

The LUREC® process—key to economic smelter acid plant operation

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Synopsis

Over many decades, numerous feasibility studies have demonstrated, that the production of sulphuric acid remains the most viable option of sulphur recovery from smelter off gas and abatement of SO_2 emissions to the atmosphere. This is particularly more pronounced as smaller but more concentrated off-gas flows are to be treated from smelters, and enhanced sulphuric acid processes become available.

Off-gas handling systems represent a significant capital and operating cost burden to the metallurgical operation. Modern pyrometallurgical smelter processes for sulphide ores based on the use of oxygen-enriched air, produce relatively small off-gas flows with high SO_2 concentrations in the smelter gas of 30–60%-vol. of SO_2. This is a prerequisite for substantial cost reductions in the smelter off-gas handling and treatment system.

As an alternative to sulphuric acid production, numerous scrubbing concepts with alkali or dual-alkali combinations as well as organic absorbents have been proposed. Also the reduction of the SO_2 to elemental sulphur has frequently been studied. Very few of those alternative processes have been built in industrial scale, but all were generally characterized by none-sustainable operation due to cost reasons, problems with issues related to chemicals used and by-products or poor availability.

Thus the traditional concept of converting the SO_2 to sulphuric acid is most common, although regarded as uneconomic, but is at least a proven, environmentally sustainable and reliable way of sulphur gas processing. With the high acid price levels these days, operating companies are even able to generate significant revenue with their otherwise 'fatal' acid.

Even though smelter gas is available at high SO_2 concentrations, in a conventional acid plant one would add large amounts of air to dilute the gas down to a suitable concentration of 12–13 (14)%-vol. SO_2. Dealing with higher SO_2 concentrations in smelter acid plants is not feasible, as the gas exit temperature of the catalytic oxidation step would exceed the allowable limit for the catalyst. Hence those conventional acid plants are characterized by the use of large equipment, i.e. high capital cost, high energy consumption and limited flexibility.

The newly developed LUREC® process can handle off-gas with significantly higher gas concentrations, even in excess of 25%-vol. SO_2. It is entirely based on well-proven equipment and unit operations. The first industrial application, operating with 16–18%-vol. SO_2 will be presented in this paper, along with the fundamentals of the process. It will be demonstrated that the process requires inherently lower capital cost, fewer operating costs, and offers better energy recovery and lower emis-sions as compared to conventional design.

While most existing smelter acid plants would have some built-in spare capacity, any significant increase of smelter capacity, say of 30%, can basically not be accommodated without installing an additional parallel new acid plant unit. An add-on LUREC® module can remove any such restriction, while simultaneously debottlenecking the existing acid plant. The paper also discusses the present status of the add-on technology for smelter acid plant expansions with respect to technical boundary conditions of different process alternatives as well as economic and environmental aspects.

Application of the LUREC® process to modern smelter acid plants operating with high SO_2 gas concentration, will lead to a substantial reduction of the specific size of the gas processing equipment and equivalent savings in capital and operating cost.

The LUREC® process is patented worldwide by OUTOTEC.

Converting of high grade SO_2 gas in a sulphuric acid plant—the challenge

At pyrometallurgical smelters based on sulphidic ore concentrates of copper, nickel, lead or zinc, it is traditional practice to process the SO_2 off-gas to sulphuric acid. The growing awareness of environmental protection has lead to increasingly stringent regulations, primarily aiming at the reduction of SO_2 emissions in defined off-gas streams, such as the tailgas of sulphuric acid plants, which are attached to smelters as a means of desulphurization system.

Consequently, one of the main objectives in process development for metallurgical sulphuric acid plants is to enable the operation with very high SO_2 concentrations, while simultaneously avoiding the generation of diluted gas from e.g. Pierce Smith converters. Higher grade SO_2 gas obviously leads to an effective reduction in specific equipment size of the gas handling system, i.e. gas purification and sulphuric acid plant. This, however, is ideally only possible with the application of completely continuous smelter and converter processes and without any batch operating steps, so that the acid plant can be designed for constant gas conditions.

The availability of such modern and continuous processes using technical oxygen or oxygen enriched air, which subsequently leads to high grade SO_2 smelter-and converter off-gas, opens up new possibilities for more economic solutions of environmental control.

The perspective of high-grade smelter off-gas thus represents an interesting challenge for the development of enhanced sulphuric acid process technology.

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Modern smelters offer 30–60%-vol. SO_2 off-gas, which is traditionally diluted with air to suitable 12–14%-vol. prior to being processed in the sulphuric acid plant. This dilution in turn enlarges the acid plant equipment by a factor of 2–3 and hence causes an equivalent increase of the related capital and operating cost. As the smelter gas concentration increases, so will the feed gas concentration to the acid plant's SO_2 converter. The catalytic oxidation of SO_2 to SO_3 is highly exothermic and the adiabatic reaction does lead to an increased gas temperature as the conversion proceeds. The higher the feed-gas concentration and the more favourable the O_2/SO_2 ratio, the higher is the resulting adiabatic reaction temperature. Figure 1 demonstrates the bed 1 exit gas temperature as a function of both said parameters.

Figure 2 presents the bed 1 exit temperature as a function of the SO_2 concentration and the use of conventional or caesium promoted catalyst at this bed at a fixed O_2/SO_2 ratio of 1.1. It demonstrates that the Cs-catalyst provides a bit more tolerance to high SO_2 concentration. The development of caesium promoted catalyst in recent years does enable the inlet temperature to the first bed to be decreased from a conventional 420°C by a further 20–40°C, while the activity is maintained. In this case, the first bed is equipped with Cs-promoted catalyst as a top layer, say 30–50%, followed by conventional catalyst for the rest of the bed. This offers an additional small 'push' towards higher feed-gas concentrations, though marginally only. This feature is quite often used for slight plant capacity increases. The difference to the gas exit temperature is compared to an all-conventional filling.

Another important aspect of SO_2 converter operation is the ratio of O_2 to SO_2. This is particularly important with respect to the final SO_2 stack emissions. Ever increasing restrictions on emissions have lead to the introduction of 5-bed converters early on, which offer the ability to further reduce said emissions. It also provides better conversion at lower O_2/SO_2 ratios, while at larger ratios, the effect becomes marginal. Figure 3 shows the achievable overall conversion vs. converter configuration (3+1 and 3+2) and O_2/SO_2 ratio for a feed-gas of 12.5%-vol. SO_2.

A diagram typically used in the sulphuric acid industry is shown in Figure 4, i.e. temperature vs. conversion of SO_2. It is obvious that gas temperatures in excess of 640 °C will occur once the converter feed-gas concentration shifts beyond say 13–14%-vol. SO_2.

Generally, the upper thermo-stability limit of commercially available vanadium-pentoxide catalysts is specified to be around 630°C. While in some cases operators are using 640° or even 650°C as upper limit, provided the mechanical integrity of the SO_2 converter does support it, the catalyst stability will suffer from such exposure and screening losses increase substantially. In many cases, a feed-gas of 16–18%-vol. SO_2 or even higher would be available with the right parameters to achieve the required overall conversion (i.e. O_2/SO_2 ratio) with a conventional 5-bed converter system. However, the resulting gas temperature of more than say 670 °C does not tolerate such design with respect to the application of conventional catalyst.

The exothermic oxidation of SO_2 to SO_3 generally leads to an excess of energy in the system, which has to be removed, preferably in form of high pressure steam useful for e.g. generation of electrical energy or other process use. A significant part of this energy is transferred with the gas as sensitive energy into the absorbers and eventually transferred to the environment via cooling water.

The operation of a sulphuric acid plant with high grade SO_2 gas would not only significantly reduce this part of wasted energy, but would rather make it available for suitable recovery. The situation is presented for a 1 000 mtpd acid plant in Figure 5 as a function of the SO_2 concentration of the feed-gas to the converter.

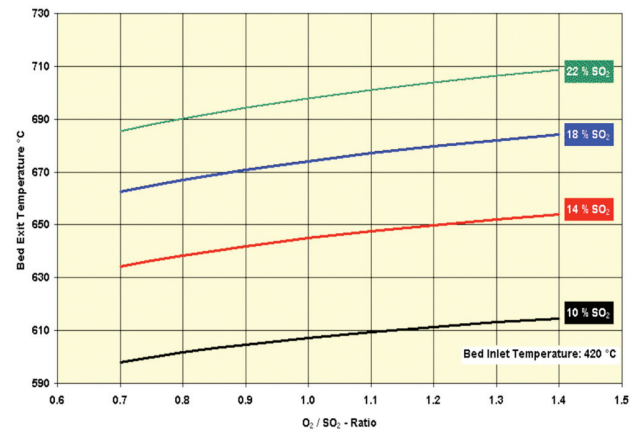

Figure 1—Bed 1 exit temperature vs. SO_2-concentration and O_2/SO_2-ratio

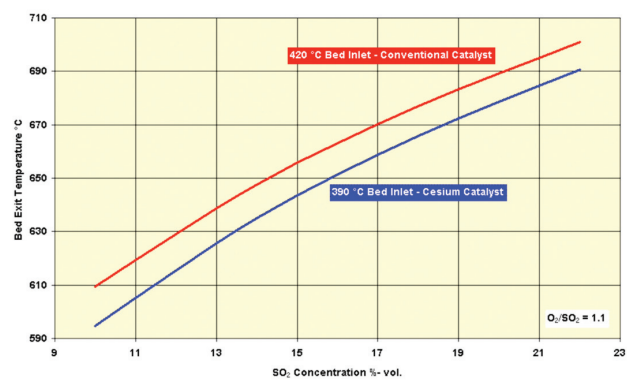

Figure 2—Bed 1 exit temperature vs. SO_2-concentration and catalyst type

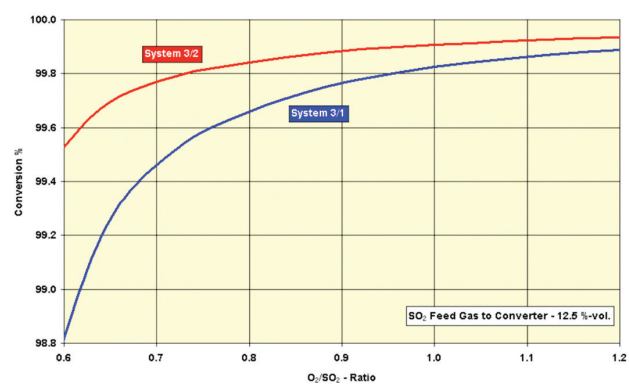

Figure 3—Bed 1 exit temperature vs. SO_2-concentration and catalyst type

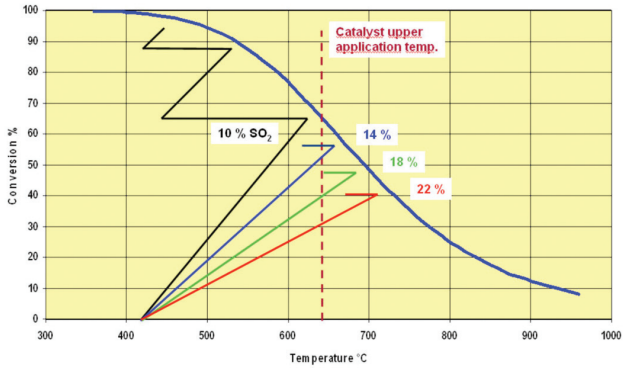

Figure 4—Bed 1 conversion vs. SO_2-concentration

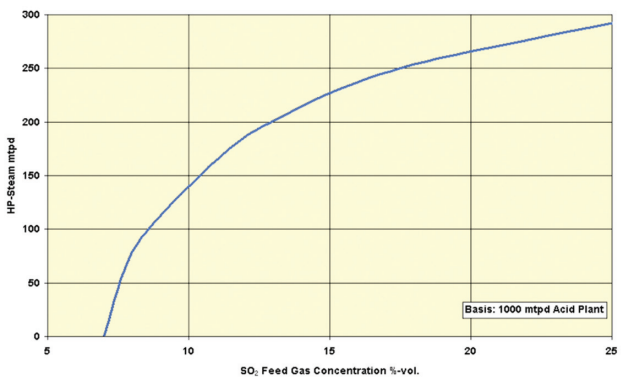

Figure 5—High pressure steam production vs. SO_2-concentration

The LUREC® plant—the right technology for processing of high grade SO_2 gas

In a conventional smelter operation, it appears that the necessity to process the copper converter off-gas simultaneously with the high-grade smelter gas in a single plant, does not justify any attempt to design such an acid plant for any higher SO_2 concentration than approximately 10–12%-vol. This is based on typical Pierce-Smith converter off-gas, containing some fluctuating 5–10% SO_2, while the smelter would discharge a typical 25% SO_2. Minimizing the emission of fugitive gases leads to more sophisticated converter hoods and comprehensive gas collection, which in turn reduces the dilution with ingress air and hence the feed gas to the sulphuric acid plant is less diluted and can readily reach 15–16% SO_2. In that case, the LUREC® technology can offer significant savings as compared to traditional 'dilution' techniques.

This becomes even more pronounced in the case where also high grade SO_2 gas from continuous oxygen enriched converting of copper matte is available. Then the application of direct processing of high-grade gas with the LUREC® process becomes a very viable and attractive option. OUTOTEC has filed patents for this technology, which can process very high grade SO_2 gas while using conventional equipment and catalysts. OUTOTEC'S LUREC® process is equally attractive for new installations, as well as for smelter expansion projects.

A generic flow sheet of the LUREC® process is presented in Figure 6. It shows a typical 5-bed arrangement (3+2) and a recirculation of SO_3 containing gas leaving the bed 3 towards the interabsorption. This recirculation of SO_3 is the key to the LUREC® process. It is used to suppress or limit the oxidation reaction of the incoming SO_2 and hence limits the gas exit temperature to an acceptable level, even when treating very high grade gas.

The recirculation effect can be demonstrated at the operating chart Figure 7, showing gas temperature vs. SO_2 conversion. It is based on an 18% SO_2 feed gas, also containing 13%O_2.

As an effect of the recirculation of SO_3, the thermodynamic equilibrium of the reaction $SO_2 + ½\ O_2 \leftrightarrow SO_3$ is used to limit the SO_2 conversion. At this graph, the operating line for bed 1 is simply shifted upwards as a function of the degree of recirculation and thus limits itself to a lower degree of conversion and equivalent reaction temperature by approaching the equilibrium line. Higher recirculation pushes the operating line upwards and lower recirculation downwards respectively. Based on this simple mechanism, the process can easily adapt varying feed-gas concentrations. It is simply a matter of controlling the first bed exit temperature by automatically adjusting the recirculation rate.

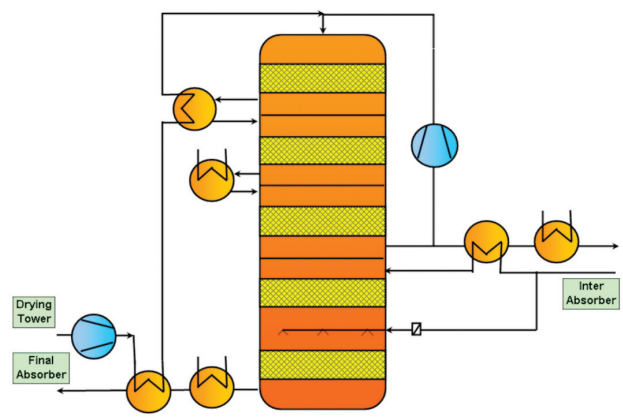

Figure 6—Generic flowsheet LUREC® process

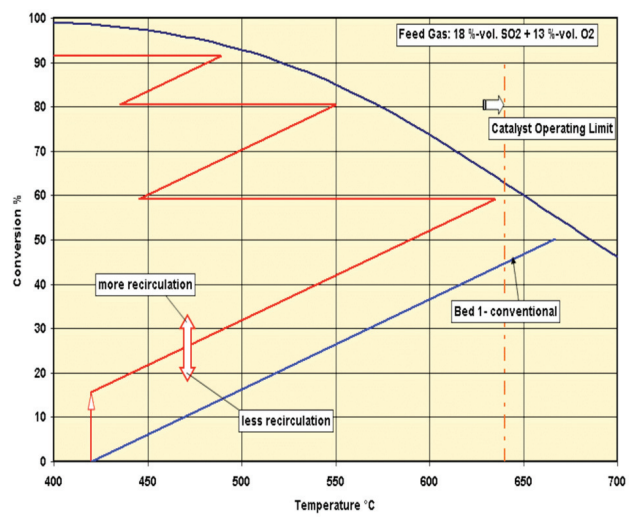

Figure 7—LUREC® recirculation influence on bed 1 exit temperature

The LUREC® process—key to economic smelter acid plant operation

A detailed flowsheet of a LUREC® process for the processing of an extreme 25%-vol. SO_2 gas is presented in Figure 8. The process is practically arranged as a triple absorption plant with seven catalytic beds. The first catalytic section consists of two beds (beds 1,2), which also incorporates the recirculation and is followed by the first interabsorption or pre-absorption. The subsequent five beds are identical with a traditional 5-bed double absorption plant, i.e. arranged in 3/2 sequence (beds 3, 4, 5/beds 6, 7).

Inversely we can also consider this flowsheet as a conventional 3/2 plant with a 2-bed pre-converter/pre-absorber, which takes away such a portion of SO_2 (say from 25 to 12%-vol.) as to enable the more conventional part to operate in a traditional manner. The physical arrangement of the converters/heat exchangers is naturally optimized with respect to minimization of equipment and gas ducts. It makes use of the formidable temperature distribution within the plant and thus two converters with integrated hot heat exchangers and only three external cold heat exchangers are required to match the process requirements. Steam generation equipment, i.e. evaporator and economizer are auxiliaries to maximize the recovery of excess heat.

The application of the LUREC® process offers additional advantages for equipment cost savings, improved operability and other potential credits resulting from better product quality or value added by-products.

The gas originating from the wet gas purification plant and gas drying tower must be heated by gas/gas heat exchangers to the catalyst ignition temperature at approximately 420°C. The capital and operating cost of those heat exchangers are substantial and can be significantly reduced, not only based on a lower gas-flow, but also thanks to the recirculation of hot SO_3 containing gas and the resulting favourable temperature driving forces.

The SO_3 gas generated gas is usually removed from the process gas in packed bed absorbers. The dimensions of those absorbers are generally dictated by the gas volume throughput. This is not the case, however, at very high SO_3 concentrations, e.g. above approximately 12%-vol., where very large acid volumes are required and the dimensioning of the absorber is governed by the flooding limit of the packing size and material, i.e. the liquid to gas ratio L/G. This offers the advantageous application of OUTOTEC's traditional venturi absorbers, which are not sensitive to the said L/G ratio and thus present a cost-effective alternative. OUTOTEC has successfully installed its patented combined absorbers with a venturi-type pre-absorber located in the centre of the vessel with a post-absorber part as an annulus around the venturi. These are shown at the Figure 7 as inter-absorbers. At large plants in particular, the radial gas feeding to the packed section part ensures good and uniform distribution of gas to the entire packing section, which enables a very small packing height and high gas velocities. This in turn results in comparatively small equipment size.

The process can easily cope with fluctuations of the SO_2 concentration in the feed gas. A partial bypass around the first catalytic section (pre-section) ensures that the subsequent 3/2 section always receives sufficient SO_2 gas to maintain autothermal operation, while the pre-section can be kept going by means of the recirculation.

Operation with high grade SO_2 gas naturally produces high-grade SO_3 gas. This can be perfectly used to directly produce oleum with very high concentration, e.g. well above 35% free SO_3, which otherwise can only be obtained by additional process steps, such as distillation and condensation.

Despite enormous efforts in the gas cleaning section, purified smelter off-gas still contains some impurities, which eventually report to the product acid as contaminants. At smelter operations, the acid is often declared as 'fatal acid', a by-product thanks to environmental restrictions and must be marketed as a competitive product to the sulphur burning acid. The quality of the latter is naturally superior, without the impurities usually observed from smelter operations. To match the quality requirements, additional investment in gas purification is necessary, beyond the requirements of the acid

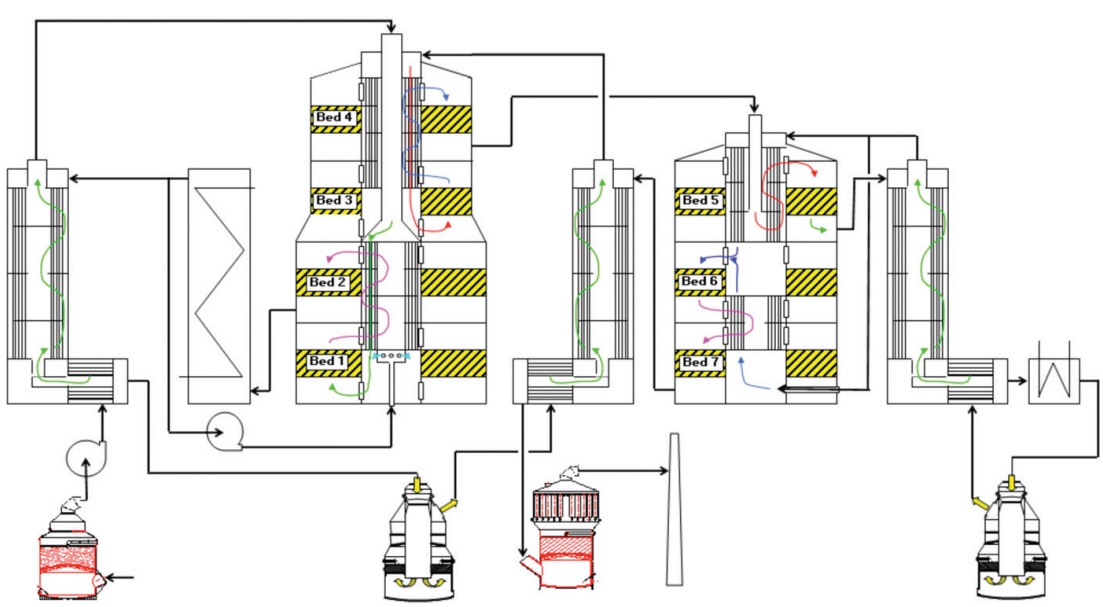

Figure 8—LUREC® process flowsheet –25%-vol.SO_2

plant. This ensures that the smelter acid can be marketed at all and the operator can achieve a small benefit/revenue. The LUREC® process in this configuration offers the withdrawal of two types of product acid, a conventional 'contaminated' portion and an enhanced quality 'clean' por-tion, subject to the discharge location i.e. at the pre-absorber or the inter-absorber. This enables the operator to gain a better market position.

The use of technical oxygen at the smelter has been discussed earlier and it should be recognized that this is an absolute prerequisite for the application of any of the above-discussed processes, including the LUREC®. It becomes increasingly economical to operate an acid plant with a decreasing O_2/SO_2 ratio, so as not to discharge valuable oxygen with the tailgas to the stack. Conventional metallurgical acid plants are operating at ratios of 0.9 to 1.1, or even 1.5 with pure air dilution and can achieve good results regarding the residual SO_2 in the stack, despite the ups and downs in the upstream metallurgical operation. Sulphur burning plants on the other hand operate with ratios down to 0.73 and achieve similar stack results. However, these operate on a steady basis, which enables a more favourable conversion efficiency. The LUREC® process does tolerate an even lower O_2/SO_2 ratio while achieving comparable stack emissions. Hence, the economics of the addition of technical oxygen becomes increasingly viable.

The LUREC® process is also ideally suited for capacity increase of existing smelter operations. It has been discussed earlier, that the process can be regarded as a conventional 3/1 or 3/2 plant with a 1- or 2-bed pre-contact unit. Any existing acid plant can be equipped with such pre-converter unit; in fact OUTOTEC has installed such pre-converters at a number of acid plants to boost the capacity. Subject to the gas composition, it may be fit with or without a pre-absorber unit. Although most acid plants would inherently have some spare capacity, any significant increase of smelter capacity, say of 30% can usually not be accommodated without actually installing an additional parallel acid plant unit.

An add-on LUREC® module can remove any such restriction, while simultaneously debottlenecking the existing acid plant. An example flowsheet is provided in Figure 9. The grey shaded areas are supposed to be the existing acid plant, in this case a 3/2 installation, while the clear part represents the add-on LUREC® module. The latter would process and remove such an amount of SO_2, to ensure that the residual conventional concentration to the existing acid plant is left at say 8-10%-vol. SO_2. The latter can thus be operated in the usual way, without interfering with the new add-on LUREC™ module. An appropriate bypass system would enable sufficient gas to be available at all times to the conventional plant to ensure autothermal operation, while the add-on LUREC® module would be operated at low load, just to keep it ready to kick in at any time.

Figure 10 shows a pre-converter, with the integrated heat exchanger clearly visible.

Economic and environmental review of the LUREC®

Add-on LUREC® module installation to existing facilities

Table I compares the capital cost for 2 cases of capacity increase, namely by 50 and 100%, based on an existing sulphuric acid plant (3 + 1 double catalysis) and a simultaneous reduction of emission from 500 mg/Nm³ to 300 mg/Nm³.



Figure 10—Pre-converter

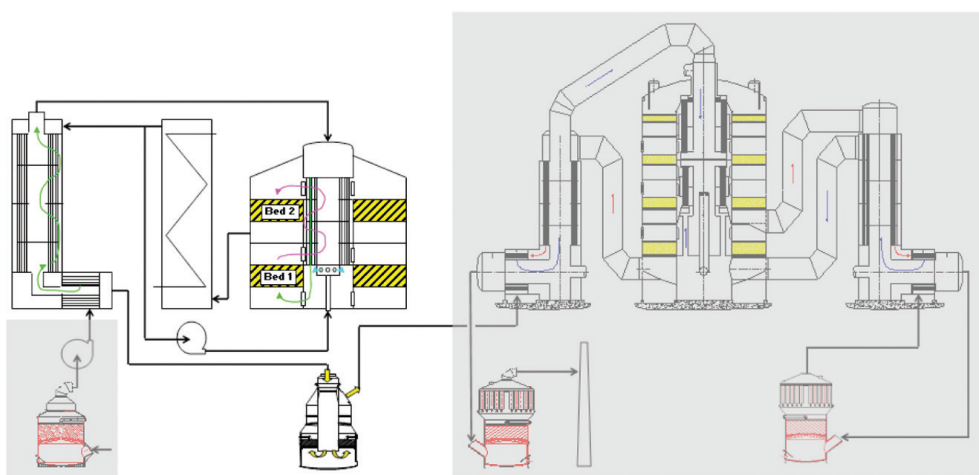

Figure 9—LUREC® add-on module

Table I

Comparing the capital cost for 2 cases of capacity increase

Item	Unit	LUREC® add-on module		Conventional new acid plant 3 + 2 double catalysis	
Capacity increase		**100%**	**50%**	**50%**	**100%**
Capacity total	**mtpd**	**2480**	**1860**	**1860**	**2480**
Capacity of the new plant	mtpd	1420	1100	740	1360
Gas amount	Nm^3/h	110 000	100 000	60 000	110 000
SO_2	%-vol	21.0	17.3	11.5	11.5
O_2	%-vol	17.6	12.6	12.6	12.6
Additional amount of O_2	Nm^3/h	5 500	-	Dilution air	Dilution air
Emission	mg/Nm^3	-	-	300	300
Estimated capital cost	€	88%	77%	100%	115%
Gas to existing plant					
Capacity of existing	mtpd	1060	760	1120	1120
Gas amount	Nm^3/h	95 000	85 000	100 000	100 000
SO_2	%-vol	10.5	8.4	10.5	10.5
O_2	%-vol	12.6	8.7	12.6	12.6
Emission	mg/Nm^3	300	300	300	300
Gas to stack					
Gas amount total	Nm^3/h	80 000	74 500	133 800	175 100
Emission	mg kg SO_2/t H_2SO_4	300 0.235	300 0.290	300 0.520	300 0.515
Conversion	%	99.96	99.95	99.92	99.92

Table II

Cost savings involved at the gas cleaning section

Data of the existing plant with a LUREC® add-on module					Existing plant			
	SO_2-conc. (%)	O_2-conc. (%)	Gas amount (Nm^3/h)	Capacity mtpd	SO_2-conc (%)	O_2-conc (%)	Gas amount (Nm^3/h)	Emission (Nm^3/h)
Existing Plant 3+1	11.5	12.6	100 000	1 240	11.5	12.6	100 000	300
With LUREC® add-on module 1+3+1	21.0	15.7	100 000	2 260	10.6	11.4	82 500	300
	21.0	17.6	100 000	2 260	10.5	12.6	82 000	300
	17.2	12.6	100 000	1 860	8.4	8.7	85 000	300
	15.0	12.6	100 000	1 620	7.7	9.6	87 000	200

Basically, the existing plant with a capacity of 1 240 mtpd is operating at a gas amount of 100.000 Nm^3/h with 11.5% SO_2 and 12.6% O_2. In both cases, the options considered are the installation of an additional new conventional plant with the necessary capacity, or the installation of an add-on LUREC® module to the existing plant.

It is obvious that in case the smelter capacity increase is boosted not only by proportional increase in off-gas production, but by the application of larger proportion of oxygen and hence result in a lower off-gas stream with higher SO_2 concentration (or a combination thereof), the advantages for the use of an add-on LUREC® module are even more pronounced.

The process varieties using the recycling concept are numerous and each application would be tailor made. As an example, the concept can be applied also if the gas concentration would increase from say 13%-vol. to 15%-vol. SO_2, where a complete module as sketched in Figure 8 would not be necessary. In this case, a pre-absorber unit may not be required and the pre-converter may exist of only one single bed including heat removal equipment. This is of course subject to the availability and cost of technical oxygen, emission regulations and other factors and must be investigated on a case-by-case basis

Some typical configurations are shown in Table II.

Any additional cost savings involved at the gas cleaning section for both cases is neglected for the purpose of the above comparison.

At the LUREC® case, it should be noted that the existing plant could be operated in future at significant lower gas flow and feed-gas concentration as compared to the existing design conditions which reduces the total SO_2-emissions dramatically.

The LUREC® process—key to economic smelter acid plant operation

New LUREC® module installation—greenfield

Table III compares the capital cost of conventional sulphuric acid plants, using air dilution to operate with 12%-vol. SO_2 at the converter inlet, with a plant based on LUREC® process with 18%-vol. feed gas. The raw gas basis is: 100 000 Nm³/h with 18%-vol SO_2, resulting in a nominal capacity of 1930 t/d. The gas flows to the various catalytic beds are shown for clarity.

The process varieties using the recycling concept are numerous and each application would be tailor made. All possibilities of design are of course subject to the availability and cost of technical oxygen, emission regulations and other factors and must be investigated on a case-by-case basis. The above shown figures are only the result of typical examples.

As a further variety, Table IV compares the capital and operating cost of a conventional sulphuric acid plant of 3 000 mtpd, using air dilution to operate with 12 %-vol. SO_2 at the converter inlet, with a plant based on the LUREC® process with a 25%-vol. feed-gas. The comparison is based on an off-gas consisting of 36%-vol. SO_2 plus 4.5%-vol. O_2 originating from the smelter operation.

It appears that the operating cost of both alternatives are at a comparable level, despite the use of oxygen at the LUREC® process. Labour and other running cost are considered identical for both options. No credit for the superior product acid quality and lower SO_2 emissions have been included for the LUREC® process. Significant savings will result from annual maintenance and capital cost, estimated to be approximately 25% less in favour of the LUREC® process, while the gas cleaning section may offer additional savings, which is not considered in this comparison.

Table III

Comparative the capital cost of conventional sulphuric acid plants

Item		Unit	Convent. 3 + 1	LUREC® 3 + 2	Convent. 3 + 2	LUREC® 4 + 2	LUREC® 1 + 3 + 1
Gas amount	Bed 1	Nm³/h	150 000	130 000	150 000	130 000	130 000
	Bed 2	Nm³/h	145 000	95 000	145 000	95 000	83 000
	Bed 3	Nm³/h	142 000	92 000	142 000	92 000	80 000
	Bed 4	Nm³/h	124 000	75 000	124 000	91 000	79 000
	Bed 5	Nm³/h	-	74 000	123 000	75 000	73 000
	Bed 6	Nm³/h	-	-	-	74 000	-
SO_2		%-vol.	12.0	18.0	12.0	18.0	18.0
O_2		%-vol.	13.3	15.4	13.3	15.4	14.4
Emission		mg/Nm³	500	500	300	300	300
Capital cost		€	125%	100 %	133%	107%	110%

Table IV

Comparative the capital and operating costs of a conventional sulphuric acid plant

Item	Unit	Amount		Specific cost	Annual cost € m	
		Conventional	LUREC®		Conventional	LUREC®
Acid plant capacity	mtpd	3 000	3 000	-	-	-
Gas flow from smelter	Nm³/h	77 500	77 500	-	-	-
Gas flow to converter	Nm³/h	232 500	109 600	-	-	-
Dilution air added	Nm³/h	155 000	24 800	-	-	-
Technical oxygen added	Nm³/h	0	7 300	-	-	-
SO_2 conc. to converter	%-vol.	12.0	25.4	-	-	-
Gas flow to stack	Nm³/h	190 600	67 700	-	-	-
SO_2 stack emission	ppmv	150	200	-	-	-
SO_2 stack discharge	kg/h	84	40	-	-	-
SO_2 stack discharge	kg/mtpd	0.67	0.32	-	-	-
SO_3 stack discharge	kg/mtpd	0.0381	0.0135	-	-	-
Electrical energy	kW	6 650	5 320	0.08 €/kWh	4.469	3.575
Cooling water	m³/h	4 474	3 628	0.04 €/m³	1.503	1.219
Boiler feed water	t/h	23.6	35.7	2.00 €/t	0.395	0.599
Process water	t/h	16.9	19.3	0.50 €/t	0.071	0.081
HP steam export	t/h	23.0	35.1	18.00 €/t	-3.478	-5.307
Oxygen	t/h	0.0	10.5	31.00 €/t	0	2.734
Total operating cost	€m/a				2.960	2.901
Estimated capital cost		125%	100%	-	-	-

Table V presents another case. This is to compare a greenfield new installation of 2 000 mtpd capacity with a moderate SO_2 concentration of 18% versus a conventional plant design with 12%-vol. SO_2.

It is obvious from the figures that the LUREC® process offers significantly advantages over a conventional acid plant. It results in reduced gas flows throughout the plant, which in turn lead to adequate savings in capital cost in the order of 20%. The savings in operating cost, particularly for electrical energy and cooling water, are equally important and in excess of 20%. A considerable higher potential heat recovery, i.e. production of high pressure steam, can be realized, thus contributing additional revenues for the acid plant.

With ever increasing and more stringent environmental restrictions, the amount of gas discharged to the atmosphere, together with the loading of SO_2 and SO_3 become of vital importance and their further reduction can cause enormous capex and opex. Here the LUREC® does inherently offer substantial advantages, reducing the stack gas amount by 40% and the total SO_2 and SO_3 discharge by 25 and 40% respectively.

Greenfield installation of a LUREC® plant in Yanggu Xiangguang Copper, Shangdong Province, China (nominal capacity 2 340 t/d)

In 2005, OUTOTEC was awarded a contract for the design and delivery of key equipment for the copper flash smelter, the flash converter (OUTOTEC-KENNECOTT), the tankhouse and the sulphuric acid contact section. The construction of the plant was finished in July 2007. Cold commissioning and testing took place in July and August 2007. The hot commissioning commenced at 8.8.2007 and was completed after 6 weeks in September.

The plant has operated up to full acid capacity, and has operated with SO_2 concentrations of 14 to 17% vol. The technology has been validated and the plant is now OUTOTEC'S first existing reference.

Design basis for the acid plant

The sulphuric acid contact section is designed to process 136 000 Nm³/h metallurgical off-gases with a SO_2 content of approx. 16–18% vol. entering the cold heat exchanger from the drying section. Table VI provides typical feed gas composition, which represents the design basis for the acid plant.

Table V

Compare a greenfield new installation

Basis 2,000 mtpd		12 %-vol. SO_2 conventional	18 %-vol. SO_2 LUREC® process	LUREC® savings (%)
Gas flow drying tower Nm³/h		158 000	105 000	- 34%
Gasf low absorbers Nm³/h		130 000	80 000	- 38
Gas flow converter Nm³/h	Beds 1+2	158 000	135 000	- 15%
	Bed 3	149 000	96 000	- 36%
	Beds 4+5	130 000	79 000	- 39%
Stack gas Nm³/h		129 000	76 000	- 41%
Heat exchanger surface m²		13 500	9,900	- 26%
Plant cost installed		100%	79%	- 21%
Power consumption kWh/t H_2SO_4		55	43	- 22%
Cooling water consumption m³/t H_2SO_4		37	29	- 21%
Heat recovery kWh/t H_2SO_4	165	205	+ 24%	
Emission kg SO_2/t H_2SO_4	1.20	0.90	- 25%	
Emission kg SO_3/t H_2SO_4 (Basis 30 mg/Nm³)		0.076	0.046	- 39%

Table VI

Typical feed gas composition

Inlet drying tower (without dilution air)	SO_2	CO_2	O_2	N2	Total
Volume flow (Nm³/h)	21.882	4.269	10.418	66.448	103.017
Concentration (% vol.)	21.24	4.14	10.11	64.51	100
Temperature (°C)	45				
Pressure (kPa)	-13.8				
SO_3 (mg/Nm³)	< 20				
Dust [mg/Nm³)	< 1				
Exit Drying tower (incl. dilution air)	**SO_2**	**CO_2**	**O_2**	**N2**	**Total**
Volume flow (Nm³/h)	21.882	4.269	17.299	92.550	136.000
Concentration (% vol.)	16.06	3.11	12.72	68.11	100
Temperature (°C)	100				
Pressure (kPa)	144.8				

The LUREC® process—key to economic smelter acid plant operation

Process description with flowsheet

Refer to the following simplified process flowsheet, Figure 11.

OUTOTEC'S LUREC® process—featuring recirculation of a small processed gas stream branched off after the 3rd bed of the converter—allows the direct feeding of process gases with a SO_2 content of 16–18 % vol. With a substantial reduced amount of dilution air at the inlet of the plant—compared with a conventional design—the engineering design applying LUREC® technology allows downsizing of all installed equipment up to the stack. Beside the savings on capital cost, the operating costs are significantly reduced by lower power requirement at the main blower based on handling a significantly lower amount of process gas.

This section comprises the following main equipment based on the double absorption process:

- Stainless steel converter vessel with 3 integrated gas/gas heat exchangers:
  - Hot heat exchanger made of stainless steel
  - Hot reheat exchanger made of stainless steel
  - Intermediate reheat exchanger made of stainless steel.
- External cold heat exchanger made of boiler steel with sacrificial part (not shown) made of stainless steel
- External cold reheat exchanger made of boiler steel with sacrificial part (not shown) made of stainless steel
- Heat recovery unit 1: waste heat boiler (water tube type) with economizer
- Heat recovery unit 2: waste heat boiler (water tube type) with economizer
- LUREC® recirculation blower.

The purpose of the converter section is to convert gaseous SO_2 to SO_3 promoted by a vanadium pentoxide catalyst. The governing chemical reaction is described as follows:

$$SO_2 + \tfrac{1}{2}O_2 \leftrightarrow SO_3 \quad -99 \text{ kJ/mole}$$

This reaction is highly exothermic and is governed by the thermodynamic equilibrium, subject to temperature, gas composition and pressure. The heat generated will be mainly used to heat up the process gas exiting the SO_2 gas blower, while the excess heat is transferred to high pressure steam of 25 bar.

The entire converter including all of the internal heat exchangers are made of stainless steel, to continuously withstand high operating temperatures up to 640°C.

In order to achieve and ensure sustainable high conversion rates, two main issues must be focused on:

- Positive isolation of catalytic beds to avoid gas bypassing any catalyst layer. This is ensured by a completely welded design, e.g. all dividing plates inside the converter are fully welded at the shell and core tube, and hence absolutely gas-tight.
- Uniform gas distribution to the individual catalyst beds. Radial flow gas inlets at all beds will ensure good gas distribution.

From the blower exit, the cold gas is first directed through the external cold heat exchanger (shell side) and the internal hot heat exchanger (tube side) to the first catalyst bed of the converter, entering this bed with approximately 390–400°C. A caesium-promoted catalyst is used here as an ignition layer.

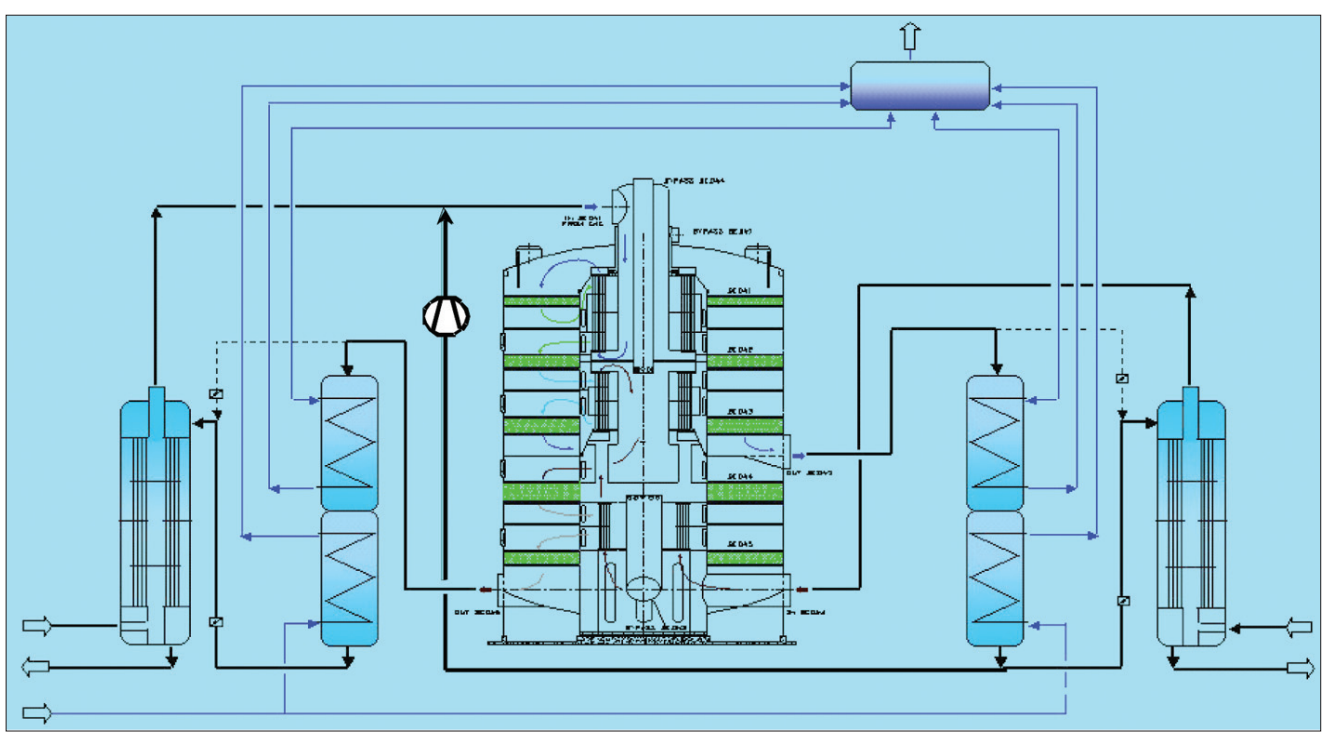

Figure 11—Simplified process flowsheet—Yanggu converter section



Figure 12—Yanggu contact section

At this first catalyst bed, 57–60 % of the SO_2 is converted to SO_3. The exothermic reaction would lead to a bed outlet temperature in excess of 660°C, well beyond the tolerable operating limit of vanadium catalysts. To lower the bed outlet temperature to an acceptable level of 620–630°C, a part-stream of precooled SO_3 gas is recirculated from the outlet of waste heat boiler 2 to the first bed of the converter via the recirculation blower. The SO_3 enriched gas suppresses the adiabatic reaction temperature in the first bed to a maximum of 620–630°C.

To enable optimum conversion conditions for the second catalyst bed, the outlet gas from the first catalyst bed is cooled down at the integrated hot heat exchanger (shell side) to a temperature of approximately 440°C prior to entering the second catalyst bed.

In bed 2 approximately 82–83 % (cumulative) of the total SO_2 is converted to SO_3 with the resulting adiabatic reaction temperature of approximately 535–540°C. The cooling of the gas from this second catalyst bed down to approximately 450°C takes place in the internal hot reheat exchanger (shell side) heating up SO_2 feed gas for the fourth catalyst bed on the tube-side. The cooled process gas from the second bed enters the third catalyst bed.

At the third catalyst bed further SO_2 is converted to SO_3 achieving an adiabatic gas outlet temperature of approximately 485°C. After bed 3 the total resulting conversion is approximately 91–92 %. Leaving the third catalyst bed, the SO_3 gas is cooled to approximately 280°C in the waste heat boiler and economizer 2, where the excess heat is recovered as 25 barg steam. A minor part of the gas is branched off to establish the LUREC® recirculation. After further cooling at the cold reheat exchanger (incl. sacrificial part), the process gas enters the intermediate absorber at approximately 165°C.

The SO_3, which is contained in the gas, is absorbed in the intermediate absorber and transformed to sulphuric acid.

Downstream the intermediate absorption, the process gas with a temperature of approx. 80°C is returned through the shell-side of the cold reheat exchanger (incl. sacrificial part) and further heated up to the inlet temperature of the fourth catalyst bed of approximately 415°C in the both integrated intermediate (tube side) and hot reheat exchanger (tube side). Leaving bed 4, an overall conversion rate of approximately 99.6% is already achieved.

The process gas stream leaving the fourth bed at approximately 460°C is cooled to the required fifth bed inlet temperature of approximately 380°C by the intermediate reheat exchanger (shell side).

Bed 5 features caesium promoted catalyst to achieve an overall conversion efficiency in excess of 99.9 % of the total SO_2.

Leaving the fifth catalyst bed, the SO_3 gas with an outlet temperature of approx. 382 °C is led to waste heat boiler 1 with economizer, followed by further cooling at the cold heat exchanger (with sacrificial part) prior to entering the final absorber at approximately 160°C.

The SO_3, which is contained in the gas, is absorbed in the final absorber and transformed to sulphuric acid.

Temperature control

The converter system is equipped with appropriate gas bypasses (not shown in Figure 9) to control the gas inlet temperatures at each of the individual catalyst beds. Those bypasses are arranged both external and internal of the converter, while all gas dampers are located outside.

The exit temperature of the first bed is controlled by the LUREC® process gas recirculation. To keep this temperature constant, the recirculation blower is equipped with a variable speed drive, controlled by the said exit temperature. When the exit temperature increases, e.g. in case of higher SO_2 concentration feed gas, the amount of recirculation gas is increased and vice versa.

Gas bypasses around the waste heat boiler/economizer systems are installed to facilitate start-up operation as well as situations where the gas concentration may fall below the normal design level. This enables autothermal operation of the plant with SO_2 concentrations as low as 8%-vol.

Computer simulation

Large size SO_2 converters and complicated gas flows and patterns do require careful design and the use of computer flow- and stress modelling to ensure that the plant and

process will perform in accordance with the requirements and the predictions. At selected areas at each vessel, such investigations using finite element methods are conducted intermittently during the design and the results used as feedback to adjust the relevant elements if required. Figure 13 presents typical output profiles for gas velocity and temperature distribution at certain locations within the SO_2 converter.

Converters are generally designed in accordance with the applicable pressure vessel code, irrespective of the declaration as pressure vessel, which varies from one to another location. The finite element method can determine the most critical mechanical items under load and temperature. It is also used, to simulate various plant operating conditions, such as high and low SO_2 concentration (and hence temperatures), start-up and shutdown, preheating and the like, in order to ensure that all elements within the converter can freely move under all circumstances, based on their individual thermal expansion. Figure 14 presents a typical stress output of some converter elements.

Extensive process investigations and simulations must be performed prior to the definition of the equipment and plant design parameters. A number of operating scenarios are simulated throughout the entire plant, obviously with emphasize on the SO_2 conversion. Those calculations are made by using in-house computer programs, which are used to determine the impact of variations of process parameters, such as process gas flow, dilution air or oxygen flow, catalyst operating temperatures, emissions, effects of recirculation and the like. It can also be used to simulate catalyst activity loss and its impact on the operation. The program is available for potential clients who want to familiarize with the process and its application to their specific operation. It can also later be used for operator training. Figure 15 shows a typical input screen of the program.

Summary

The newly developed LUREC® process can directly handle off-gas with significantly higher gas concentrations as compared to conventional smelter acid plants. While the latter are limited to 12–13 (14) %-vol. of SO_2, the LUREC® can process up to 25 %-vol. SO_2 directly, without excessive air dilution. It is entirely based on well-proven equipment and unit operations. The first industrial application, a greenfield plant operating with 16–18 %-vol. SO_2 is in operation since the summer of 2007.

In the meantime, OUTOTEC does realize and enjoy an enormous interest of the metallurgical sulphuric acid industry in this technology. A second installation, also using the LUREC® process to cope with higher SO_2 concentrations based on increased smelter capacity will soon be demonstrating the superiority of the process over a conventional arrangement.

The application of the new LUREC® process to modern smelter acid plants operating with high SO_2 gas concentrations, does lead to a substantial reduction of the specific size of the gas processing equipment and related savings in capital and operating cost as well as inherently lower emissions. ◆

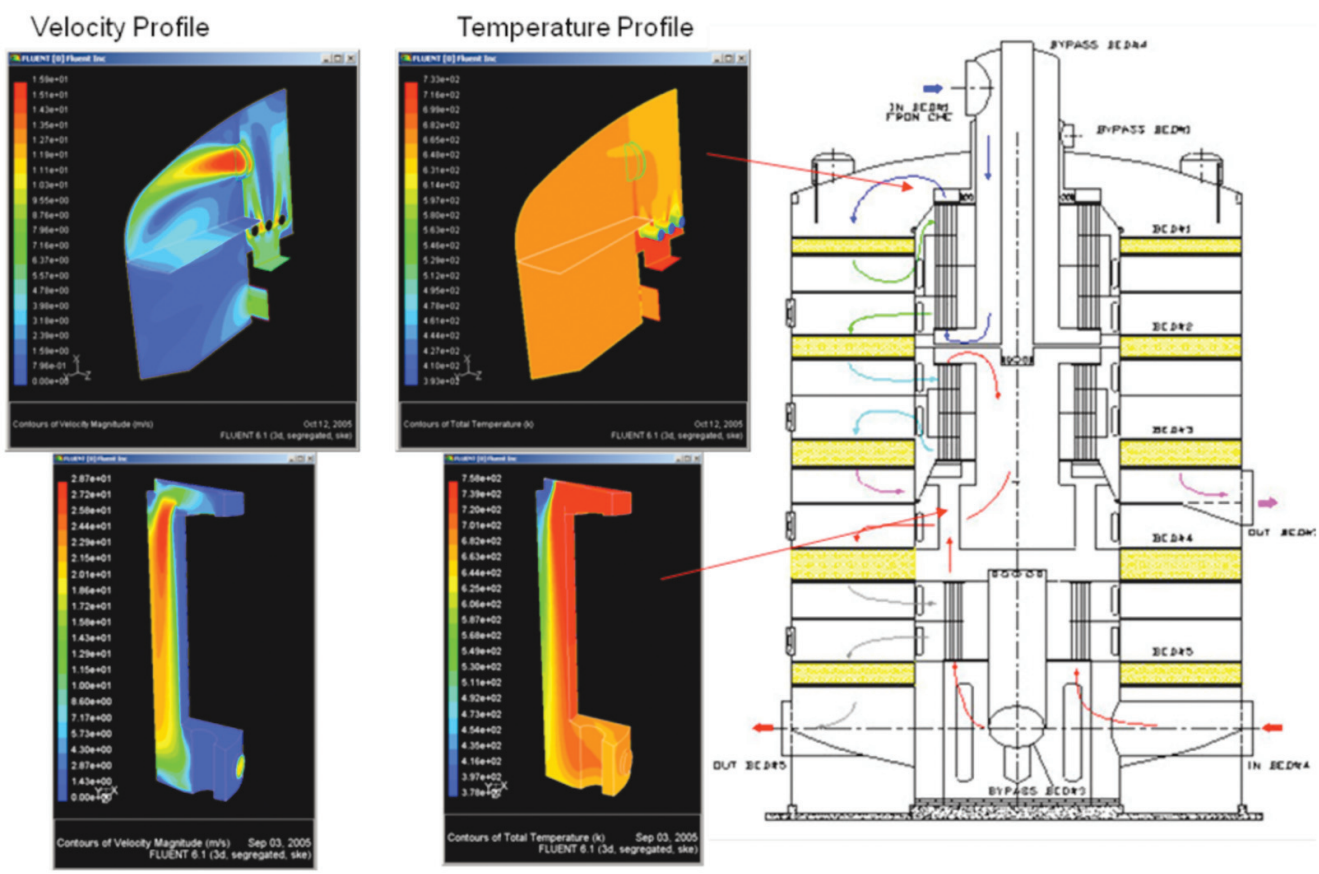

Figure 13—Converter flow simulation

The LUREC® process—key to economic smelter acid plant operation

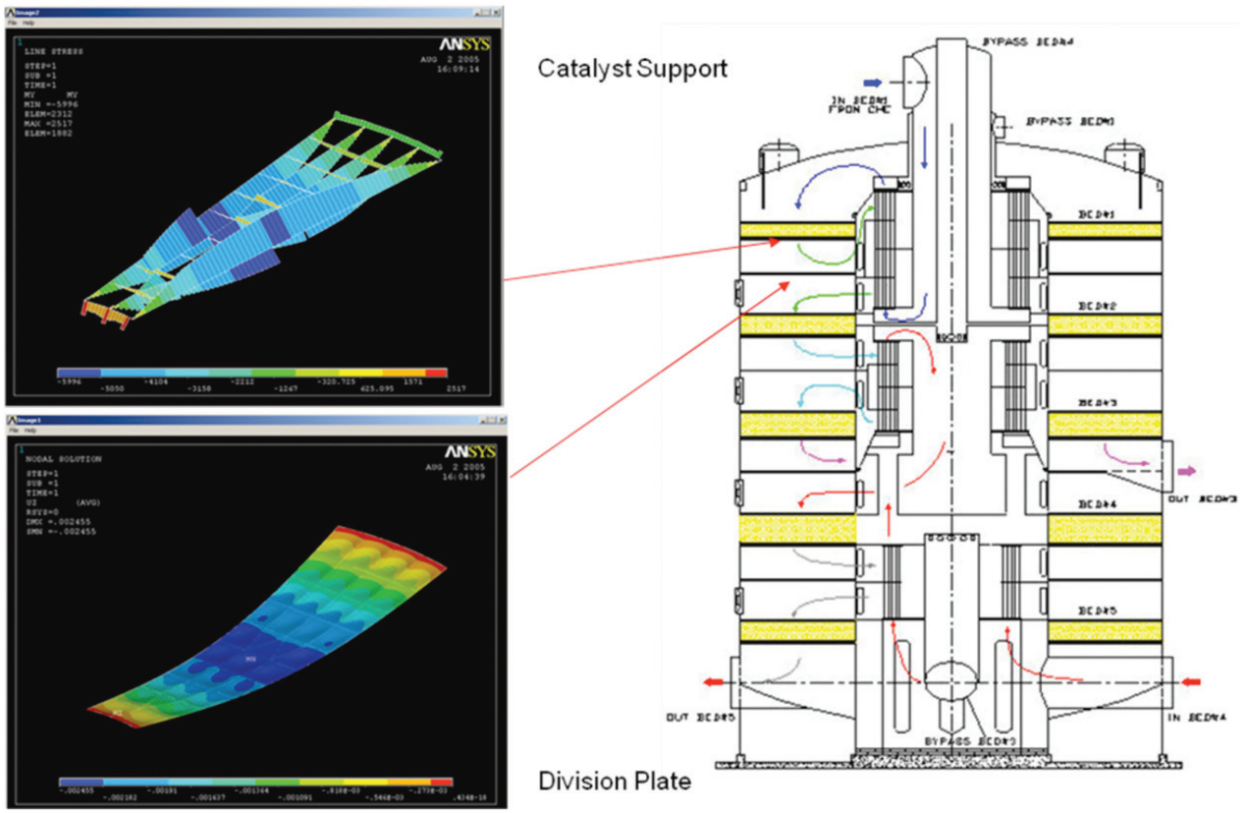

Figure 14—Converter stress calculation

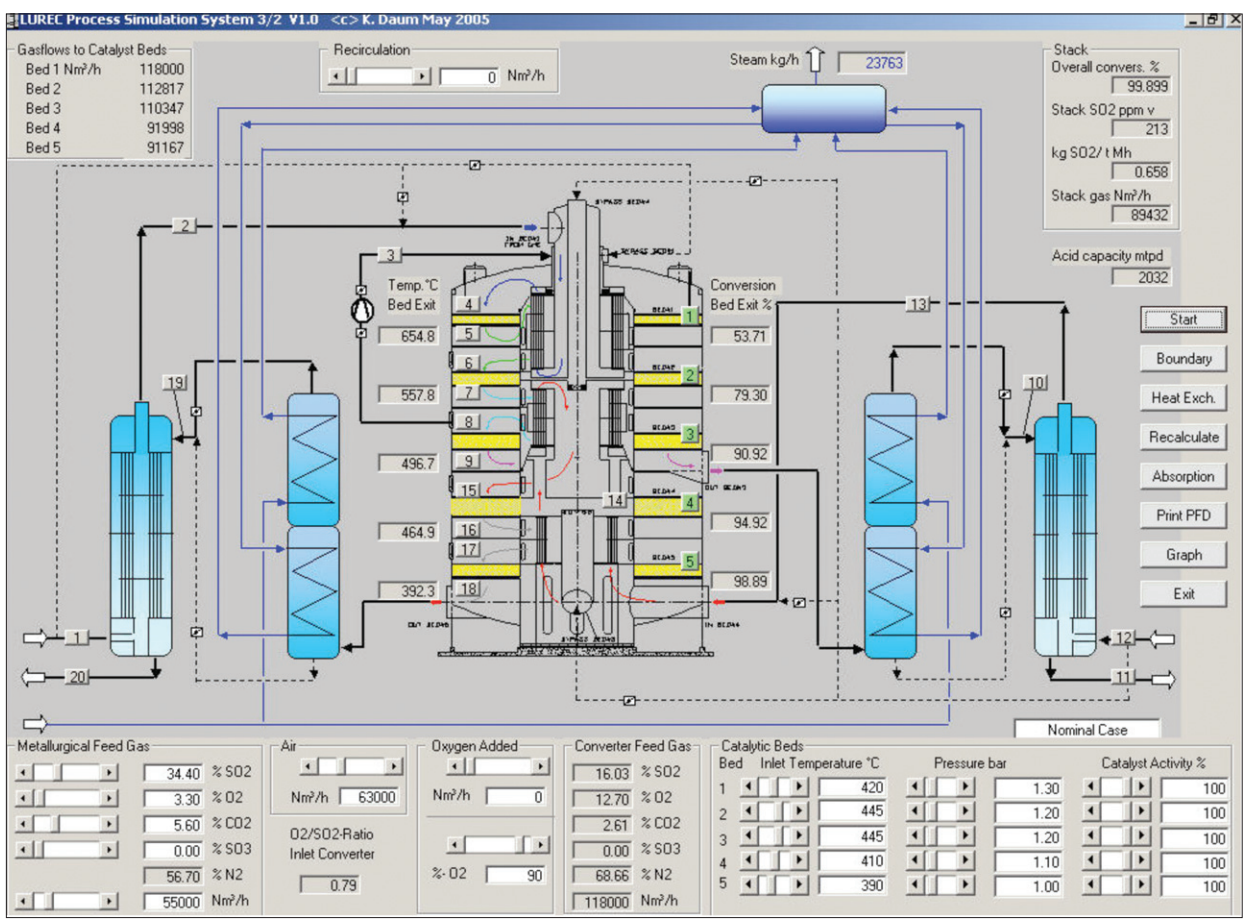

Figure 15—Computer simulation program