

LESSONS LEARNED FROM SCR EXPERIENCE OF COAL FIRED UNITS IN JAPAN, EUROPE AND USA; ARE THESE ENOUGH?

Dr. Anupam Sanyal

International Environmental & Energy Consultants, Inc.

Naperville, Illinois 60563

And

William Ellison

Ellison Consultants

Monrovia, Maryland 21770

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Introduction

Development of SCR in Japan, Europe & USA

- ~23,100 MW Total Japanese Utility Capacity Comprising 61 Plants
- ~55,000 MW Total European Utility Capacity
- ~Until end 90's, 2,211 MW of US Capacity. By 2004, this is Expected to Increase to over 100,000 MW \equiv Over 33% of the Total Coal Fired Capacity of the USA

Pioneers of Japan, Europe & USA for SCR Installations on Coal Fired Commercial Units Sequence of Milestones

1975	Japanese – Pilot plants firing oil, gas & refinery gas. 1 st coal fired Japanese demo. on ½ x 175 MW of Chogoku Electric's Shimoneski Unit followed by Hokkaido Electric's Tomato - Atsuma (1/4 x 250 MW) & EPDC's Takehara 250 MW Unit 1 Low Dust Demo.
1980	Fed. Republic of Germany's ordinances for 111 ppm (0.12 lb/Mbtu) (200 mg/Nm ³) from large boiler installations
1985	Arsenic Poisoning reported in German pilot plants. 1 st coal fired German <u>Dry Bottom</u> Commercial Unit is Neckerwerk's Altbach/Deizisau 420 MW Unit 5 & 1 st coal fired German slag tap 175 MW unit of Knepper C
1990	1 st <u>New</u> US P.C. Fired PG&E's 265 MW Carney's Point Cogen Plant & 1 st <u>Retrofit</u> Merrimack Unit 2 (1 st US Gas Unit is LADWP Haynes)
1995	1 st PRB Application Feb 2000 to AECI's 600 MW New Madrid #2 E NO _x 93%, NH ₃ Slip 3ppmvdc 1 st largest cyclone unit (700 MW) firing high sulfur bit coal to TVA's Paradise #2 Spring 2000; ENO _x 90% NH ₃ Slip 2 ppmvdc
2000	1 st Arsenic poisoning in Stanton #2 of OUC

Performance of Earlier Japanese SCR Plants on Coal Fired Utility Boilers

Plant Owner	Plant Site	<u>Boiler</u> No. MW		Catalyst Type	Pitch (mm)	SV {h ⁻¹ }	<u>NO_x (ppm)</u> In Out		DeNO _x (%)	NH ₃ (ppm) Slip	Up Year
EPDC	Takehara	1	250 x ½	Plate	10	2,300	350	67	81	4	1981
EPDC	Takehara	2	250 x ½	Tube	-	2,000	350	67	81	4	1981
EPDC	Takehara	3	700	Plate	10	2,370	250	48	81	2	1983
Chugoku	Shimonoseki	1	175	Honeycomb	10	3,000	420	180	57	1	1980
Chugoku	Shin-Ube	3	156	Honeycomb	7	4,000	400	140	65	2	1981
Chugoku	Mizushima	2	156	Plate	6	2,200	350	120	65	2	1984
Shikoku	Saijo	1	156	Honeycomb	7	4,000	380	130	65	1	1983
Shikoku	Saijo	2	250	Honeycomb	7.5	-	330	90	70	1	1983
Tokyo	Yokosuka	1,2	265	Honeycomb	7	4,000	200	60	70	1	1984
Chugoku	Shin-Onada	1	500	Honeycomb	7.4	2,800	200	40	80		
Tohoku Electric	Sendai	1	175	Plate		2,600	400	160	60	2	1983

Typical European Units Providing NO_x Reduction >85% (High Performance SCR)

Unit	NO _x %	Initial Space Velocity (h ⁻¹)	Final Space Velocity (h ⁻¹)	Reactor Size (MW)	Comment
Knepper C	90	Not defined	1412	175	Combined high initial NO _x , small reactor size, and relatively low space velocity provide ideal conditions for maximum NO _x removal
Walheim	85	2449	1839	140 + 40	Same as Knepper
Staudinger ½	84	-	-	63	Extremely small reactor size
Staudinger 3	85	-	-	293	Reactor size is one of the largest for high performance SCR Catalyst Management
Tiefstack	85	1257	943	100	Excess of catalyst, small reactor size
Bremen	85	3152	2364	170	Catalyst volume is not excessive, but tower design boiler and small reactor promote good gas distribution and mixing
Kiel	83	2308	1528	350	Relatively large reactor, with excess of catalyst, APH corrosion/SO ₃
Franken	83	1785	1250	220	Excess catalyst volume

The background is a solid dark blue. A thin, light blue curved line starts from the top left and arcs towards the right. On the right side, there is a wedge-shaped area of a lighter blue color, pointing towards the center.

Japanese & European Coal Quality Indigenous and Imported

Analyses of Typical Japanese Coals

Area	Mining District or Seam	As Received		Moisture and Ash-Free HHV			Hardgrove Grind.
		% Ash	% H ₂ O	% VM	Btu/lb	MJ/kg	
Kyushu	Tagawa	35.4	3.9	51.2	13,660	31.81	45
	Iizuka	22.1	2.2	48.3	14,410	33.5	44
	Hiyoski	24.5	3.0	40.0	14,700	34.2	49
	Kokura	39.1	6.1	53.6	13,160	30.6	37
	Onoura	23.8	2.4	46.2	14,440	33.6	46
	Yamano	51.3	2.2	45.8	13,400	31.2	43
	Meiji-Saga	15.4	2.2	51.7	14,800	34.4	47
	Shinkokusko	23.3	3.6	46.9	14,690	34.2	51
Shikoku	Takamatsu	29.7	4.4	47.6	14,040	32.6	38
Yamaguchi	Sanyo	31.7	2.2	9.5	14,900	34.7	72
Hokkaido	Sunagawa	38.0	1.6	49.7	13,710	31.9	49
	Bihai	15.1	3.6	45.9	14,330	33.3	47
	Yubetsu	21.6	2.4	44.9	14,740	34.3	53
	Akama	30.8	2.6	46.4	14,590	33.9	54
	Hahoro	16.8	12.9	55.8	14,510	33.7	36
	Horonai	8.2	3.4	48.4	14,490	33.7	38

Characteristics of Typical European Indigenous & Coals Imported by Europe & Japan

Analysis	Germany Ruhr Valley	Australia N.S. Wales	S. Africa Transvaal	Columbia Guasare	Poland
Ash	9.5%	8.0%	14.0%	7.0%	21.0%
VM	37.7%	25.0%	24.0%	36.0%	28.0%
FC	0%	54.0%	54.0%	48.0%	43.5%
Moisture	4.90%	13.00%	8.00%	9.00%	7.50%
Heating Value	12,858	10,854	11,240	11,755	9,926
Sulfur	0.90%	0.30%	0.60%	0.60%	0.90%
C	69.0%	64.0%	65.0%	69.0%	55.7%
H	5.0%	4.0%	4.0%	5.0%	4.4%
N	1.9%	1.4%	1.6%	1.2%	1.1%
Ash Chemistry	Pct of Ash	Pct of Ash	Pct of Ash	Pct of Ash	Pct of Ash
Si	38.90%	70.00%	43.00%	57.00%	51.00%
Al	23.20%	21.00%	32.00%	24.00%	25.00%
Fe	11.60%	3.00%	4.00%	9.00%	7.00%
CA	9.70%	1.00%	9.00%	1.10%	3.40%
Mg	2.53%	0.60%	1.70%	0.11%	2.20%
Na	0.48%	0.40%	2.00%	0.90%	1.00%
K	1.185%	1.00%	0.50%	1.50%	2.20%
Ti	0.80%	1.00%	1.50%	1.00%	1.60%
P	0.10%	0.50%	2.00%	0.40%	0.50%
SO ₃	8.34%	0.20%	6.00%	1.00%	2.00%
Error	0.41%	0.71%	-0.81%	1.30%	1.30%
Total Alkali	14.56%	3.00%	13.20%	3.61%	8.80%
Total Alkali/S	16.18	10.00	22.00	6.02	9.78
Total Ca/S	10.78	3.33	15.00	1.83	3.78
Sulfur (%)	0.9%	0.3%	0.6%	0.6%	0.9%

Highlights of SCR Experience of Japan & Europe

- **Coal** – Mostly imported, med. Sulfur, low arsenic sub-bit.

Design Features	Remarks
Inlet NO _x lb/Mbtu	0.50-1.25
deNO _x E	50-90%
NH ₃ Slip ppmvdc	1-10
SO ₂ -SO ₃ Conv. %	<2%
SV hr ⁻¹	1,257-4,000
Low/High Dust	Both
• Catalyst Pitch & Geometry Pitch	7-10 mm; pipe, honeycomb, plate, (graduated) fiber-reinforced corrugated
Composition	V ₂ O ₅ , TiO ₂ , MoO ₃ on S.S. or Ceramic
Management	By coupon testing, inspection, cleaning regularly, poisoning/blockage

Highlights of SCR Experience of Japan & Europe (Cont'd.)

- | | |
|---|---|
| ● SCR Process | Parameters |
| Gas Velocity Dist ^N | 15-20% SD |
| NH ₃ / NO Ratio SD | 4-6% |
| Physical & CFD Modeling | Yes |
| ● Reactor Layout | Various – e.g. For simple tower boilers
“Elbow” to complicated layouts |
| achieved by prior modeling of range | specific needs – e.g. wide load |
| ● Reagent Type & Application | Anhydrous NH ₃ (Mostly)
AIG Designs – High Density Injectors & Low Density Injectors w/Mixing Devices |
| ● Reagent Monitoring | From flyash; shiftwise or weekly |

Highlights of SCR Experience of Japan & Europe (Cont'd.)

- Air Heater European mostly Enamel Coated DP up to 8 in. w.g., Deposition
- Flue Gas Handling Typically 3-7 in. w.g. Equipment upgrade during FGD retrofit mostly accommodated SCR requirement
- I & C Feed forward Signal for reagent demand from boiler NO_x with a feedback signal based on outlet

NO_x

Summary of European & Japanese SCR Experience

- Coal sulfur mostly within 1.7%
- Av. Max. 85% NO_x removal
- Significant number of tower boilers with a few SCR operating problems
- Routine use of combustion controls
- Graduated catalyst geometry
- Inconsequential Am. Sulfate/bisulfate deposition – low SO₃ - <2 ppm NH₃ slip – enameled APH surfaces
- Alkali injection – arsenic bonding and SO₃ control
- Catalyst channel blockage by unburned carbon and coarse fly ash
- Ammonia slip monitoring
- Catalyst performance monitoring & regeneration

Evaluation of Transferability of Japanese & European Experience to U.S. SCR Application

- Japanese & European experience mostly transferable to US with respect to deNO_x effy., NH₃ Slip, SO₂/SO₃ Conv. E, SV, High/Low Dust, Catalyst Composition, Geometry & Management
 - Gas Velocity Distribution
 - NH₃/NO Ratio SD
 - Air Heater Performance

US Situation Differs With Respect To:

- Reactor layout due to boiler design difference
- Reagent – Aq. Ammonia & Urea
- Higher deNO_x effy. – advanced goal
- Most importantly Coal Type & its concomitant issues – e.g. Arsenic Poisoning, Pore Blockage by PRB Coal Ash & SO₃ Plume

Typical U.S. SCR Experience

Typical U.S. Performance

Serial #	1	2	3	4	5	6	7	8	9	10	11	12
Unit	New Madrid #2	Gavin	Somerset	Paradise #1&2	Bowen 1 & 2	Bowen 3 & 4	Gorgas #10	Birchwood	Roxboro #4	Hawthorn #5	Brandon Shores	Logan
Size Mwe	640	2 x 1,300	675	700	756	950	780	250	2 x 735	500	1370	200
Utility	AECI	AEP	AES	TVA	GA PWR	GA PWR	AL Pwr	So Energy	CP&L	KCP&L	Constellation	PG&E
Date Commissioned	2/7/00	5/01	7/99	Spring 2000 & 2001	5/01	5/03	5/02	11/96	7/01	5/01	5/01	9/94
Firing Mode	Cyclone	Wall	Wall	Cyclone	Corner	Corner	Corner	Corner	PC	PC	PC	PC
Coal Type	100% PRB		High Vol. Bit.	Bit./PRB	Bit.	Bit.	AL Bit		Bit.	PRB	Low S Bit.	
Inlet NOx lb/Mbtu	1.5		0.55	0.86-1.6	0.45	0.45	0.55		0.46	0.225		
Outlet NOx lb/Mbtu	0.105		0.05		0.07	0.07	0.08	0.1	0.097	0.099		
DeNOx Eff. %	93	90	90	90	85	85	85		79	56	90	63
NH ₃ Slip PPM	3	<0.2	3	2	2	2	2	5	2	2	2 (1 achieved initially)	
SO ₂ lb/Mbtu	0.4	4.75-6.5			2	2	3.0					
% Sulfur	.23	3.4	1.5-4..60	2-3	1	1	1.5		1.5	0.56	<1.0	
SO ₂ to SO ₃ Conv. %	3.0	1.6	0.75	0.75					<1.0	<0.75		
Catalyst Type	Plate	Plate	Plate	Plate	Honeycomb	Honeycomb	Honeycomb	Plate	Plate	Plate		Plate
Pitch mm	9.2	5.6	6.0	8.2								
Reagent	Ammonia	Urea	NH ₃ anhy.	NH ₃	NH ₃ anhy.	NH ₃ anhy.	NH ₃ anhy.	NH ₃ anhy.	Ammonia	Ammonia		Am.Aq.
SCR/deltaP (Iwg)		8	2.8									
Catalyst Life Hrs.	20,000		24,000	20,000					24,000			
Catalyst m ³			897						314	477		
Arsenic ppm				<20					2			
deltaP Catalyst (Iwg)									1.3	2.0		New
General Remarks	World's 1 st SCR for 100% PRB				Lime addition			Continuous	Low Dust	Continuous	Low Dust	High Dust

U.S. Coals

Coal Quality Parameters Affecting SCR Performance

Coal Parameter:	<ul style="list-style-type: none">• Impact
Sulfur:	<ul style="list-style-type: none">• SO₃ Formation in catalyst chamber by V-oxide• Catalyst pluggage by am. sulfate/bisulfate• High ammonia usage• Catalyst surface masking• Airheater corrosion & pluggage• Catalyst surface & pore blockage
Arsenic:	<ul style="list-style-type: none">• Catalyst poisoning by arsenic oxide.
Calcium:	<ul style="list-style-type: none">• Reduction in available catalyst sites by fine lime particles from PRB coal
Sodium & Potassium:	<ul style="list-style-type: none">• Blockage also by Na&K-sulfates
Chlorine & Fluorine:	<ul style="list-style-type: none">• Reduction in active catalyst site by formation of vanadium chloride/fluoride• Catalyst pore blockage by Am-chloride/fluoride
Unburnt Carbon:	<ul style="list-style-type: none">• Increased ammonia usage
Ash Burden	<ul style="list-style-type: none">• Blockage of initial catalyst layers• Higher the ash burden, higher catalyst deactivation & air heater corrosion/blockage

Table 1
Typical Mineral Ash Analysis
Of U.S. Bituminous Coals

Constituent weight percent of ash

Origin	Sample No.	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	TiO ₂	CaO	MgO	Na ₂ O	K ₂ O	SO ₃	Sample No.	Comments
Kentucky Deane	2	59.6	26.7	4.2	3.4	2.3	0.7	0.8	0.1	1.8	2	High in silica
	3	68.5	20.8	2.6	3.6	1.4	0.4	0.6	<0.1	1.7	3	
Illinois Carrier	26	17.5	9.2	64.1	0.4	4.1	0.4	0.3	1.0	1.8	26	Low in silica, sample 26, exceptionally high in iron; sample 67 moderately high in iron and calcium
Mills	67	25.6	21.0	20.5	0.6	15.8	0.7	2.0	0.2	10.9	67	
Utah Horse Canyon												
Utah Horse Canyon	124	42.0	26.5	7.5	1.2	7.9	1.6	1.2	1.6	9.5	124	Moderately high in silica
West Virginia, Bickmore	127	46.1	34.5	7.5	1.5	2.5	0.7	0.3	2.0	3.0	127	
Pennsylvania, Ebensburg	142	19.4	10.5	36.6	0.4	8.9	5.8	0.4	1.1	15.9	142	Low in silica, exceptionally high in iron
Oklahoma, Redstone	143	29.2	14.2	27.8	0.5	7.1	5.1	0.6	1.5	14.0	143	

Table 2
Typical Mineral Ash Analysis
Of U.S. Sub-Bituminous/Lignite

Constituent weight percent of ash

Origin	Sample No.	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	TiO ₂	CaO	MgO	Na ₂ O	K ₂ O	SO ₃	Sample No.	Comments
North Dakota, Zap lignite	87	20.0	9.3	7.5	0.5	20.8	6.0	11.5	0.4	21.9	87	Exceptionally high in sodium, fairly high in calcium, low in silica and alumina
	88	23.5	13.6	5.7	0.5	18.3	5.4	8.3	0.3	21.7	88	
Montana, Savage lignite	91	17.0	14.6	1.2	0.4	34.6	13.3	0.3	0.1	14.3	91	Low in sodium, exceptionally high in calcium, low in silica and alumina
	92	12.5	13.3	6.8	0.3	26.1	10.4	0.3	0.1	26.17	92	
Wyoming, Glenrock and Gilette sub-bituminous	99	39.8	19.4	3.9	0.9	15.0	2.4	0.2	1.1	16.2	99	Glenrock fuel-ash sample moderately high in calcium; Gilette sample exceptionally high in calcium and exceptionally low in silica and alumina
	100	9.8	12.9	6.7	1.0	33.4	7.9	1.6	0.2	21.7	100	
Texas; Darco lignite	140	38.4	21.0	11.2	1.1	10.0	1.7	0.7	0.5	14.6	140	Sample 140 moderately high in calcium and iron; sample 141 exceptionally high in silica
	141	62.1	16.8	3.9	0.9	5.2	1.1	0.5	0.6	8.9	141	

**A Typical Example of
The Difference in the Form of
Presence of Alakaline & Alkaline Earth Components in
US bituminous & Sub-Bituminous Coal**

% Total In <u>Each Portion</u>	<u>Illinois Bituminous</u>		<u>Wyoming Sub-Bituminous</u>	
	<u>Mineral Matter</u>	<u>Organically Associated</u>	<u>Mineral Matter</u>	<u>Organically Associated</u>
Coal Ash	80	20	46	55
CaO	70	30	3	97
MgO	46	54	6	94
Na₂O	56	44	0	100
K₂O	84	16	31	69
S_iO₂	100	0	100	0
Al₂O₃	91	9	55	45
TiO₂	100	0	72	28
Fe₂O₃	34	66	15	85

Main Features of U.S. Coals With Adverse Impact on SCR Technology

Bituminous

- Higher sulfur than the coals used in Europe & Japan
- Higher arsenic (comes with sulfur)
- Higher ash burden (compared to sub-bituminous)
- More slagging type coal

Sub-Bituminous/Lignite

- Higher calcium & Magnesium mostly coal bound as in PRB
- (These in European & Japanese cases are in form of minerals)
- Higher sodium & potassium
- These pose perhaps the greatest challenge to U.S. SCR installations

Eastern/Western Blends

- Still contains coal bound calcium, magnesium
- Little European & Japanese experience (blends are of imported coals low in sulfur, calcium & sodium)

Mechanism of Catalyst Deactivation by PRB Fly Ash

- **PRB coal contains high (up to 30% of ash) calcium & high (up to 10% of ash) magnesium**
- **These are mostly (up to 80%) present in the coal matrix**
- **These are readily released in the furnace as fine particles of reflective calcium oxide & magnesium oxides & silicates which combine with sulfur trioxide in the convective pass to form calcium (and magnesium) sulfates which plug the catalyst pores.**
- **Sodium (and potassium) of PRB coals form sticky sulfates and phosphates on the catalyst sites promoting additional deposition of fly ash.**

What Are Required?

And What Should Be Done?

Use Chemicals to Improve Catalyst Pore Blockage using PRB Coal, Reduce LOI and Catalyst Surface Masking Firing Bituminous Coal

- **Convert fine calcium oxides & magnesium oxides into compounds prior to contact with SO_3 and thereby minimize formation of Ca & Mg sulfates and reduce catalyst pore blockage**
- **A chemical has been developed to control PRB's reflective ash generated slagging. This is considered to be able to minimize Ca & Mg sulfate formation & thereby reduce catalyst pore blockage**
- **Used over a decade, a chemical is used by the U.S. utility industry firing bituminous coals to reduce LOI and improve cycle efficiency by recovering superheat & reheat steam temperatures.**

Improved Understanding of Coal and Ash Quality

**Laboratory Studies for Inorganic Coal
Constituents & Ash Deposit evaluation**

Physical

**Computer controlled scanning electron
microscopy & X-ray Diffraction**

Chemical

**Fractionation for modes of occurrence of
calcium, magnesium, sodium & potassium in
coal**

Arsenic Poisoning Mitigation

Mean Arsenic content of U.S. Coals

Source: U.S. Geological Survey

	As PPM
• Appalachian Basin n 2835	22
• Illinois Basin n 298	10
• Powder River Basin n 160	2.6
• Colorado Plateau n 180	3.1
• Gulf Coast Lignites n 135	5.1

n represents number of measurements

Physical & Chemical Deactivation Mechanisms

Physical – Arsenic Oxide (As_2O_3) Molecules Condense in Catalyst Pores

Chemical – As_2O_3 Reacts with V_2O_5 to form Non-Catalytic Arsenic Vanadate Compounds

Mitigating

Measures: 1) Molybdenum Oxide (MoO_3) Added to Catalyst Composition which form Arsenic Molybdate –
More de- reactive than As-Vanadate which slows down activation of active V_2O_5 sites

2) Calcium Oxide (CaO) added in furnace form Ca-Arsenide [$\text{Ca}_3(\text{AsO}_4)_2$] which reduces As_2O_3 conc. in boiler

Reliable method of measurement of coal arsenic by EPA 2051 or ASTM D6357 required & NOT by ASTM D4606

SO_3 Plume Visibility Prevention

TABLE 1

(Ref.. Mineral Impurities in Coal Combustion – E. Raask P. 404

Table 19.1 SO₃ concentration, dewpoint temperature, and acid deposition in coal-fired boilers

Type of Coal	Sulfur In Coal (%)	CaO In Ash (%)	SO ₃ in Flue Gas (ppm v/v)	Dewpoint Temperature K (F)	Maximum Acid Deposition Rate (mg m ⁻²)
High sulfur, low calcium	>2.5	2-5	10-25	400-410 (261-279)	5-10
Medium sulfur, low calcium	1-2.5	2-5	5-10	295-400 (72-261)	2.5-5
Medium sulfur, Medium calcium	1-2.5	5-10	1-5	285-295 (54-72)	1-2.5
Low sulfur, high calcium	<1	>10	<1	<285 (<54)	<1

**Development of Improved Prediction Method
Required For SO₃ Concentration
Across High Sulfur Flue Gas System
(Current Methods Found
Inapplicable To Field Performance)**

- Coal Sulfur, Ash Chemistry, Flue Gas SO₃ Conc. & Acid Dew Point Relationship need review and development
- Mechanism of SO₃ Concentration Enhancement by ESP needs to be Ascertained
- Multi-Point Application of Chemicals for SO₃ Mitigation Should be Considered

- **Alternative Catalyst Development Without Oxides of Vanadium, Titanium, Molybdenum & Tungsten**
- **Catalyst regeneration**
- **Reliable on line ammonia slip monitoring**

Conclusions

- Over 100,000 MW of coal fired units in the USA might need SCR retrofit by 2004 in response to EPA's SIP Call rule
- Evolved from two generations of design in the 70's & 80's in Japan & Europe, SCR technology has made great strides W.R.T. NO_x removal, NH₃ slip, catalyst, reactor design, process conditions, etc.
- While this experience has been utilized by the US SCR industry, it is not adequately transferable to meet the challenges of US boiler designs and especially the unique US coal quality
- High sulfur & high arsenic bituminous & high coal-bound calcium based PRB coal have not been experienced in Europe & Japan

Conclusions (Cont'd.)

- Elimination of SO₃ plume formation firing high sulfur bituminous coal in units with wet FGD needs:
 - Development of improved SO₃ formation prediction methods
 - Improved understanding of the role of process equipment/conditions
 - Multiple control strategy
- Use of additives to control catalyst deactivation by calcium oxide & arsenic needs immediate consideration since the deactivation mechanisms are understood & additives effectiveness are proven
- Development of non-vanadium based catalyst formulations for high dust application needs promotion

These goals need to be achieved as

“Necessity is the Mother of Invention”