rate will be intermediate between the larger rate which would occur without the presence of a funnel 40, and the somewhat reduced rate which would prevail if the insert were only cylindrical in configuration. It is theorized that the effect of the funnel is to enhance contact of the ammonia gas with the liquid medium so that more of the ammonia will be dissolved into the liquid and hence be made available for reaction with the nitric acid. According to this theory, the major portion of the ammonia will react with the nitric acid at the point of highest ammonia concentration, i.e., in proximity to the entrance of the second reaction zone 11. The heat from the exothermic reaction will generate steam which will carry some of the ammonia up through the funnel 40. The sudden expansion of the gases at the exit end of the funnel will slow the rate of gas flow and/or will cause increased turbulence thereby increasing the distribution and dissolution of the ammonia gas into the liquid.

When a funnel type configuration is used, it is desirable that the sloped portions of the conical section 42 be extended beyond the walls 10 of the second reaction zone 11 in order to reduce bypassing of the gas around the reaction zone inlet 9. The angle of the slope of the extended section may be the same or different than the angle of slope of the conical walls.

In one embodiment of this invention, it has been found that good results are obtainable when the inside diameter of the second reaction zone 1 is 24 inches, the diameter of the conical portion 42 of the funnel 40 is 18 inches, the slope of the conical section is 40 degrees, and the walls of the conical section extend about 2 inches beyond the walls 10 of the second reaction zone 11.

These particular sizes, however, are merely illustrative and are not intended to be construed as limiting of the invention. Another method of recovering entrapped ammonia from the generated steam, is to insert suitably spaced spiral type baffles (not shown) within the second reaction zone 11, or within the cylindrical portion 44 of the funnel 40, which will cause the rising liquid-gas mixture to swirl. The swirling motion imparted to the mixture by the baffles will increase turbulence and hence will enhance the gas-liquid contact. Instead of baffles, propeller type vanes 50 mounted on supports 51 may be emplaced within the second reaction zone 11 as shown in Figure 3, or within the cylindrical portion 44 of the funnel 40 (not shown) to accomplish the same type of swirling motion.

In another modification of this invention, means can be inserted into the vicinity of the ammonia inlet 17 which will reduce the extent of eddy current formation at the walls of the inlet 17, thereby smoothing the flow of circulating liquid into the second reaction zone 11. As shown in Figure 4, this means may take the form of a conically shaped skirt 53 surrounding the inlet pipe 17.

Having now generally described the invention, a further understanding can be obtained by reference to certain specific Examples which are provided herein for purposes of illustration only and are not intended to be limiting in any manner.

The following Examples will be discussed with respect to the schematic of Figure 2. Nitric acid of about 55% concentration enters the system 1 through line 31. Gaseous ammonia enters through line 32. A secondary feedstream 37 feeds material recovered from the separator scrubber (not shown). This feedstream is, of course, optional to the process.

Product ammonium nitrate is recovered through line 35 and steam and gaseous by-products are exhausted through line 36. A water stream 34 may be fed into the exhaust stream to desuperheat that stream when using the optional scrubber. Stream 33 is exhausted into the atmosphere.

The results of several runs under acidic and basic conditions are shown in the following Tables.

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TABLE I

Process Flows With Product Slightly Acidic (0.1% by Weight)

Stream		31	32	33	34	35	36	37
Components								
NH ₃	lb/hr.	—	4,852	10	—	—	10	—
HNO ₃	lb/hr.	17,678	—	—	—	34	—	8
H ₂ O	lb/hr.	13,537	2,546	9,598	—	8,734	9,598	2,249
CO ₂	lb/hr.	—	783	783	—	—	783	—
NH ₄ NO ₃	lb/hr.	—	—	10	—	25,518	10	3,117
Air	lb/hr.	—	—	—	—	—	—	—
Total (Norm.)		31,215	8,181	10,401	—	34,286	10,401	5,374
Total (Max.)		49,706	23,750	18,700	401	51,000	18,700	6,600
Temp. (°F.)		105	140	258	80	258	258	200
Pressure Psig		—	—	0.2	—	—	0.2	—
Conc. %		56.63	—	—	—	75	—	58
Sp. Gr.		1.326	—	—	1.0	1.295	—	1.23
M.P. °F.		—	—	—	32	—	—	—
B.P. °F.		—	—	—	212	258	—	237
Flow G.P.M.*		47	—	—	—	53	—	7.3
Flow A.C.F.M.**		—	3,345	4,750	—	—	4,750	—

* G.P.M. means gallons per minute

** A.C.F.M. means actual cubic feet per minute

TABLE II

Process Flows With Product Slightly Ammoniacal

Stream		31	32	33	34	35	36	37
Components								
NH ₃	lb/hr.	—	4,852	100	—	0.3	100	—
HNO ₃	lb/hr.	17,678	—	—	—	—	—	86
H ₂ O	lb/hr.	13,537	2,546	9,598	—	8,656	9,598	2,171
CO ₂	lb/hr.	—	783	783	—	—	783	—
NH ₄ NO ₃	lb/hr.	—	—	3	—	25,656	3	3,117
Air	lb/hr.	—	—	—	—	—	—	—
Total	(Norm.)	31,215	8,181	10,484	—	34,312	10,484	5,374
Total	(Max.)	49,706	23,750	18,700	401	51,000	18,700	6,600
Temp.	°F.	105	140	258	80	258	258	200
Pressure	Psig	—	—	0.2	—	—	0.2	—
Conc.	%	56.63	—	—	—	75	—	58
Sp. Gr.		1.326	—	—	1.0	1.295	—	1.23
M.P.	°P.	—	—	—	32	—	—	—
B.P.	°F.	—	—	—	212	258	—	237
Flow	G.P.M.	47	—	—	—	53.4	—	7.3
Flow	A.C.F.M.	—	3,345	4,800	—	—	4,800	—

The quantity of ammonium nitrate, ammonia and nitric acid emitted into the atmosphere by the apparatus and process of the present invention was compared with that emitted from the atmosphere by conventional neutralization equipment under similar reaction conditions, and the following data compiled.

TABLE IV

Pilot Unit Stack Loss Data for Various Neutralization Schemes

	Free HNO ₃ , Free NH ₃ in Product wt. %	HNO ₃ Conc. Entering Reaction Zone wt. %	Stack Gas Composition Weight %
			%HNO ₃ , %NH ₃ , %NH ₄ NO ₃
Case 1. Conventional neutralizer, product slightly acidic	0.1	0.0	not measured 0.0 0.26 0.31
Case 2. Conventional neutralizer, product slightly ammoniacal	0.0	0.037	not measured 0.0 0.50 0.20
Case 3. Thermal Syphon neutralizer, acid introduced inside central pipe, product slightly acid	0.08	0.0	not measured 0.0 0.80 0.22
Case 4. Thermal Syphon neutralizer, acid introduced near entrance to central pipe, product slightly acidic	0.08	0.0	not measured 0.0 0.21 0.24
Case 5. Thermal Syphon neutralizer, optimized design, product slightly acidic	0.08	0.0	0.36 0.0 0.10 0.097
Case 6. Thermal Syphon neutralizer, optimized design, product slightly ammoniacal	0.0	0.003	0.21 0.0 0.67 0.025

N.B. In Table IV Cases 1 and 2 are prior art systems and Cases 3 to 6 illustrate embodiments of the present invention.

WHAT WE CLAIM IS:—

1. A thermal siphon-pressure pump neutralizer for neutralizing nitric acid with ammonia, which comprises:
 - 5 a reaction vessel suitable for containing an aqueous reaction medium and having a vapour outlet and a product outlet, said product outlet defining the level of said aqueous reaction medium in said vessel when the apparatus is in normal use.
 - 10 an elongated fluid impervious hollow cylindrical member positioned within said vessel and defining a second reaction zone within said member and a first reaction zone within said vessel outside said member, said member having an inlet positioned below the level of said product outlet and an outlet positioned above the level of said product outlet,
 - 15 ammonia inlet means for introducing ammonia and/or ammonium hydroxide into said vessel in proximity to said member inlet and for directing said ammonia and/or ammonium hydroxide towards said inlet, and
 - 20 nitric acid inlet means for introducing nitric acid into said first reaction zone, said means being positioned between said product outlet of said vessel and said inlet of said cylindrical member.
 - 25
 - 30 2. A neutralizer according to claim 1, wherein the walls defining said ammonia inlet means form a venturi-type constriction with the walls defining said cylindrical member inlet so as to increase the rate of circulation between said first and second reaction zones.
 - 35
 - 40 3. A neutralizer according to claim 1 or claim 2 which further comprises a water or a weak ammonium nitrate liquor inlet for enabling a quantity of the water vaporized from the system to be continuously replenished.
 - 45 4. A neutralizer according to any preceding claim wherein the nitric acid inlet means is positioned closer to said product outlet than to said cylindrical member inlet.
 - 50 5. A neutralizer according to any preceding claim wherein said cylindrical member outlet is disposed vertically with respect to the reaction vessel so that the thermal and pressure heads created by the exothermal ammonia-nitric acid reaction will cause the aqueous solution within said second reaction zone to fountain upwardly through said member outlet, above the surface of the aqueous solution contained in said first reaction zone.
 - 55 6. A neutralizer according to claim 5, wherein a curvilinear deflector is supported above the cylindrical member outlet such that the solution fountaining from said member will be deflected downwardly back to said first reaction zone, and vapors emitted from said member outlet will circumvent said deflector and pass through the vapor outlet.
 - 60 7. A neutralizer according to claim 6, which further contains a mesh mist remover situated within the vapor outlet which is capable of separating entrained liquid from the vapor stream passing through the outlet and permitting said liquid to drop back into said first reaction zone.
 - 65 8. A neutralizer according to any preceding claim, wherein a plurality of elongated fluid impervious cylindrical members are positioned within said vessel so as to define said second reaction zone.
 - 70 9. A neutralizer according to any preceding claim further containing means emplaced within said second reaction zone for enhancing contact between the liquid medium and ammonia gas entrapped within the steam generated by said heat of neutralization of said ammonia-nitric acid reaction.
 - 75 10. A neutralizer according to claim 9, wherein said means for enhancing the contact is a funnel which is fixedly emplaced within said second reaction zone.
 - 80 11. A neutralizer according to claim 10, wherein said funnel has an inlet portion which is substantially frusto-conical in configuration, and has an outlet portion which is substantially cylindrical in configuration at the apex of said conical portion.
 - 85 12. A neutralizer according to claim 11, wherein the walls of said conical portion extend beyond said second reaction zone.
 - 90 13. A neutralizer according to claim 9, wherein said means for enhancing the contact are propeller vanes rotatably mounted within said second reaction zone such that the rotation of said vanes imparts a swirling and mixing motion to the gas and liquid contents within said second reaction zone.
 - 95 14. A neutralizer according to any preceding claim further containing means emplaced within said first reaction zone for reducing eddy current formation which would disrupt the flow of liquid into the second reaction zone.
 - 100 15. A neutralizer substantially as described herein with reference to and as illustrated in the accompanying drawings.
 - 105 16. A method of neutralizing nitric acid with ammonia in a thermal siphon-pressure pump neutralizer according to claim 1, which comprises:
 - 110 maintaining a body of aqueous liquid within said reaction vessel to a level defined by said product outlet of said vessel,
 - 115 introducing nitric acid into said first reaction zone via said nitric acid inlet means,
 - 120 introducing ammonia and/or ammonium hydroxide into said vessel via said ammonia inlet means, and
 - 125 removing excess ammonium nitrate-containing liquid from said product outlet of said vessel,
 - 130 and wherein the method further comprises introducing sufficient amounts of said nitric acid and said ammonia and/or ammonium hydroxide to maintain a thermal siphon-pressure pump effect within said apparatus and



introducing said nitric acid in concentrations and amounts insufficiently high to raise the concentration of nitric acid entering said second reaction zone to 15% or more.

- 5 17. A method as claimed in claim 16 substantially as described herein with reference to the accompanying drawings.

R. G. C. JENKINS & CO.,
Chartered Patent Agents,
Chancery House,
53/64 Chancery Lane,
London, WC2A 1QU.
Agents for the Applicants.

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