Feature Article. . .

REFRACTORY LINER MATERIALS USED IN SLAGGING GASIFIERS

James P. Bennett, Ceramic Engineer and Kyei-Sing Kwong, Ceramic Engineer, U.S. Dept. of Energy - Albany Research Center, 1450 Queen Ave SW, Albany, OR, USA 97321, jbennett@alrc.doe.gov

ABSTRACT

Refractory liners are used on the working face of entrained-flow slagging gasifiers that react coal, petroleum coke, or other carbon feedstock with oxygen and water. The refractory liners protect the gasifier shell from elevated temperatures, corrosive slags, and thermal cycling during gasification. Refractory failure is primarily by two means, corrosive dissolution and spalling. High chrome oxide refractory materials have evolved as the material of choice to line the hot face of gasifiers, yet the performance of these materials does not meet the service requirements of industry. A review of gasifier liner materials, their evolution, issues impacting their performance, and future research direction are discussed.

Keywords: Gasifiers, Slagging gasifiers, Chrome oxide refractories, Coal slag, Petroleum coke slag, Slag corrosion, Spalling

INTRODUCTION

Gasifiers are used to produce power and chemicals used in other industrial processes. A gasifier acts as a containment vessel to react liquid fuels, a carbon source (coal and petroleum coke are the most commonly used), water, and oxygen at elevated temperatures and under reducing conditions; producing CO and H_2, as the primary gases, along with CO_2, CH_4, HS and other trace gases. Gasifiers fall into one of three varieties: fluidized-bed, moving-bed, or entrained-flow systems; with only the entrained-flow gasifier producing a fluid ash (slag) from impurities in the carbon source, the other two gasifier varieties producing a "dry" ash by-product. In an entrained-flow gasifier, the bulk of this molten slag flows down the gasifier sidewalls, with only minor amounts associated with the excess carbon and existing as fused particles. Slag is considered one of the primary by-products of the gasification process, varying greatly in chemistry and quantity depending on the gasifier feedstock and operation conditions. A coal slag typically contains SiO_2, FeO, CaO, and Al_2O_3; with vanadium oxides also present in a petroleum coke slag. Slag quantities in excess of five or more tons per hour can be generated in a gasifier, depending on the carbon source, the quantity of impurities present in it, and gasifier throughput. The ability of a slag to flow and to cause predictable refractory wear in an entrained-flow system is critical to its operation. Industrial and government interest in gasifiers has varied over time, driven by high energy prices, perceived/real shortages of energy producing raw materials, efficiency of the gasification process, low water consumption compared to other processes, and the ability of a gasifier to generate very low levels of pollutants. The low level of pollutants is achievable because gasification is a controllable closed circuit process that has been designed to produce low levels of NOx, and because other low level emissions such as sulfur and mercury from the gasification process can be captured and/or processed for reuse as by-products. Research is underway to sequester and/or find uses for CO_2, a future environmental concern and a thermodynamic by-product of the gasification process that is easily captured.

The gasification chamber of an entrained-flow gasifier typically operates at temperatures between 1250º and 1550ºC, at pressures of 400 psi or higher, and is lined with refractory materials to contain the severe environment and to protect the outer steel shell from erosion, corrosion, and high temperature. As mentioned, the slag is liquefied in the gasification chamber, and can corrode, penetrate, and react with the refractory liner at elevated temperatures, severely limiting refractory service life and gasifier operation. Two types of entrained-flow gasifiers are used in industry, water cooled and air cooled. Water cooled gasifiers have a working face lining of Al_2O_3-SiC refractory, and have a satisfactory service life because slag freezes on the refractory surface, restricting slag penetration and corrosion. The long refractory service life of the water cooled design comes at a performance cost, with slightly lower energy efficiency versus an air cooled design. Air cooled entrained-flow gasifiers are lined with refractory materials found to give the best service life and on-line performance – a high chrome oxide material that contains alumina and may contain other additives like zirconia. Refractory liners in an air cooled entrained-flow gasifier last anywhere from three

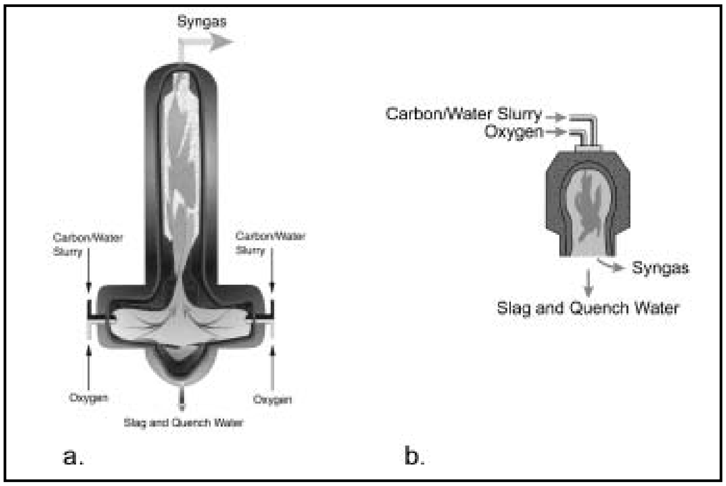


Figure 1. Two types of commercial entrained-flow slagging gasifier designs; a) opposing flow burner, and b) single burner.

months to two and one-half years. Cross sections of two types of air cooled entrained-flow gasification chambers used in industry are shown in Figure 1. These gasification chambers can be up to approximately 82 feet in length and 16 feet in diameter [1]. In spite of the superior performance of chrome oxide based refractory liners over other material, gasifier operators and designers using air cooled systems have identified refractory service life as the most important factor limiting their on-line availability [2]. This paper will review refractory liner materials currently used in gasifiers and their evolution, analyze failed refractory materials, and discuss trends in future refractory research for gasifiers.

REFRACTORY MATERIALS USED IN GASIFIERS AND THEIR EVOLUTION

Because of the severe environment in an air cooled entrained-flow slagging gasifier, the material challenges for a refractory liner are many, and include: elevated temperature; large and/or rapid changes in temperature; erosion by particulates; molten slag attack; variable slag composition resulting from the feed stock; attack by hot corrosive gases; alkali vapor attack; and variable oxidizing and/or reducing conditions [3-5]. Refractory materials that can withstand these environments for long periods of time are necessary for a continuous, efficient, and reliable gasification process. A number of refractory compositions have been evaluated historically in these harsh thermal environments; and include sintered and/or fused cast alumina-silicate, high alumina, chromia-alumina, chrome-magnesia spinels, alumina and magnesia, alumina and chrome, and SiC refractory compositions [6-10]. Thermodynamic and phase diagram studies must be taken into account when choosing the composition of hot face or of back-up refractory materials. Compounds like Fe_2O_3 and SiO_2 can form reactions with CO, H_2, or H_2O in the gasifier environment as follows:

1. $2CO \rightarrow CO_2 + C$ (in presence of Fe_2O_3, T ~ 570ºC) [11]
2. $SiO_2 + H_2 \rightarrow H_2O + SiO\uparrow$ [11]
3. $2SiO_2 + 3\ H_2O \rightarrow Si_2O(OH)_6\uparrow$ [12]

These and other gasification reactions are possible or have been theorized [1, 11-14] to occur on either the surface or within a gasifier refractory (due to the thermal gradient). Caution must be taken to minimize possible gaseous interactions; such as with CO, H_2, or H_2O; by controlling refractory components. Wear by corrosion was evaluated and noted in refractory materials containing high levels of Al_2O_3 or MgO/Al_2O_3 spinels during simulated gasifier environment testing using a synthetic coal type slag [6-9]. Other refractory materials, like SiC or Si_3N_4, were found to react with components in the slag, causing severe material wear. SiC, for instance, interacts with FeO in the slag, yielding volatile gases (CO, SiO and/or others) and metallic iron. Fuse cast refractory materials with little or no porosity often had low chemical wear, but suffered from thermal shock, a threat to refractories because of constant gasifier shutdowns.

In general, chrome oxide additions to a refractory composition were found to improve their slag resistance. The high chrome oxide material used in most slagging gasifiers today has its roots in DOE and Electric Power Research Institute (EPRI) funded efforts traceable back to the 70's and 80's [3-9, 13, 15]. Research and industrial experience indicated that only $Cr_2O_3 - Al_2O_3$, $Cr_2O_3 - Al_2O_3 - ZrO_2$, and $Cr_2O_3 - MgO$ compositions could withstand gasifier environments long enough to be economically feasible [3, 10, 14]. A strong desire has always existed in industry to develop an improved performance refractory with a low cost. Bakker [16] indicated that a minimum Cr_2O_3 level of 75 wt.% is necessary in a refractory material for sustained material performance in slagging gasifiers. One problem in evaluating materials is that good tests simulating the gasifier environment do not exist, making it difficult to equate laboratory results with material performance in industry. Taber [1] discusses general factors to take into account when choosing a gasifier refractory, including chemical compatibility, thermal and mechanical concerns, and design issues.

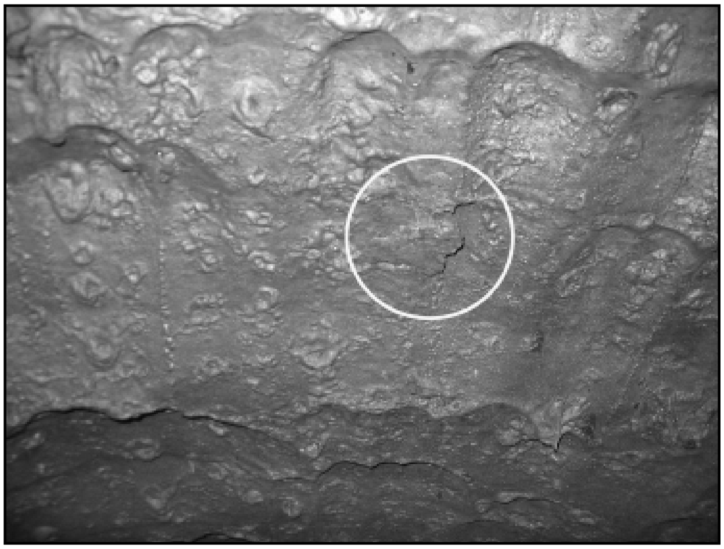


Figure 2. Hot face of high chrome oxide refractory material in a slagging gasifier (spalled refractory material circled).

The failure of a refractory lining in a gasifier is expensive, both in terms of the refractory replacement costs (as high as one million USD, depending on gasifier size and the extent of rebuild required), but also in lost production down time. Relining a gasifier requires that the system be completely shut down, and under the best of circumstances takes about 10 days for a partial rebuild, longer for a complete rebuild. A rebuild involves cooldown (up to 5 days) and teardown and repairs (3 days for a partial rebuild and 7-10 days or longer for a full rebuild, depending on the extent of repairs necessary). Some gasification facilities maintain a second gasifier for use while repairs are being made, reducing system downtime and increasing on-line service and availability of the gasification system. Even then, the time to switch gasifiers can vary from hours to days, depending on if the spare gasifier is available in pre-heat mode. Because of the long down times required for repair, gasifier operators would like to install refractory linings with a reliable life of at least three years. The current generation refractory liners installed in gasifier systems have yet to meet this requirement, failing in as little as three months in high wear areas. Refractory service life is highly dependent on variables such as the carbon feedstock, the temperature of operation, material throughput, and the frequency and quality of

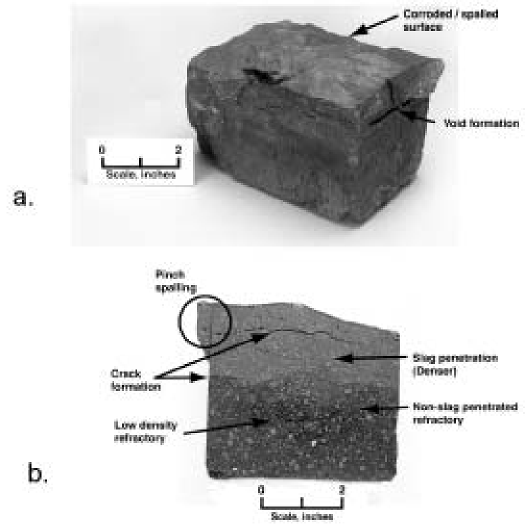


Figure 3. High chrome oxide refractory removed from a slagging gasifier; (a) hot face and (b) cross section of refractory.

plant maintenance. Differences in these variables at gasifier sites may indicate that the development or customization of high chrome refractories for each plant site may be necessary in order to achieve optimum refractory performance. Nonetheless, reliable refractory liners are needed to ensure that gasification fulfills its potential as a clean and efficient means of generating power.

POSTMORTEM ANALYSIS

High chrome oxide refractory materials at the hot face of a slagging gasifier after shutdown is shown in Figure 2. Note that the slag (and possibly feedstock and/or gas velocity) has created an irregular worn surface on the refractory liner. Refractory materials used to line a gasifier are typically dense firebrick composed of chromium oxide as the primary component, along with smaller quantities of other refractory oxides (typically aluminum and/or zirconium oxide). The wear of refractory from the hot face is thought to be caused by two principle mechanisms, corrosion and spalling. Corrosion can occur from dissolution of refractory material into the flowing slag or by dissolution of the bond phase at the hot face, followed by freeing of refractory grains into the flowing slag. This type of wear occurs continuously throughout the gasifier's service life and is gradual and predictable in a high chrome oxide refractory liner. Spalling occurs in stages, and is the irregular removal of large chunks of the refractory materials hot face into the flowing slag. Physical evidence of corrosion and spalling are both indicated in Figure 2. Note the washed-out appearance of the refractory surface (corrosion) and the spalled refractory fragment (circled in Figure 2) sliding down the gasifier sidewall.

A high chrome refractory brick removed from the hot face of a gasifier sidewall and the cross section of it are shown in Figure 3. The surface of the hot face (shown in Figure 3a) is in the latter stages of spalling, which would lead to the rapid and physical removal of a large portion of the refractory's surface, leading to a shorter refractory service life. Bakker [16] indicated spalling as a refractory wear mechanism, and demonstrated how the repeated occurrence of it during a gasifier's operation would led to an incremental and rapid shortening of refractory service life versus the predictable and gradual linear wear caused by corrosion.

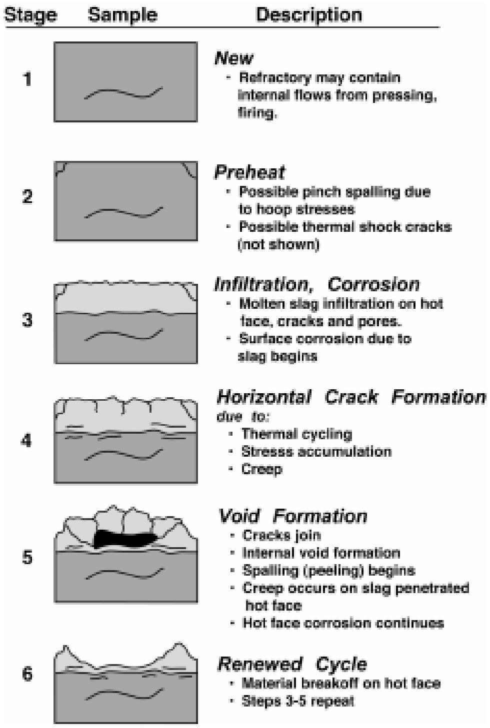


Figure 4. Stages of refractory wear.

Spalling and crack growth at different areas associated with slag penetration into a refractory are shown in Figure 3b. Cracks form and link together in the slag penetrated or slag penetrated/non-penetrated interface of the refractory, creating a situation where large surface portions of a refractory may become freed from the base refractory (spalled). Creep, corrosion, thermal cycling of the gasifier, an oxidizing/reducing atmosphere, or new mineral phase formation brought about by slag interactions with the refractory may all contribute to spalling. The creation, growth, and joining of cracks appears to be a reoccurring process during the operation of a gasifier. Spalling has occurred in many refractory applications and has many causes [17].

The stages of a gasifier refractory wear at different times and how these stages lead to refractory failure by corrosion and spalling is shown in Figure 4. The first stage of the process indicates when a refractory lining has been installed, and shows internal laminations or low-density areas associated with the material's manufacture. Flaws such as these may exist in the refractory because of manufacturing practices, but they can be eliminated or minimized through inspection of the refractory material before installation. Stage 2 indicates hoop stress cracking occurring at the refractory hot face edge (a rare phenomenon) and the possible formation of thermal shock cracks (not shown because this type of flaw is location specific). Once a gasifier is in service and slag has contacted the brick's hot face, it causes surface corrosion and begins to penetrate the pores of the refractory, as shown in stage 3.

The molten slag attack leading to corrosion and spalling of a chrome oxide refractory hot face starting in stage 3 and build-

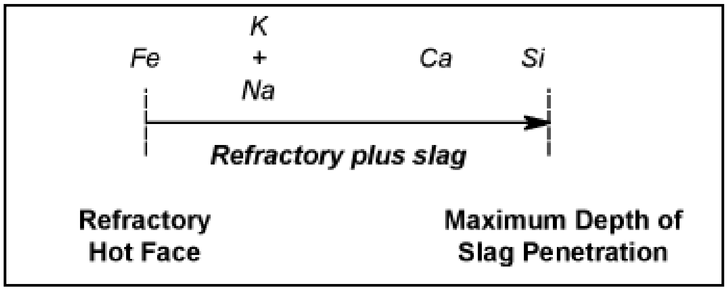


Figure 5. Relative penetration of slag elements into the microstructure of a high chrome oxide refractory.

ing up to spalling shown in stage 5 occurs throughout a gasifiers operation. Corrosion is limited to the interaction between slag components and the refractory, and is influenced by gasifier operational temperature and the melting point, solubility, and fluidity of new phases that are formed. Chrome oxide works well preventing corrosion because it interacts with the slag to form high melting phases in situ. In doing so, it also raises the silica content (and thus viscosity) of the slag that has penetrated the refractory, limiting further slag penetration. It should be noted that particulate wear (abrasion) from carbon feedstock caused by burner alignment or feedstock throughput may appear to be corrosive wear on the refractory surface rather than abrasion. Wear of this type can be difficult to ascertain.

Spalling (stages 3-5 in Figure 4) may be initiated by many causes. Slag penetration and interactions between the slag and the refractory form new phases with different thermal expansion that could lead to crack formation and stresses. Once slag has penetrated a refractory surface, density/expansion differences between the penetrated/non-penetrated refractory also exist that could lead to crack formation and spalling. Spalling may also be influenced by iron compounds that interact in the slag/refractory, forming spinel or solid solution phases with an expansion behavior different in the refractory or that may change with the oxidizing/reducing state of the gasifier. Other operational practices, like depressurization of the gasifier while the hot face is at or near operating temperature, rapid or uncontrolled gasifier shutdown, oxidizing preheats/shutdowns of the gasifier, gasification vessel design/construction, refractory creep, or thermal cycling of the gasifier could be additional causes or contributing factors to hot face spalling. Figure 4 does not discuss these causes of refractory failure, only indicating the stages of failure.

Previous work on samples penetrated by slag [18] have shown that iron penetration from slag into a refractory generally did not occur to any appreciable depth, being limited to the surface, while calcium, aluminum, and silica penetrate much deeper into the refractory structure. The relative depth of slag element penetration into a high chrome oxide/alumina refractory is shown in Figure 5. Most elements in the slag react with the Cr_2O_3/Al_2O_3 refractory matrix to form glass, solid solutions, or spinels, or other new phases. Based on microstructural analysis, the role of aluminum in the refractory matrix is unclear since it is already present in high chrome oxide refractories. Evidence suggests alumina may be removed from matrix aggregate into the slag in certain situations, although additional research is needed to clarify its behavior. Calcium oxides react with silica to form calcium silicates, tending to form fluid, low melting slag compositions that penetrate deep into the refractory. The high silica slag that penetrates furthest into the hot face of the refractory is thought to increase in viscosity as other oxide components of the slag interact with the refractory matrix, causing the slag to become so viscous that further penetration effectively stops. The thermal gradient, ΔT, across the hot face refractory is small, making it unlikely that it alone is the cause of the slag" freezing."

Although spalling and corrosion are thought to be the primary causes of refractory failure, evidence that other mechanisms of

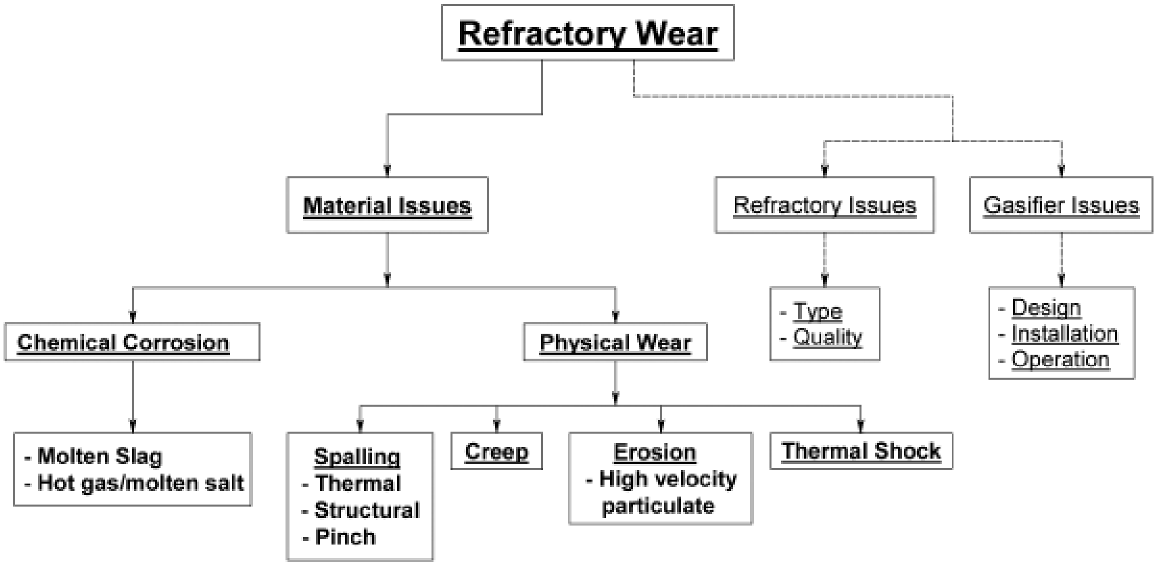


Figure 6. Causes of refractory wear in a slagging gasifier.

refractory wear such as hot gas corrosion or salt buildup in the refractory have been noted and may occur in specific gasifier locations.

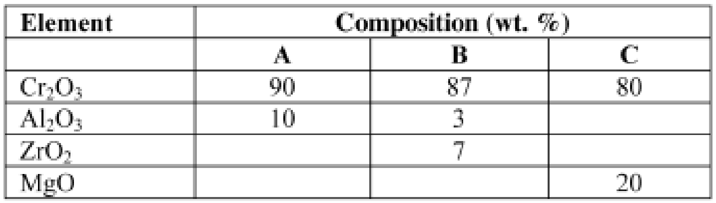
A flowsheet breaking down the major types of refractory wear in a slagging gasifier is shown in Figure 6, with the causes categorized as material, refractory and gasifier issues. Gasifier and refractory issues are indicated by a dashed line in Figure 6 as they tend to remain fixed gasifier issues over a period of time, being determined by economic and production issues. Material issues that lead to gasifier wear during its operation are impacted by chemical corrosion and/or physical wear. It is important to make a study of all possible causes of wear in a gasifier to determine how they can be impacted by system changes. All causes of refractory wear tend to be lumped together into a single category of refractory failure, complicating corrective action.

TRENDS IN REFRACTORY MATERIAL DEVELOPMENT

The major types of high chrome oxide refractories used in an entrained-flow air cooled slagging gasifiers are indicated in Table 1. Differences exist in the performance of the different types of refractories, with the chrome-alumina and chrome-alumina-zirconia based compositions (A and B) now predominately used in industry. High chrome oxide content refractories (greater than 85 wt.% Cr_2O_3) are used in the severe wear areas of a gasifiers, with lower chrome oxide content materials used to line less severe wear areas of the gasifier, resulting in a zoned lining. This is done in part because of high material cost of the high chrome oxide materials. Variations in some refractory compositions are made to give special properties to a material, such as thermal shock resistance. Changes such as these have resulted in many different linings being used in gasifiers as users seek the longest lasting materials at the best price. Gao [14] has indicated one lining makeup, for instance, as a high chrome oxide material on the hot face, followed by an 85 wt.% Al_2O_3/13 wt.% Cr_2O_3 backup lining, although up to six layers of refractory insulation have been reported [1]. Regardless of the refractory lining or its makeup in a gasifier, worn linings or voids in a lining can result in dangerous hot spots on the gasifier containment shell, a situation which must be prevented.

Research is on-going to develop new or improved refractory liner materials for use in slagging gasifiers, especially in high chrome oxide compositions. The Albany Research Center, for instance, has developed and is testing the performance of a high chrome oxide refractory material with phosphate additions [19]. Other future research trends could be towards developing refractory materials coatings, monolithic linings, or refractory materials that do not contain chrome oxide or are low in chrome oxide. Research is underway at the Albany Research Center into non-chrome oxide refractories for the following reasons: 1) the high cost of chrome oxide refractories, 2) the potential for the formation of hexavalent chrome during service (not currently known to be an area of concern), 3) the difficulty in fabricating and sintering high chrome oxide materials, 4) handling difficulties of high chrome oxide refractories due to their density, 5) possible supply issues associated with high chrome oxide refractories and 6) the fact that high Cr_2O_3 refractories have not met the performance requirements of gasifiers. Greater recycling of spent linings from gasifiers could also be encouraged. Recycling is limited because of frequency of generation issues, shipping costs and distances, storage costs, and mixed lining issues.

Table 1. High chrome oxide refractory material composition (approximate) used in air cooled entrained-flow slagging gasifiers



Element	Composition (wt. %)		
	A	B	C
Cr_2O_3	90	87	80
Al_2O_3	10	3	
ZrO_2		7	
MgO			20

CONCLUSIONS

Air cooled entrained-flow slagging gasifiers are lined with high chrome oxide refractory material. The gasifier acts as a containment vessel to react carbon feedstock (typically from coal or petroleum coke) with water and oxygen in a reducing environment to produce CO and H_2, along with lower levels of other gases. Gasifiers are used for power generation or to produce chemicals for other processes. Slagging gasifiers have service lives ranging from three months to two and one-half years, with longer service life desired. The service life of a gasifier refractory is dependent upon feedstock, the temperature of operation, material throughput, and the frequency and quality of plant maintenance. Failure is typically by chemical corrosion or spalling. Improvements are being sought in refractory performance by developing new or improved refractory materials. These improvements are being sought through product reformulation, through phosphate additions to the refractory, or through the development of low or no chrome oxide refractory materials.

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TRI News Continued from Page 5

Group and the Institute's Regulatory Committee will be missed, but we welcome Greg to the refractories community. Greg can be reached at: Tel: 412-294-1023, Fax: 412-293-1123, and E-mail: Greg.McDonough@rescoproducts.com.

ALMATIS TO UPGRADE PLANT

Almatis has announced a major investment in a fully integrated tabular alumina facility at their existing Qingdao plant in the Shangdong province of China. The $18 million investment will expand existing tabular capability, supporting the installation of two shaft kilns for tabular alumina sintering and more than doubling the existing crushing and screening capacity. Under the terms of Almatis' long-term supply agreement with Alcoa, the Qingdao plant will be supplied with calcined alumina feedstock from Alcoa, Australia.

"China is the fastest growing high alumina refractory raw materials market in the world and Almatis' China sales are growing as rapidly. Our independent ownership allows Almatis to invest now, providing the Chinese refractory market continued full supply of the high quality, high alumina raw materials they need to support the shift to modern steel manufacturing and steel capacity expansion. High quality steel production demands high quality and high performance raw materials, such as Almatis' tabular alumina", stated Gangolf Kriechbaum, Almatis Chief Commercial Officer.

In addition to tabular alumina, the brownfield expansion will produce spinel and bonite, other high-grade raw materials used as aggregates in shaped and unshaped high performance refractories.

Almatis was formerly known as Alcoa World Chemicals and now operates as an independent alumina materials company, headquartered in Frankfurt, Germany.

TRI SAFETY AWARDS

TRI Chairman John Turner announced the winners of the 2004 TRI Chairman's Safety Awards at the TRI Spring Meeting. Winner in the small company category was The Nock and Son Company. The mid-size category award went to Riverside Refractories, and the large company recognition went to Unifrax Corporation.

Rob Crolius also read the list of the winners of the TRI President's Awards which were presented to facilities that operated in 2003 without a lost work time accident. They are: Inland Refractories' Avon, OH plant; Minteq International's Bryan, OH, Baton Rouge, LA, Dover, DE, and Slippery Rock, PA plants; New Castle Refractories' Newell, WV plant; The Nock and Son Oak Hill, OH plant; Refco Incorporated's Wellstone, OH plant; Refractarios Peruanos' Callao Plant; Resco Products' Cedar Heights Clay, Oak Hill, OH plant; Riverside Refractories' Pell City, AL and Nanticoke, Ontario plants; the RPC Division of Thermal Ceramics in Elgin, IL; and, the Amherst, NY and Sanborn, NY plants of Unifrax Corporation.

REGULATORY

OSHA Agenda: Silica and Hex Chrome

In the Regulatory Agenda published June 28, 2004, the Occupational Safety and Health Administration affirmed its intent to publish a final rule on hexavalent chromium by October 4, 2004. That date had been ordered by the Third Circuit Court of Appeals.

Regarding crystalline silica, OSHA indicated that a peer review of the risk assessment will be completed by February 2005. It is unclear how the agency will proceed after that. It may move expeditiously on a comprehensive regulation, or focus on some specific issues like modernizing PELs or standardizing sampling methods.